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Temperature effect on exchange coupling and magnetization reversal in antiferromagnetically coupled (Co/Pd) multilayers

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Magnetization reversal of antiferromagnetically coupled (AFC) soft and hard (Co/Pd) multilayers was studied as a function of temperature. While the hard $[\text{Co}(0.3\text{ nm})/\text{Pd}(0.8\text{ nm})]_{\times 10}$ was kept unchanged, the softness of the $[\text{Co}(t)/\text{Pd}(0.8\text{ nm})]_{\times 3}$ was controlled by varying the thickness t of the Co sublayer. Clear two-step hysteresis loops were observed for all the investigated multilayers with t ranging between 0.4 and 1 nm. The spin reorientation of the soft layer magnetization from in-plane direction to out-of-plane direction was investigated from 50 to 300 K. The antiferromagnetic field H_{AFC} measured from the shift of the minor hysteresis loop reveals a good agreement to the quantum-well model. From the out-of-plane hysteresis loop of the uncoupled soft layer, its magnetization shows an in-plane orientation for $t \geq 0.6\text{ nm}$. The strong H_{AFC} helps to induce an out-of-plane orientation of the soft layer with a linear decrease of its coercivity with temperature. These investigated structures show the possibility to reduce the unwanted stray field and improving the out-of-plane anisotropy even for relatively thicker soft layer. © 2015 AIP Publishing LLC.

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I. INTRODUCTION

Enormous efforts are devoted to the development of magnetic materials with perpendicular magnetic anisotropy (PMA) for bit-patterned media (BPM)^{1–7} and magnetic random access memory (MRAM).^{8–15} In fact, materials with PMA have higher magnetic anisotropy energy compared to their counterparts with in-plane anisotropy. Thus, they are suitable for higher storage density without compromising their thermal stability. For magnetoresistive devices used for MRAM, the key component is a magnetic tunnel junction (MTJ) where a magnetically hard layer acting as a reference is separated by a thin tunnel barrier of less than 1 nm in thickness from a magnetically soft layer (free layer). As the size of the device is reduced, the magnetostatic field from the reference layer could reach values higher than 100 mT and consequently results in an asymmetrical reversal of the magnetization of the free layer for either magnetic field or spin transfer torque switching.^{8–16} This magnetostatic field is inversely proportional to the size of the device and becomes a barrier toward down-sizing the memory device.¹⁷ For BPM application, the small separation between each bit ($<5\text{ nm}$ for 1T bit/in.² recording density) leads to a strong magnetic stray field acting on the neighboring bits. This undesirable reversal of magnetization by this strong stray field represents a serious challenge for BPM with PMA.

In order to overcome this problem, antiferromagnetically coupled (AFC) structures have been proposed. In such a structure, two ferromagnetic layers are antiferromagnetically coupled through a non-magnetic spacer, usually Ru, with a thickness between 0.4 and 1.0 nm. In this configuration, by properly adjusting the relative thickness of the two ferromagnetic layers, the net stray fields generated from this structure can be greatly reduced (Fig. 1). Although for MRAM, the net stray field can be reduced to zero, it is important for BPM application to only minimize it, so that the reading by a magnetoresistive field sensor of the recorded bits is still possible. It is important to mention that vortex-based structures have also been proposed in order to reduce the dipolar coupling between magnetic nanostructures in MRAM devices.^{18,19}

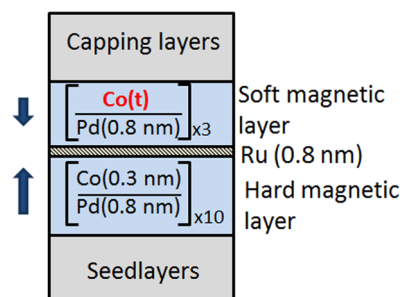


FIG. 1. Schematic representation of the antiferromagnetic structure studied. The thickness of the top multilayer was varied by changing the Co thickness t .

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Multilayers of (Co/Pd), (Co/Pt), or (Co/Ni) were widely used as materials with PMA. Their anisotropy is originating from the interface between the two layers.²⁰ Controlling the magnitude of the anisotropy energy can be easily achieved by adjusting the relative thickness of the two layers forming the stack and also the number of repeats. Therefore, the soft and hard magnetic layers can be prepared using the same set of material. We have recently reported that the AFC can be achieved in $(\text{Co/Pd})_{\times m}/\text{Ru}/(\text{Co/Pd})_{\times n}$ systems even when Co layers in the thinner magnetic layer are very thick and that the PMA in such systems are achieved for Co thicknesses that would normally not produce a PMA.²¹ We also explained that the PMA for such thick Co bilayers is observed due to the AFC. It is therefore, interesting to carry out detailed investigations on the AFC in such systems. In this study, we report on the effect of temperature on the antiferromagnetic coupling for different thickness of the soft layer. This was done by changing the Co layer thickness in the stack.

II. EXPERIMENTS

All the samples were deposited on thermally oxidized Si substrate using DC-magnetron sputtering in a chamber with a base pressure below 4×10^{-6} Pa (3×10^{-8} Torr) at ambient temperature. The investigated stacks consist of substrate/Ta(3)/Cu(5)/Pd(3)/[Co(0.3)/Pt(0.8)] $_{\times 10}$ /Ru(0.8)/[Co(*t*)/Pt(0.8)] $_{\times 3}$ /capping layers where the numbers in brackets are thickness in nanometer. The thicknesses of all single layer films were determined by x-ray reflectometry and the film thicknesses of each layer in the final stacks were estimated from the deposition rate and deposition time. The capping layer is a lamination of 3 nm Pd and 3 nm Ta to protect the whole stack from oxidation. The bottom multilayer had a thinner Co layer and a thicker lamination and hence was harder in nature. In this study, the thickness *t* of Co in the

top multilayer was varied from 0.4 nm to 1 nm with a step of 0.2 nm. The magnetic measurements were carried out using Quantum Design VersaLab magnetometer and superconducting quantum interference device (SQUID) magnetometer in temperature range between 50 K and 300 K. The magnetic field applied perpendicular to the film plane was carried out identically in all measurements. The antiferromagnetic coupling field H_{AFC} was evaluated from the shift of the minor loop of the soft multilayer with 3 repeats.

III. RESULTS AND DISCUSSION

Fig. 2 shows major hysteresis loop of AFC structure in the out-of-plane direction for different Co thickness in the soft multilayer and at different temperatures. The measurements were carried out at several temperatures ranging from 50 K to 300 K. Two steps magnetization switching can be seen for all the samples with a decrease of the coercivity field of the hard layer $H_{\text{c,H}}$ at higher temperature. In contrast, the switching of the soft layer magnetization from parallel state to antiparallel state occurs at low field which shows no significant dependence on the temperature. It is important to note that $H_{\text{c,H}}$ changes by about 25% due to the presence of the soft layer. Without coupling, the coercivity of [Co(0.3 nm)/Pt(0.8 nm)] $_{\times 10}$ was around 95 mT and increased to about 0.12 T when it is coupled to [Co(*t*)/Pt(0.8 nm)] $_{\times 3}$. It seems that the antiferromagnetic coupling hold the magnetization of the hard layer until both the coercivity and the coupling strength are overcome. The small and large arrows in Fig. 2(a) indicate the magnetization direction of the soft layer and the hard layer, respectively. More details on the reversal of magnetization using very small magnetic steps are shown in Fig. 3 for the antiferromagnetically coupled structure with Co thickness of 0.8 nm and measured at 300 K.

To analyze deeply the effect of the antiferromagnetic coupling on the soft layer, we measured the minor hysteresis

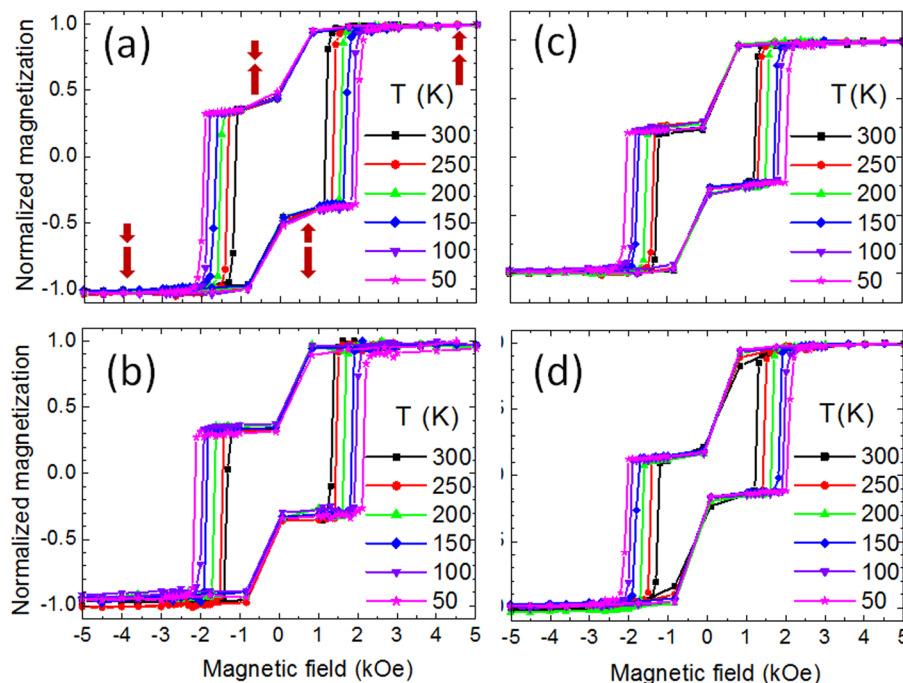


FIG. 2. Major Out-of-plane hysteresis loops of antiferromagnetically coupled [Co(*t*)/Pd(0.8 nm)] $_{\times 3}$ and [Co(0.3 nm)/Pd(0.8 nm)] $_{\times 10}$ multilayers at different temperatures and for different values of Co thickness *t*. (a) for *t* = 0.4 nm, (b) for *t* = 0.6 nm, (c) for *t* = 0.8 nm, and (d) for *t* = 1.0 nm.

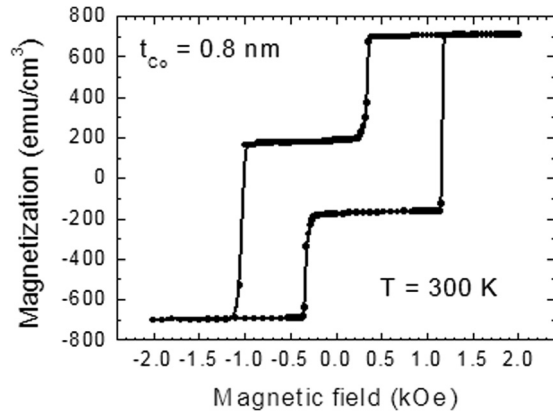


FIG. 3. Major Out-of-plane hysteresis loop of antiferromagnetically coupled $[\text{Co}(0.8 \text{ nm})/\text{Pd}(0.8 \text{ nm})]_{\times 3}$ and $[\text{Co}(0.3 \text{ nm})/\text{Pd}(0.8 \text{ nm})]_{\times 10}$ at 300 K.

loops for the four samples at different temperatures. Fig. 4 represents the out-of-plane minor hysteresis loops of the soft layer when the bottom layer magnetization direction was kept unchanged as indicated by the arrows. Fig. 4(a) is for the AFC structure with Co thickness of 0.6 nm and Fig. 4(b) is for the case where $t = 0.8 \text{ nm}$. The antiferromagnetic coupling field H_{AFC} is evaluated from the shift of the minor loop. H_{AFC} indicated in Fig. 4 is only for $T = 300 \text{ K}$. It can be seen clearly that for all the samples investigated, the switching field from up direction to down direction did not change much with the measurement temperature. This can be understood as this field corresponds to the difference

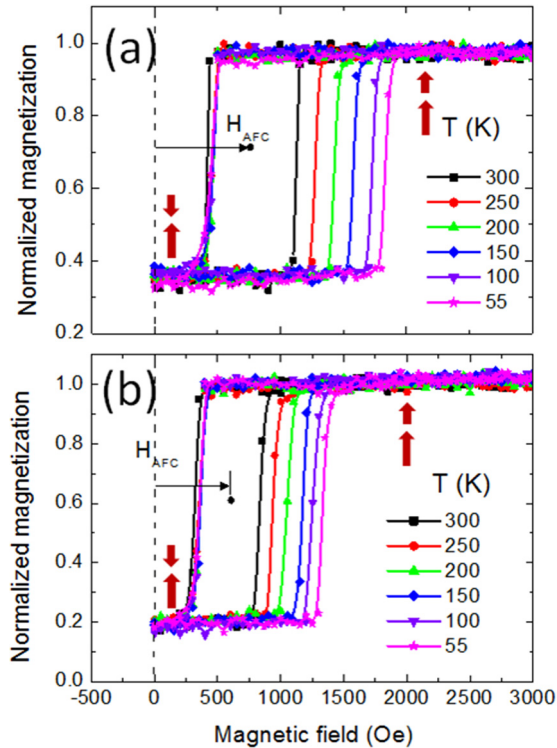


FIG. 4. Minor hysteresis loops of $[\text{Co}(t)/\text{Pd}(0.8 \text{ nm})]_{\times 3}$ multilayer antiferromagnetically coupled to $[\text{Co}(0.3 \text{ nm})/\text{Pd}(0.8 \text{ nm})]_{\times 10}$ multilayer at different temperature values. The magnetic field is applied perpendicular to film plane. (a) For $t = 0.6 \text{ nm}$ and (b) for $t = 0.8 \text{ nm}$. The antiferromagnetic field H_{AFC} was defined from the shift of the hysteresis loop for each temperature. The case of $T = 300 \text{ K}$ is shown as example for both cases a and b.

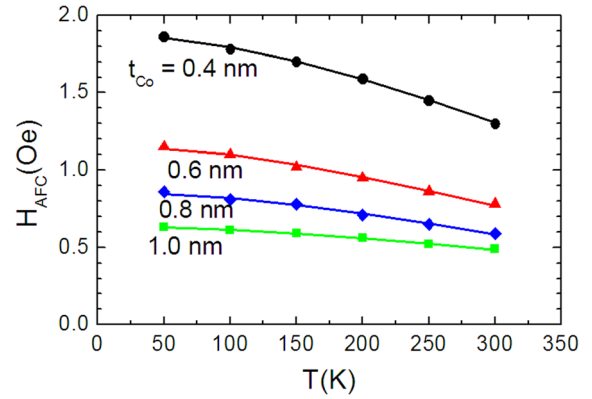


FIG. 5. Temperature dependence of the antiferromagnetic coupling field for different Co thickness values taken from the shift of the hysteresis loop of the $[\text{Co}(t)/\text{Pd}(0.8 \text{ nm})]_{\times 3}$ multilayer when it is exchange coupled to $[\text{Co}(0.3 \text{ nm})/\text{Pd}(0.8 \text{ nm})]_{\times 10}$ multilayer.

$H_{\text{AFC}} - H_c$. The decrease of H_{AFC} with temperature (plotted in Fig. 5) is balanced by a decrease of H_c with approximately the same amount. For reversing the soft layer magnetization from down direction to up direction, the antiferromagnetic coupling field has been added to the intrinsic coercivity of the layer and both are temperature-dependent. In Fig. 4, the minor loops for $t = 0.4 \text{ nm}$ and $t = 1.0 \text{ nm}$ are not shown for clarity, but H_{AFC} has the same trend as the cases (a) and (b). The temperature dependence of H_{AFC} is plotted in Fig. 5 for different thickness values of Co in the soft layer. It can be noticed that the H_{AFC} increases with a decrease in temperature. It is known that the H_{AFC} is proportional to the antiferromagnetic coupling constant J and inversely proportional to the saturation magnetization M_s . As M_s increases with a decrease of temperature, the increase of H_{AFC} at lower temperatures is expected to be due to an increase of J at lower temperature, at a rate much higher than that of $M_s(T)$. The field H_{AFC} could be well fitted with the following formula:^{22,23}

$$H_{\text{AFC}}(T) = H_0(T/T_0) \sinh(T/T_0), \quad (1)$$

where H_0 is the antiferromagnetic coupling field at 0 K and T_0 is a characteristic temperature given by^{23,24}

$$\hbar v_F / 2\pi k_B L. \quad (2)$$

Here, \hbar is the reduced Planck constant, v_F is the Fermi velocity, and L is Ru spacer thickness which is fixed to 0.8 nm. The values of H_0 and T_0 for different samples are reported in Table I. It can be noticed that the field H_0 follows an exponential decay function with temperature. This is understandable, as H_{AFC} itself is inversely proportional to t , as

TABLE I. The antiferromagnetic coupling field at 0 K H_0 and the characteristic temperature T_0 for different samples investigated are summarized.

t_{Co} (nm)	H_0 (kOe)	T_0 (K)
0.4	1.87	196.9
0.6	1.15	185.1
0.8	0.85	191.0
1.0	0.63	233.2

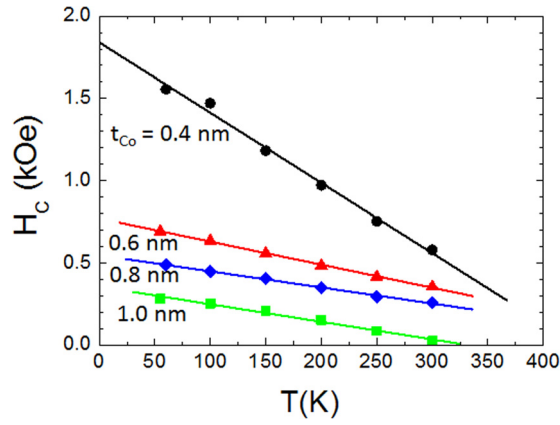


FIG. 6. Temperature dependence of the coercive field of the $[\text{Co}(t)/\text{Pd}(0.8\text{ nm})]_{\times 3}$ multilayer when it is exchange coupled to $[\text{Co}(0.3\text{ nm})/\text{Pd}(0.8\text{ nm})]_{\times 10}$ multilayer.

discussed earlier. However, this dependence is valid only in a small range of film thickness. For $t \geq 1.2\text{ nm}$, the antiferromagnetic coupling vanishes. By increasing the thickness of Co in the top soft layer, saturation magnetization becomes larger and the effective magnetic anisotropy energy K_{eff} is reduced. This reduction of K_{eff} is a consequence of an improvement of the volume contribution in the total energy which is given by

$$K_{\text{eff}} = K_v - \frac{M_s^2}{2\mu_0} + \frac{K_{\text{int}}}{t}, \quad (3)$$

where K_v is the volume anisotropy energy, K_{int} is the interface anisotropy energy, and M_s is the saturation magnetization. Although the coercivity of the hard layer did not change much for all the studied samples as expected, the soft layer coercivity was strongly dependent on the magnitude of the exchange field. Fig. 6 shows the soft layer coercivity H_C as a function of the temperature when it is antiferromagnetically coupled to $[\text{Co}(0.3\text{ nm})/\text{Pd}(0.8\text{ nm})]_{\times 10}$ multilayer. A linear decay of H_C with temperature was observed for Co thickness between 0.4 nm and 1.0 nm. In exchange coupled structures, H_C can be fitted to $H_{C0} - \chi T$ (where H_{C0} is the coercivity at 0 K and χ is a constant that depend on materials properties such as saturation magnetization M_s). For small values of M_s ($t = 0.4\text{ nm}$), χ has the largest value of 0.43 mT/K and decreases to 0.09 mT/K for $t = 1\text{ nm}$. It is important to note that for the structures selected in this study, the magnetization of the soft layer has an out-of-plane orientation although it is not the case for thicker single layer. Fig. 7 represents a comparison of M-H plots for $[\text{Co}(t)/\text{Pd}(0.8\text{ nm})]_{\times 3}$ multilayer with different Co thickness values (black dots) and exchange coupled to $[\text{Co}(0.3\text{ nm})/\text{Pd}(0.8\text{ nm})]_{\times 10}$ multilayer (red triangles). It is only for $t = 0.4\text{ nm}$ where an out-of-plane orientation of the magnetization could be seen for

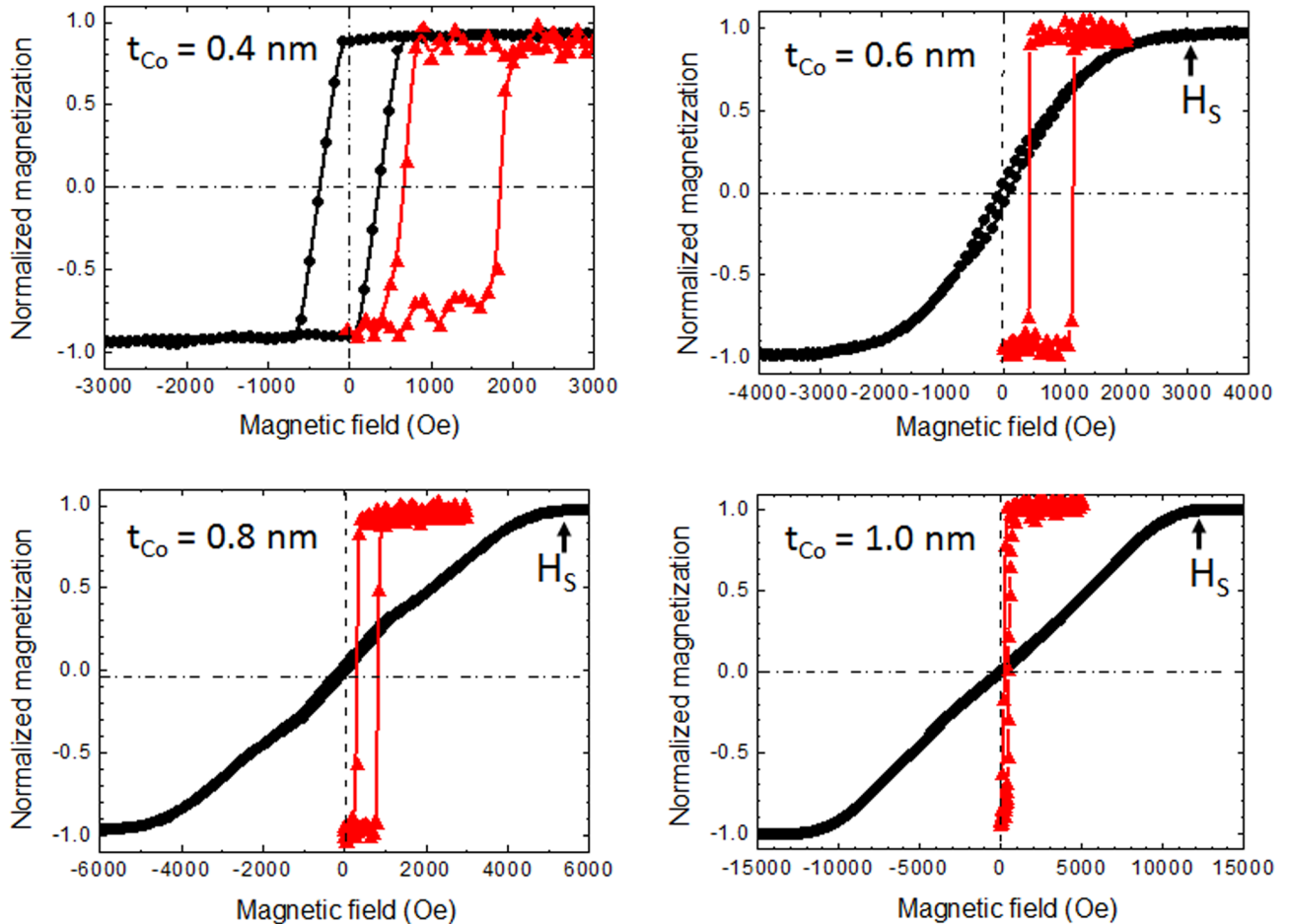


FIG. 7. Perpendicular hysteresis loops of $[\text{Co}(t)/\text{Pd}(0.8\text{ nm})]_{\times 3}$ multilayer for different Co thicknesses. The symbols \blacktriangle are when the multilayer is measured alone and \bullet when it is antiferromagnetically coupled to $[\text{Co}(0.3\text{ nm})/\text{Pd}(0.8\text{ nm})]_{\times 10}$ multilayer. The measurements were carried out at $T = 300\text{ K}$.

non-exchange coupled case. As t increases, the magnetization becomes aligned in the film plane with an increase of the saturation field H_S with thickness of Co which is an indication of a larger M_s ($H_S = 4\pi M_s$). The saturation field increases from about 0.36 T for $t = 0.6$ nm to 1.23 T for $t = 1$ nm. Two interesting results could be taken from Fig. 7. First, for $t = 0.4$ nm, an increase of the coercivity of the soft layer from 35 mT to 59 mT is observed. Second, the antiferromagnetic coupling induces a reorientation of the soft layer magnetization from in-plane to out-of-plane direction (cases of $t = 0.6, 0.8$, and 1.0 nm). It is known that for (Co/Pd) or (Co/Pt) multilayers, it is desirable to increase the thickness of Co sublayer in order to obtain a reasonably good spin polarization for magnetoresistive device application. However, the magnetization of the multilayer stack lies in the film plane due to a reduction in the interface anisotropy term as seen in Fig. 7 for t larger than 0.4 nm. Tuning the thickness of the soft layer to achieve perpendicular to plane magnetization and good spin polarization remains a challenge. As shown in this study, the antiferromagnetic coupling could help to reach these two conflicting requirements. In addition, the antiferromagnetically coupled structure is effective in minimizing the magnetic stray field especially for nanoscale devices. It is important to note that although the coercivity of coupled soft layer with t larger than 0.4 nm is only few hundreds Oersteds high, this value could be much larger at nano-scale after patterning.^{1,25}

IV. CONCLUSION

Temperature dependence of magnetization reversal of antiferromagnetically coupled (Co/Pd) multilayers was investigated. The antiferromagnetic coupling field was measured from the shift of the minor hysteresis loop of the soft layer in the out-of plane direction. The antiferromagnetic field H_{AFC} could be fitted to an analytical formula deduced the quantum-well model. The characteristic temperature and the coupling at frozen temperature could be obtained for each structure. The large values of antiferromagnetic field were the origin of magnetization reorientation of the soft layer for Co thickness larger than 0.4 nm. Achieving both relatively thicker soft layer and perpendicular magnetic orientation makes these structures useful for spintronics and patterned media applications.

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