Interfacial tuning of perpendicular magnetic anisotropy and spin magnetic moment in CoFe/Pd multilayers

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The [CoFe 4Å/Pd t]6 (t = 10−20Å) multilayers with strong perpendicular magnetic anisotropy were fabricated. Increasing the Pd spacing thickness resulted in the exponential decreases in saturation magnetization, M_s, and uniaxial anisotropy, K_u, to 155 emu/cc and 1.14×10^5 J/m^3, respectively. The X-ray absorption and X-ray magnetic circular dichroism measurements revealed a spin polarization level higher for very thin film than those of very thick Fe and Co films and the modification of the CoFe/Pd interfacial magnetism of the multilayers by the Pd spacers. It is suggested that our CoFe/Pd multilayers are excellent candidates for narrow-bit and low current spin-torque devices.

PACS numbers: 73.21.Ac, 75.30.Gw, 75.70.Cn, 72.25.Ba

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Perpendicular magnetic anisotropy (PMA) films\(^1\) have been drawn much intention because of their potential applications in magnetic recording technology and spintronics. The PMA films with high saturation magnetization ($M_s$) have been used for perpendicular magnetic recording, bit-patterned media recording technology.\(^2\) In spintronics, the PMA films have been exploited in low-energy devices, such as spin-transfer torque MRAM,\(^5\) nanowire memory,\(^6\) spin logic devices,\(^7\) etc. committed to create spin-torque devices operating with low energy consumption. Recently, the PMA films with unique magnetic properties have been extensively studied for spin-torque domain walls (DWs) nanowire devices (non-volatile memory, spin logic devices, etc).\(^6,7\) One of the most important issues of such devices is to reduce the controlled current density, of which the critical current density is essentially governed by the intrinsic parameters of the films:\(^8\)

$$j_c = \frac{e\alpha M_s^2 \Delta}{g\mu_B P} \frac{1}{|\beta - \alpha|}$$

(1)

Where, $e$ is the electron charge, $g$ is the gyromagnetic ratio, $\mu_B$ is the Bohr magneton, respectively; $\alpha$, $\beta$, $M_s$, $\Delta$ and $P$ are the Gilbert damping factor, non-adiabatic spin transfer parameter, saturation magnetization, domain wall thickness and spin polarization of the film, respectively. In the PMA films, the Bloch-type walls with narrower thickness form instead of Néel-type walls of very large wall thickness which normally exist in the thin films with in-plane anisotropy. The narrow Bloch wall is also favorable for non-adiabaticity that is expected to enhance the wall motion.\(^9\) Therefore, a PMA film with low $M_s$, low damping, narrow DW and high spin polarization would be desirable for a spin-torque DW device working with low current density.

In this letter, we present an important achievement that strong PMA multilayer films with low $M_s$, high uniaxial anisotropy and spin polarization would be made basing on fcc-Co\(_{70}\)Fe\(_{30}\) layers.
Thin Co$_{70}$Fe$_{30}$ film was chosen as the magnetic layer of the multilayer because of its high spin polarization$^{10-13}$ and low damping.$^{14}$ Although Co$_{70}$Fe$_{30}$ composition possesses the highest $M_s$ in the CoFe alloys, low $M_s$ of the CoFe/Pd multilayer would be realized using non-magnetic Pd spacing layers.

The multilayers Ta 50Å/Pd 50Å[Co$_{70}$Fe$_{30}$ 4Å/Pd $t$]$_6$/Ta 20Å ($t = 10\div20Å$) were grown on oxidized Si substrates by DC sputtering in an ultrahigh vacuum chamber of the Bestec UHV Deposition Cluster. The base pressure was better than 2.5×$10^{-9}$ Torr whereas Ar pressure was remained at 1.5 mTorr during deposition. Magnetic properties of the films were characterized using an alternating gradient force magnetometer (AGM, Micromag) with a maximum magnetic field of 20 kOe. X-ray absorption (XAS) and X-ray magnetic circular dichroism (XMCD) at Co $L_{3,2}$ and Fe $L_{3,2}$ edges were obtained by recording the sample current (total yield mode) as a function of photon energy at the SINS beam line of the Singapore Synchrotron Light Source (SSLS).$^{15}$ Elliptically polarized light with a degree of circular polarization $p = 80$ and an energy resolution of 0.25 eV was employed for the XMCD measurements. To measure the out-of-plane spin and orbital magnetic moments, the light was incident at a normal angle from the sample surface, with its propagation direction along the sample out-of-plane magnetisation direction. An external saturation magnetic field of $\pm10$ kOe was applied to magnetize the sample along the out-of-plane direction. The XCMD were done by changing the direction of applied magnetic field while kept the helicity of the light. We intentionally measured XMCD with spin polarization along out-of-plane magnetic moments to follow an easy-axis magnetization shown by the magnetometer measurements. Micromagnetic simulation of domain structure was performed using LLG Micromagnetic Simulator$^\text{TM}$.16
Figure 1 shows a typical picture of hysteresis loops measured on two directions for CoFe/Pd multilayers \((t = 10\sim20\,\text{Å})\): parallel (or in-plane) and perpendicular (or out-of-plane) to the film plane. The hysteresis loop measured in-plane of the film exhibited a hard axis behavior characterized with rotation mechanism of the magnetization reversal denoted by an S-shape loop. Whilst it was an easy-axis loop measured out-of-plane reflected by the square-shape hysteresis loop. Smooth hysteresis loops indicated that unique magnetic properties occurred over the film as a single phase behavior due to strong ferromagnetic coupling between the magnetic CoFe layers. These proved that the magnetic anisotropy of such CoFe/Pd multilayer has been well aligned normal to the film plane. Interestingly, such strong PMA was also observed in range of Pd spacing thickness from 10 to 20 Angstroms.

Usually, the Co-Fe alloy with a concentration of Fe less than 35 at.% is favorable for the formation of body-centered cubic crystal structure,\(^{17}\) and the magnetic anisotropy should appear in-plane. However, the thick Pd seed layer in face-centered cubic (fcc) with a strong \(<111>\) texture normal to the film plane proved previously\(^{18}\) would induce the \(<111>\)-texture fcc structure in the CoFe to support the out-of-plane anisotropy.\(^{17,19}\) It was also attributed that the interfacial anisotropy of the CoFe/Pd interface, in which the CoFe layer was in \(<111>\)-texture fcc, would be an important factor governing the perpendicular magnetic anisotropy of the CoFe/Pd multilayers.\(^{20}\) The contribution of the CoFe/Pd interfacial magnetism to the magnetic properties of the films will be revealed using XMCD measurements as discussed later.

It is attractive to visualize the modification of magnetic properties of the CoFe/Pd multilayers by changing the thickness of the Pd as a spacing layer. Fig. 2(a) illustrates the saturation magnetization, \(M_s\), of the CoFe/Pd multilayer as a function of Pd spacing thickness. Obviously, \(M_s\) appeared to decrease with the Pd thickness due to diluting the magnetic interaction by the
non-magnetic Pd layers. With Pd spacing layers of 10Å thick, the $M_s$ value was measured to be 280 emu/cc, whereas this value exponentially decreased to 155 emu/cc as the thickness of the spacing layers increased to 20Å. Such low $M_s$ values were significantly lower than that in other common Co-based PMA films used for spin-torque domain wall applications, such as Pt/Co multilayers (~900 emu/cc),\textsuperscript{21} Co/Ni multilayers (660 emu/cc),\textsuperscript{6} perpendicular magnetized CoFeB films (1200 emu/cc),\textsuperscript{22,23} and comparable with low-$M_s$ TbFeCo film.\textsuperscript{24} The $M_s$ of the studied CoFe/Pd multilayers was definitely much lower than that of pure fcc-Co\textsubscript{70}Fe\textsubscript{30} alloy film (1900 emu/cc).\textsuperscript{25} This reduction was attributed to the weakening of ferromagnetic coupling between CoFe layers by the Pd spacing layer.\textsuperscript{18}

In contrary to the tendency of the $M_s$, the anisotropy field $H_k$, which was derived from the hysteresis loops as $H_k = H_s + 4\pi M_s$ ($H_s$ is the saturation field),\textsuperscript{22} increased as a linear function of the Pd layer thickness [see Fig. 2(b)] from 12.6 kOe to 14.8 kOe. From $H_k$ and $M_s$, the uniaxial anisotropy $K_u$ would be determined as $K_u = H_k M_s / 2$ and shown in Fig. 2(c). Figure 2(c) presents how the uniaxial anisotropy constant, $K_u$, varies with the Pd thickness. It is clearly seen that the insertion of Pd space layers supported to reduce the uniaxial anisotropy of the multilayers. Namely, with the Pd space layers of 10Å thick, the $K_u$ was obtained as high as $1.77 \times 10^5$ J/m$^3$. Increasing the Pd thickness to 20Å unexpectedly led to an exponential reduction of the $K_u$ to $1.14 \times 10^5$ J/m$^3$. Fortunately, such a high uniaxial anisotropy could be comparable to other common materials. Furthermore, the uniaxial anisotropy allows the domain wall thickness to be estimated using the relation: $\Delta = \pi \sqrt{A / K_u}$ with $A$ is the exchange constant. Assume $A \sim 10$ pJ/m (typically for Co-based magnetic thin films), the domain wall thickness would be estimated to be around 20-25 nm in scale of the Co/Pd multilayers and other Co-based multilayers.\textsuperscript{26} The exchange interaction length could be deduced from the $M_s$ by using the
relation $l_{\text{ex}} = \sqrt{2A / \mu_0 M_s^2}$. This value was in range of 7-12 nm, which is just about the overall thickness of the multilayers, supporting the thin magnetic layers ferromagnetically to couple to each other over the thickness scale as discussed above.

The commercial simulation package LLG$^\text{TM}$ was used to simulate the domain pattern and wall structure of the films. Fig. 3 depicts a typical picture of the simulated domain pattern in the multilayer with the Pd thickness of 10Å. A stripe-like domain structure, which looks similar to that of the PMA FePd film reported previously, was observed in all the multilayer samples with a very well-defined period of about 400 nm. This was also the size of the domains in some PMA multilayers based on Co. For the stripe domains, it is important to take into account the quality factor which is given by:

$$Q = \frac{2K_u}{\mu_0 M_s^2}$$ (2)

In our studied multilayers, the Q factor was ranging from 3.5 to 7.6, indicating that the anisotropy energy term was dominant to form a sharply defined domain state as seen in Fig. 3(a) with the Bloch-type domain walls extending right up the surface of the films with a weak Néel caps in similar size of the Bloch walls as described in previous reports. The simulated results shown in Fig. 3 somehow affirmed this suggestion. Namely, perfectly perpendicular magnetization in the domains was obviously visible via the blue-red color whilst the Néel caps would be visualized as the in-plane components located at the domain walls [Fig. 3(b)]. The Néel caps would bring a benefit that the domain walls could be easily nucleated with low local field applied to the film. This allows explaining why the out-of-plane hysteresis loops were not perfectly sharp as square shape but slightly reduced from remanent state to the switching state (Fig. 1).
The final important aspect is to discuss about the electronic structure and spin polarization of the films revealed using XAS and XMCD. Figure 4(a, b) shows the XAS and XMCD at Co $L_{3,2}$ and Fe $L_{3,2}$ edges, which are Co $2p \rightarrow 3d$ and Fe $2p \rightarrow 3d$ transitions, respectively. Thus, because of dipole selection rule, these transitions are element specific and extremely sensitive to electronic structure and spin polarization at the Co $3d$ and the Fe $3d$ bands. Furthermore, due to strong spin-orbit coupling, XAS at Co $L_{3,2}$ edges show two peaks, at ~778 eV for $L_3$ and at ~794 eV for $L_2$, while XAS at Fe $L_{3,2}$ edges shows two strong peaks, at ~708 eV for $L_3$ and ~721 eV for $L_2$ in the CoFe layers. In Figure 4(a), we present XAS and XMCD on CoFe layers of the multilayer film with a Pd spacing thickness of 10Å as a typical example. A magnetic field of ±10 kOe was applied out-of-plane direction and the XMCD signal was deduced by subtracting two XAS signals at the opposite magnetic field directions, namely positive and negative fields. Standard Co and Fe films were used as the reference samples to calibrate the XAS and XMCD signals (not shown). We have observed strong ferromagnetic properties in the films, which could be referred as two well-defined peaks at both edges, $L_3$ and $L_2$ of Co and Fe, in the XMCD signal and the ferromagnetism in the CoFe/Pd multilayer results from the Co $3d$ and Fe $3d$ states, essentially. This XMCD result is important because the observed ferromagnetism is truly from intrinsic properties of the films. The enhancement of the normalized peak in the XAS signal of the CoFe/Pd multilayer films comparing to those of Co and Fe reference signals (data not shown) indicated the strong ferromagnetic interaction between Co-Fe ions in the CoFe lattice which is well-known as the origin of the high magnetic moment in the Co-Fe alloy system.\textsuperscript{30}

One of great advantages of XMCD data is its capability to reveal the spin magnetic moment ($\mu_S$) and orbital magnetic moment ($\mu_L$).\textsuperscript{31} By applying the X-ray MCD sum rule,\textsuperscript{32,33} we have estimated $\mu_S$ and $\mu_L$ based on the following equations.
\[ m_{\text{spin}} = -\frac{1}{\cos \theta \times \text{CPD}} \times \left\{ 6 \left[ L_3 (\mu_+ - \mu_-) d\omega - 4 \right] \frac{L_3 + L_2 (\mu_+ - \mu_-) d\omega}{L_3 + L_2 (\mu_+ + \mu_-) d\omega} \right\} \times \left[ 0 - n_{3d} \left( 1 + \frac{7 T_z}{2 S_z} \right) \right]^{-1} \] (3)

\[ m_{\text{orb}} = -\frac{1}{\cos \theta \times \text{CPD}} \times \left\{ 4 \frac{L_2 + L_2 (\mu_+ - \mu_-) d\omega}{3 L_2 (\mu_+ + \mu_-) d\omega} \right\} \times \left[ 0 - n_{3d} \right] \] (4)

where \( n_{3d} \) is the 3\textit{d} electron occupation number, \(<T_z>\) is the expectation value of magnetic dipole operator, \(<S>_z\) is equal to half of the \( m_{\text{spin}} \) in Hartree atomic units, \( \theta \) is photon incident angle which is 90°, and \( \text{CPD} \) is circular polarization degree which is 0.8. Based on band structure calculations, the \(<T_z>/<S>_z\) is negligible. The results are shown in Figure 4(c,d,e) and Figure 5. 3)

The spin magnetic moment observed in the XMCD signals [Fig. 4(c), Fig. 5] reveals that spin polarization associated to the spin magnetic moment\(^{34}\) was evidenced. As a result, by normalizing the spin magnetic moment of the CoFe/Pd multilayer films to the reference Co and Fe samples, the field-induced spin polarization in the CoFe/Pd multilayer films could be estimated. By comparing to the spin magnetic moment of the Co and Fe reference films, the spin polarization in the studied CoFe/Pd multilayers was evaluated to be ~40-60\%, which is remarkably higher than those in our reference Co and Fe thicker films (~40\%).\(^{30}\) This indicated that our CoFe/Pd multilayers are superior in spin polarization to other Co-based multilayers. Interestingly, Fig. 4(c) and Fig. 5 indicated that spin magnetic moment and total magnetic moment were apparently decreased by varying the Pd spacing thickness. This is consistent with our AGM result. Because the thickness of the CoFe magnetic layers was fixed, such decreases in spin and total magnetic moments could be assigned to the contribution of the interfacial magnetism of the CoFe/Pd interfaces. This is an evidence to prove that the interfacial anisotropy significantly affected the magnetic anisotropy of the films. This is also similar to the XMCD results of the Co/Pd multilayers in which the magnetic properties were argued to be partially
contributed from the magnetic polarization of Pd 4d that was caused by hybridization with the Co 3d electron.\textsuperscript{35}

In conclusion, the Co\textsubscript{70}Fe\textsubscript{30}/Pd multilayers with strong perpendicular magnetic anisotropy and modifiable magnetic properties have been fabricated. Saturation magnetization as low as 155 emu/cc and narrow Bloch-wall type as thin as 20-25 nm were obtained by increasing the Pd spacing thickness to 20Å. Using XAS and XMCD measurements, spin polarization was observed and estimated (~40-60\%) to be higher in the very thin film CoFe than those in the thicker Co and Fe films. Furthermore, the XAS and XMCD spectra revealed the modification of electronic structure of CoFe/Pd interfacial magnetic moments by changing the Pd spacing layer, contributed to the magnetic properties of the films. These advantages indicated that our studied CoFe/Pd multilayers are of superior characteristics for spin-torque applications.

**Acknowledgements**

This work was completed by a financial support from the A*STAR (SERC) Public Sector Funding (Grant No. 092 151 0087). We also acknowledge MOE-AcRF-Tier-2 and CRP Awards No. NRF-CRP 8-2011-06 and NRF-CRP 8-2009-024 for the works performed at Singapore Synchrotron Light Source. D.-T. Ngo would like to thank Prof. Michael R. Scheinfein (Arizona State University in Tempe, Arizona) for his kind support in LLG simulation.
References


16. Available at: http://llgmicro.home.mindspring.com/


**Figure captions**

FIG. 1. M-H hysteresis loops of the CoFe/Pd multilayers with different Pd spacing thickness. The loops with circle dots were measured out-of-plane of the films, and the solid plots denoted the in-plane loops.

FIG. 2. (a) Saturation magnetization, (b) anisotropy field and (c) uniaxial anisotropy as the functions of the Pd spacing thickness.

FIG. 3. Simulated domain pattern in the CoFe/Pd multilayer with a spacing thickness of 10Å: (a) out-of-plane induction component and (b) in-plane induction components.

FIG. 4. (a,b) XAS and XMCD spectra of the Fe and Co atoms in the CoFe/Pd multilayer with a spacing thickness of 10Å, and (c-e) elementary spin and orbital magnetic moments Fe and Co in the CoFe/Pd multilayers derived from XMCD spectra.

FIG. 5. Spin, orbital and total magnetic moments of the CoFe system obtained by summing the constituent elements Co and Fe.