The study of perpendicular magnetic anisotropy in the magnetic sensors with linear sensitivity using polarized neutron reflectometry

T. Zhu
China Spallation Neutron Source, Institute of High Energy Physics, Chinese Academy of Sciences, Dongguan 523803, P.R. China
Institute of Physics, Chinese Academy of Sciences, Beijing 100190, P. R. China

E-mail: tzhu@aphy.iphy.ac.cn

Abstract. The CoFeB sandwiched by Ta and MgO layers enables a perpendicular magnetic anisotropy (PMA) and provides a pathway for such application. In this paper, we reported the origin of PMA in CoFeB using the anomalous Hall effect (AHE) and polarized neutron reflectometry (PNR). From PNR experiments, we obtained the details of the magnetic and structural depth profiles inside the film. It is found that the PMA properties of CoFeB layers deposited above and under MgO layer are different and PNR measurements confirmed that a large PMA in the CoFeB above MgO layer is related to its low magnetization. Based on this PMA mechanism, we obtain a high sensitivity of AHE in the perpendicular CoFeB, which opens a new avenue to detect ultralow magnetic field.

1. Introduction
The magnetic sensors for high sensitive magnetic detecting have been reported using magnetoresistive effect or anomalous Hall effect, where a linear and reversible response is produced [1-5]. For example, detection of specific biomolecular interactions through sensing of nanoscopic magnetic labels provides one of the most promising routes towards biosensing with solid-state devices [3-5]. Typically, these magnetic nanoparticles are superparamagnetic or non-remanent ferromagnetic particles and can be attached to target biomolecules. Under a small magnetic field, these particles acquire a magnetic moment which produces a fringe field over the sensor [3]. In each probe, a magnetic sensor or Hall sensor detects the stray field produced by the label giving an electrical signal [3,4]. There are several methods to detect such weak remanent magnetic field. The sensors for biosensing have been reported using magnetoresistive effect [3]. By using a complicated design with an orthogonal alignment between the sensing and reference magnetization, a linear and reversible giant magnetoresistance or tunneling magnetoresistance response is produced [6]. On the other hand, micro-Hall magnetic sensors based on semiconductor heterostructures have been attracting wide interests [4,5]. As a simple solution, the linear anomalous Hall effect (AHE) exhibits fast response, high sensitivity, and ease of integration due to the cheap and simple fabrication process [1,2]. The Hall resistance in a magnetic thin film consists of two terms: one presents the ordinary Hall effect and another presents the AHE. The sensitivity of linear anomalous Hall effect ($S_{AHE}$) is defined as the ratio of anomalous Hall resistance ($R_{AHE}$) to the out-of-plane saturation field ($H_s$),
$R_{\text{AHE}}/H_c$. So far, large SAHE has been observed in Co(Fe)/Pt multilayers or granular films [8,9], due to the induced perpendicular magnetic anisotropy in these thin films.

Recently, the magnetization of CoFeB has been reported to stay out of plane when it is sandwiched by MgO and Ta layers [10-12]. In this paper, we reported that the effective magnetic anisotropy of CoFeB layer in the MgO/CoFeB/Ta thin film depended on the layer deposition sequences using the polarized neutron reflectometry (PNR). And then, we demonstrate a giant linear anomalous Hall effect in the MgO/CoFeB/Ta thin film, which detects the weak magnetic field from a superparamagnetic label.

2. Method

2.1. Sample preparation

MgO (1-5)/CoFeB (0.8-3)/Ta (0.55-5) samples were deposited on a thermally oxidized silicon wafer by magnetron sputtering with a base pressure of $5\times10^{-6}$ Pa. The numbers in brackets are nominal thicknesses in nanometers. Ar was used as the sputtering gas and the sputtering pressure for metals was 0.4 Pa. MgO layers was deposited by radio-frequency (RF) sputtering and the sputtering pressure was 0.2 Pa. High purity of Ta (99.95%), MgO (99.99%), and Co$_{40}$Fe$_{40}$B$_{20}$ (99.9%, Functional Materials International, Japan) were used as the target materials. All the samples were capped with a 3-nm-thick MgO to prevent oxidation. The layer thickness was identified by an x-ray reflectometer (XRR, Brucker D8).

2.2. Anomalous Hall effect measurements

The patterned Hall bar samples were annealed from 90 to 450°C for 1 hour. Room temperature anomalous Hall resistivity, $\rho_{\text{AHE}}$, and longitudinal resistivity, $\rho_{\text{xx}}$, were measured in a commercial Hall probing system by using an electromagnet (up to 1.5 T) and high resolution multimeters (Keithley® 220 and 2182). To thoroughly eliminate the ordinary Hall effect, $R_{\text{AHE}}$ is defined as the zero field extrapolation of the high field data. The low temperature AHE measurements were carried out by using standard cross-structure four-point setting in a physical property measurement system (PPMS, Quantum Design® PPMS-14T).

2.3. Polarized neutron reflectivity measurements

The PNR technique was used to directly measure the depth-dependent magnetization profile of the samples to investigate the contribution of the demagnetization term to PMA. Room-temperature PNR measurements were carried out on the Magnetism Reflectometer (BL-4A) at the Spallation Neutron Source at Oak Ridge National Laboratory [13].

![Figure 1](image)

**Figure 1.** The schematics of the PNR measurement for a perpendicular thin film under (a) a low and (b) a high external fields.

In general, PNR is a technique sensitive to the compositional and in-plane magnetic depth profiles of thin films but not sensitive to the layer magnetization perpendicular to the sample surface, because the magnetic neutron scattering cross section is sensitive only to components of $\mathbf{M} \perp \mathbf{Q}$ [14]. $Q = \ldots$
4πsinθ/λ, where λ is the neutron wavelength and θ the incident angle of the neutron beam, respectively. Hence, it needs a high external field to saturate the film perpendicular to Q. Figure 1 shows the schematics of the PNR measurement for a perpendicular thin film. There is no in-plane component of M under the low external field (figure 1(a)). The reflectivity curves look like unpolarized neutron reflectivity. On the other hand, the magnetization will align with the external field when it is large enough (figure 1(b)), and then the magnetic depth profiles of thin films have been obtained.

3. Results and Discussions

3.1. Perpendicular magnetic anisotropy

Figure 2 shows the CoFeB thickness dependence of ρ_{AHE} and ρ_{xx} for annealed MgO (1.1)/CoFeB (t)/Ta (2.2) thin films (annealing temperature T_a = 300°C). The insets show the representative Hall resistance hysteresis loops (R_{xy}-H) with t_{CoFeB}. Similar with previous results [10,11], obvious perpendicular magnetic anisotropy has been obtained when CoFeB thickness ranging from 1.06 to 1.5 nm in the Ta capping films.

Figure 2. CoFeB thickness dependence of ρ_{AHE} (the solid circles) and ρ_{xx} (the open circles) for annealed MgO (1.1)/CoFeB (t)/Ta (2.2) thin films (T_a = 300°C) with S defined as super-paramagnetic, ⊥ as magnetic easy axis perpendicular to film, and // as magnetic easy axis in the plane. The insets show the representative Hall resistance hysteresis loops.

Furthermore, ρ_{AHE} of the samples are larger than those of ordinary transition metals (~1 μΩ cm [7]). As for longitudinal resistivity, ρ_{xx} of all the annealed samples is several hundred μΩ cm and increases with the decrease of CoFeB thickness. The resistivity of tantalum is about 200 μΩ cm due to its extremely short mean free path (1 nm) [15]. Meanwhile, the resistivity of CoFeB is also much higher (about 160 μΩ cm) [16]. Due to these layers with high resistivity, MgO/CoFeB/Ta thin film has large ρ_{xx}, which is much larger than those for a traditional Pt based multilayer or alloy. The large ρ_{xx} itself is very important, because low ρ_{xx} results in a blockade to reduce the device size [17].

The sample for PNR measurement was Ta (2.2)/CoFeB (1.04)/MgO (1.1)/CoFeB (1.22)/Ta (2.2) (T_a = 300°C), hereafter defined as Ta/CoFeB/MgO/CoFeB/Ta, with the sample size of 10×15 mm². Figure 3(a) shows the R_{xy}-H loop of the sample. Apparently, the PMA properties of CoFeB layers deposited above and under MgO layer are different. For comparison, two samples Ta (2.2)/CoFeB (1.04)/MgO (1.1) and MgO (1.1)/CoFeB (1.22)/Ta (2.2), hereafter defined as MgO/CoFeB/Ta and Ta/CoFeB/MgO, respectively, were prepared on Si/SiO₂ (T_a=300°C). As seen from Fig. 3(b), the coercivity field (H_C) of MgO/CoFeB/Ta is nearly 100 Oe, which is about 24 Oe in Ta/CoFeB/MgO, which is consistent with the H_C of the related CoFeB layers in the sample for the PNR measurement, i.e. the CoFeB above or below the MgO, as shown in Fig. 3(a). The results also confirm that the PMA properties of CoFeB are dependent on the stack deposition sequence.
Figure 3. Hall resistance hysteresis loops of (a) Ta(2.2)/CoFeB(1.04)/MgO(1.1)/CoFeB(1.22)/Ta(2.2) (blue solid circles) and (b) MgO(1.1)/CoFeB(1.22)/Ta(2.2) (red solid squares) and Ta(2.2)/CoFeB(1.04)/MgO(1.1) (red open squares) films annealed at 300 °C.

3.2. Polarized neutron reflectivity

In a reflectometry experiment, the interaction of neutrons with a magnetic film is described by the Fermi pseudopotential \( V_\pm \),

\[
V_\pm = \pm \frac{2\hbar}{m} N(b_n \pm b_m),
\]

where \( m \) denotes the neutron mass, \( N \) the atomic number density, \( b_n \) the nuclear scattering length, and \( b_m \) the magnetic scattering length, respectively [18]. For the two neutron spin states, the magnetic scattering length adds to or subtracts from the nuclear one [19], leading to different momentum transfer \( Q \) dependence of reflectivity curves, \( R^+ \) and \( R^- \), and is used to determine the magnetic moment of magnetic films. Here, \( R^+ \) denotes the reflectivity for spin-up neutrons with spin parallel to \( H_{\text{ext}} \), and \( R^- \) denotes the reflectivity for spin-down neutrons with spin antiparallel to \( H_{\text{ext}} \). The sample’s depth-dependent nuclear scattering length density \( N_{bn} \) and magnetic scattering density \( N_{bm} \), hereinafter defined as SLD\(_n\)(\( z \)) and SLD\(_m\)(\( z \)), respectively, and then the magnetization are determined by a simultaneous fit of the PNR data.

Figure 4. The experimental \( R^+ \) (open squares) and \( R^- \) (open circles) measured at room temperature and (a) \( H_{\text{ext}} = 100 \) Oe and (b) \( H_{\text{ext}} = 10 \) kOe as functions of \( Q \), respectively. The solid lines are the fitted theoretical curves.
Figure 4(a) shows $R^+$ (open squares) and $R^-$ (open circles) as a function of $Q$ measured in an external field of 100 Oe after the saturation. This external field is much smaller than the out-of-plane anisotropy field. In such low external field, the magnetization $M$ is perpendicular to the plane of the sample and parallel to $Q$. Meanwhile, the small external magnetic field $H_{\text{ext}}$ provides a guide field for the polarized neutron beam. This is confirmed by the PNR data, when the reflectivities for the two spin-states, $R^+$ and $R^-$, overlap. Thus $R^+$ and $R^-$ have the same scattering vector $Q$ dependence when $H_{\text{ext}}$ is much smaller than the out-of-plane anisotropy field of a PMA film.

To determine the magnetization profile in the sample, a large in-plane field is applied to saturate the out-of-plane magnetization into the hard in-plane direction. Figure 4(b) shows the PNR data measured at room temperature with $H_{\text{ext}} = 10$ kOe, which is sufficient to obtain the in-plane saturation of the magnetization. This is to say, $M$ is in the plane of the sample and perpendicular to $Q$ when $H_{\text{ext}} = 10$ kOe.

Figure 5 shows the structural and magnetic profiles obtained from a fit to the data. First, we fitted 100 Oe PNR data to get the parameters of the layer structure as seen in the left and middle panels of figure 5. Second, we fixed these structural parameters to fit the 10 kOe PNR data to get the magnetization of the individual layer of the sample as seen in the right panel of figure 5.

![Figure 5](image)

**Figure 5.** The depth-dependence of Im-SLD$_N(z)$ (left), SLD$_N(z)$ (middle), and magnetization of CoFeB layer from SLD$_M(z)$ (right). The thin green lines show the thicknesses of layers in the sample obtained from the fit to the data.

In the CoFeB sandwiched by MgO and tantalum layers, the effective magnetic anisotropy can be generally defined as

$$K_{\text{eff}} = (K_b - 2\pi M_{S,\text{under}}^2) + 2K_S/t_{\text{CoFeB}}$$

where $t_{\text{CoFeB}}$ is the CoFeB thickness and $M_S$ is the saturation magnetization of the CoFeB layer. $K_b$ is the bulk crystalline anisotropy, which can be neglected [10]. Therefore, the perpendicular anisotropy in the CoFeB is the result of the competition between the demagnetization energy ($2\pi M_{S,\text{above}}^2$) and the interfacial anisotropy energy ($K_S/t_{\text{CoFeB}}$). One pronounced feature from the PNR results is that the magnetization of the CoFeB layer above MgO ($M_{S,\text{above}} = 663$ emu/cm$^3$) is significantly smaller than that of the CoFeB layer under MgO ($M_{S,\text{under}} = 906$ emu/cm$^3$). According to Eq. 2, the smaller demagnetization term ($2\pi M_{S,\text{above}}^2$) will induce a larger positive $K_{\text{eff}}$ and a larger switching field. On the other hand, the demagnetization term of CoFeB under MgO ($2\pi M_{S,\text{under}}^2$) is much larger, leading to a dramatic decrease in $K_{\text{eff}}$.

The mechanism of the difference in magnetization between CoFeB layers above and below MgO layers remains unclear, but it may be related to the structural dependent formation of magnetically dead layer (MDL) at the CoFeB-Ta interface during deposition, or intermixing upon annealing, or the roughness of the underlayer on which CoFeB layer was deposited [20]. In general, a reduction of the magnetization in a thin magnetic film is normally believed to be due to the presence of a MDL at the interface. For CoFeB-Ta interface, there is an approximately 0.5-nm-thick MDL in the case of the
deposition of CoFeB above MgO (i.e. MgO/CoFeB/Ta) but no MDL in the case of the deposition of CoFeB under MgO (i.e. Ta/CoFeB/MgO) [10,21,22]. Based on the PNR fitting results, the SLDN of CoFeB above MgO is 4.35×10−6 Å−2, whereas the SLDN of CoFeB under MgO is 5.03×10−6 Å−2. It suggests the quality of CoFeB layers is one of possibilities for the magnetization reduction besides of the effect of MDL. The observed low magnetization of the CoFeB layer above MgO may be due to the formation of a loose structure in the CoFeB layer during the deposition on MgO.

Another pronounced feature from the PNR results is the observation of boron diffusion after annealing. The exceptional sensitivity of neutrons to boron because of its absorption cross section is a distinct advantage and allows accurate determination of the depth profile of the boron distribution. As shown in Fig. 5, the interface layers between the CoFeB and tantalum layers are 2 nm (near top of sample) and 2.2 nm (near bottom of sample), hereinafter defined as XT and XB, respectively. The SLDN of XT is 6.1721×10−6 + 2.2×10−7i Å−2, with the second term defined as Im-SLDN from the absorption cross section, whereas the SLDN of XB is 5.4878×10−6 + 2×10−7i Å−2. The nominal SLD of boron is 6.94×10−6 + 2.79×10−7i Å−2 [23]. From the fit to the PNR data, we observed that after the annealing, most of the boron diffused from the CoFeB layers where it was initially found to form 2-nm-thick interface layers, which are a mixture of boron and tantalum.

### 3.3. Linear anomalous Hall effect

Figure 6 shows the representative $R_{xy} - H$ loop of MgO (2.5)/CoFeB (1.2)/Ta (0.7)/MgO (3) thin film annealed at 210 °C. The magnetization turns to out-of-plane when $H$ reaching a small field, which is the saturated field $H_s$. Beyond the $H_s$, the anomalous Hall resistance accesses its saturation soon. And then a large linear $R_{xy} - H$ loop is observed. The $S_{\text{AHE}}$ at room temperature is 2376 Ω/kOe when $t_{Ta} =$ 0.7 nm, which is 21 times larger than that for the best semiconductor (110 Ω/kOe for InSb [5]).

![Figure 6. The representative curves of $R_{xy} - H$ for MgO (2.5)/CoFeB (1.2)/Ta (0.7)/MgO (3) film ($T_a =$ 210°C). The red solid line is guide for eyes.](image)

Besides that obvious PMA has been obtained when the CoFeB thickness ranging from 1.06 to 1.5 nm in the MgO/CoFeB/Ta thin films annealed at 300°C, linear $R_{xy} - H$ curves have been observed in the films with thin CoFeB layers ($t_{\text{CoFeB}} \leq 1$ nm) and thick CoFeB layers ($t_{\text{CoFeB}} > 1.5$ nm) as shown in the insets of Fig. 2. It should be pointed out that those two magnetization states are different. The film with thin CoFeB layer shows out-of-plane superparamagnetic at room temperature, because out-of-plane ferromagnetic returns at low temperature. On the other hand, the film with thick CoFeB layer shows in-plane ferromagnetic state when $t_{\text{CoFeB}} > 1.5$ nm.

According to Eq. 2, $K_{\text{eff}}$ is the result of the competition between the demagnetization energy ($2\pi M_s^2 t_{\text{CoFeB}}$) and the interfacial anisotropy energy. The magnetization of thin film shows out-of-plane when $K_{\text{eff}} > 0$. From the PNR measurement, the PMA in CoFeB above MgO layer is due to its low magnetization. Hence, it suggested the MgO/CoFeB/Ta thin film can be a good candidate to easily tune the PMA strength. Further study shows that the PMA strength can be simply tuned by MgO.
thickness due to the interface structure between CoFeB and MgO layers [24]. From the definition of \( S_{\text{AHE}} \), there are two kinds of methods to achieve the sensitivity of AHE: one is to reduce the \( H_S \) and another is to enhance the \( R_{\text{AH}} \). Thus, a large linear \( R_{xy} - H \) loop can be obtained, when \( K_{\text{eff}} < 0 \) but is very close to 0. And then, a high sensitivity of AHE can be obtained in the perpendicular CoFeB thin film.

4. Summary
In summary, the PMA properties of CoFeB sandwiched between MgO and tantalum layers have been systematically investigated using AHE and PNR. A large PMA in the CoFeB above MgO layer is due to its low magnetization. Furthermore, we demonstrate a giant linear anomalous Hall effect in the perpendicular CoFeB thin film. Our results open a new avenue to detect weak magnetic field by using highly sensitive AHE of perpendicular CoFeB

References
[1] Zhu T 2014 Chin. Phys. B 23 047504
[2] Zhu T, Chen P, Zhang Q H, Yu R C and Liu B G 2014 Appl. Phys. Lett. 104 202404
[3] Freitas P P, Ferreira R, Cardoso S and Cardoso F 2007 J. Phys.: Condens. Matter 19 165221
[4] Manandhar P et al. 2009 Nanotechnology 20 355501
[5] Gilbertson A M et al. 2011 Appl. Phys. Lett. 98 062106
[6] Teixeira J M et al. 2012 J. Appl. Phys. 111 053930
[7] Gerber A 2007 J. Magn. Magn. Mater. 310 2749
[8] Zhu Y and Cai J W 2007 Appl. Phys. Lett. 90 012104
[9] Lu Y M, Cai J W, Pan H Y and Sun L 2012 Appl. Phys. Lett. 100 022404
[10] Ikeda S et al. 2010 Nature Mater. 9 721
[11] Worledge D C et al. 2011 Appl. Phys. Lett. 98 022501
[12] Zhu T, Yang Y, Yu R C, Ambaye H, Lauter V and Xiao J Q 2012 Appl. Phys. Lett. 100 202406
[13] Lauter V, Ambaye H, Goyette R, Lee W H and Parizzi A 2009 Physica B 404 2543
[14] Bland J A C, Bateson R D, Heinrich B, Celinski Z and Lauter H J 1992 J. Magn. Magn. Mater. 104–107 1909
[15] Feldman B and Dunham S T 2009 Appl. Phys. Lett. 95 222101
[16] Jen S U et al. 2006 J. Appl. Phys. 99 053701
[17] Moritz J, Rodmacq B, Auffret S and Dieny B 2008 J. Phys. D: Appl. Phys. 41 135001
[18] Rauch H and Petraschek D 1978 Neutron Diffraction Topics in Current Physics vol 6 ed H. Dachs, (Springer, New York)
[19] Felcher G P 1993 Physica B 192 137
[20] Yamanouchi M et al. 2011 J. Appl. Phys. 109 07C712
[21] Jang S Y, Lim S H and Lee S R 2010 J. Appl. Phys. 107 09C707
[22] Meng H, Lum W H, Sbiaa R, Lua S Y H and Tan H K 2011 J. Appl. Phys. 110 033904
[23] Sears V F 1992 Neutron News 3 26
[24] Zhu T, Chen P, Zhang Q H, Yu R C and Liu B G arXiv:1405.2551

Acknowledgement
This work has been supported by the National Basic Research Program of China (2012CB933102) and National Science Foundation of China (Grants 11574375 and 11174354). Research at Oak Ridge National Laboratory’s Spallation Neutron Source was sponsored by the Scientific User Facilities Division, Office of Basic Energy Sciences, US Department of Energy. The author thanks Dr. V. Lauter and Dr. H. Ambaye from ORNL for her help with the PNR measurement and analysis.