Applied Surface Science 433 (2018) 556–559

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Full Length Article

# Interfacial chemical structure-modulated anomalous Hall effect in perpendicular Co/Pt multilayers

Qian Liu[a,*,1], Shaolong Jiang[b,*,1], Jiao Teng[c]

[a] College of Metallurgy and Energy, North China University of Science and Technology, Tangshan 063210, China
[b] Department of Materials Science and Engineering, College of Engineering, Peking University, Beijing 100871, China
[c] Department of Materials Physics and Chemistry, University of Science and Technology Beijing, Beijing 100083, China

ARTICLE INFO

*Article history:*
Received 20 August 2017
Received in revised form 5 October 2017
Accepted 14 October 2017
Available online 16 October 2017

*Keywords:*
Anomalous Hall effect
Co/Pt multilayers
Co/$HfO_2$ interface
Interface chemical states

ABSTRACT

The enhancement of anomalous Hall effect (AHE) has been observed by inserting a Hf metallic layer at the Co/$HfO_2$ interface in perpendicular $HfO_2$/[Co/Pt]$_2$/Co/$HfO_2$ multilayers. It is displayed that the saturation anomalous Hall resistivity is 46% larger than that in Co/Pt multilayers without Hf insertion. Meanwhile, thermally stable AHE property is obtained in perpendicular $HfO_2$/[Co/Pt]$_2$/Co/Hf/$HfO_2$ multilayers. The X-ray photoelectron spectroscopy analysis reveals that the improved AHE originates from the modulation of chemical states at the Co/$HfO_2$ interface, owing to the insertion of the Hf layer.



## 1. Introduction

The anomalous Hall effect (AHE) in ferromagnetic materials, which is mainly utilized to probe the magnetization orientation and closely connected with the spin Hall effect, has recently received tremendous attention due to fascinating physics and potential applications [1–5]. However, the systematic understanding of the AHE in ferromagnetic materials has still not been accomplished and its physical mechanisms remain controversial. It is now extensively accepted that the AHE based on spin-orbit coupling involves intrinsic and extrinsic mechanisms. The intrinsic mechanism is related to the Berry phase curvature of the band structure [6–8]. In contrast, the extrinsic mechanism, including skew scattering and side jump [9–11], is purely scattering effect. The AHE-based ferromagnetic materials with perpendicular magnetic anisotropy (PMA) hold promise for magnetic memories and logic devices [4,12–17]. From the application point of view, obtaining large AHE in perpendicularly magnetized ferromagnets is one of the most significant goals. To date, Co/Pt multilayers with the PMA are promising candidates for AHE-based devices, while it remains still a great challenge for them to demonstrate large AHE with good thermal stability. Thus, it is timely to investigate the conditions to make PMA-based Co/Pt multilayers show the enhanced and thermally stable AHE to meet the application requirements for AHE-based devices. To make perpendicular Co/Pt multilayers display the improved and thermally stable AHE, extensive efforts have been devoted by many researchers. For example, the improvement of the AHE has been obtained in perpendicularly magnetized [Pt/Co]$_5$/Ru/[Co/Pt]$_5$ multilayers [18], which is attributed to the strong Ru/Co interface scattering. The enhanced AHE has been realized in Co/Pt multilayers using the metal-oxide interfaces [19], while the post-annealing treatment can weaken the AHE because of Co-Pt interdiffusion [20,21]. Furthermore, the thermally stable AHE has been gained by introducing the ferromagnet/$HfO_2$ interface in Co/Pt multilayers [22,23], while the oxidation of the Co layer at the Co/$HfO_2$ interface can deteriorate the AHE. In this paper, perpendicularly magnetized $HfO_2$/[Co/Pt]$_2$/Co/Hf($t_{Hf}$)/$HfO_2$ multilayers were designed and fabricated to investigate the influence of Hf insertion on the AHE in Co/Pt multilayers. Our experimental results present that the AHE in perpendicular $HfO_2$/[Co/Pt]$_2$/Co/$HfO_2$ multilayers has been enhanced, mainly ascribed to the modulation of the interface chemical states controlled by inserting a Hf metal layer at the Co/$HfO_2$ interface. Meanwhile, the good thermal stability of the AHE has been obtained in perpendicular Co/Pt multilayers regardless of Co-Pt interdiffusion.

* Corresponding authors.
*E-mail addresses:* iwooden@163.com (Q. Liu), jiangsl-cnc@pku.edu.cn (S. Jiang).
[1] These authors contributed equally to this work.

https://doi.org/10.1016/j.apsusc.2017.10.098
0169-4332/

## 2. Experimental details

Samples with the structure of $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/Hf(t_{Hf})/HfO_2(2)$ ($0 \le t_{Hf} \le 1$ nm) (in nm) were deposited on $SiO_2$/Si substrates using magnetron sputtering at room temperature with the base pressure lower than $3 \times 10^{-7}$ Torr and the working argon pressure of 2 mTorr. The Pt, Hf and Co layers were deposited using dc sputtering, while the $HfO_2$ layer was deposited using rf sputtering. The post-annealing treatment was performed at different temperatures for 30 min in vacuum below $3 \times 10^{-7}$ Torr without external magnetic field. Magnetization measurements of the samples were carried out using a physical property measurement system (PPMS). The films were then patterned into Hall bar structure using photolithography and Ar ion etching. Transverse Hall resistivity ($\rho_{xy}$) and longitudinal resistivity ($\rho_{xx}$) were studied by PPMS, with the external magnetic field ($H$) applied perpendicular to the film plane. Anomalous Hall resistivity ($\rho_{AH}$) was gained using linear extrapolation of the data at high fields to $H = 0$. The X-ray photoelectron spectroscopy (XPS, ESCALAB 250Xi) experiments were performed to analyze the composition of the films. The XPS detectable sample depth $d$ can be obtained using the formula $d = 3\lambda \sin \alpha$ [24], where $\lambda$ and $\alpha$ are inelastic mean-free paths (IMFPs) for photoelectrons and the takeoff angle for photoelectrons with respect to the surface plane of the samples, respectively. The IMFPs can be obtained from previous report [25]. For an Al $K_\alpha$ radiation source, the IMFPs for Hf 4f in metallic Hf and Co 2p in metallic Co are 2.93 nm and 1.18 nm, respectively. The IMFPs for Hf 4f and Co 2p in their oxides are approximately 0.1–0.2 nm larger than those in metallic Hf and Co, respectively. Thus, when $\alpha$ is 90°, the information at the Co/$HfO_2$ interface for sample $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/HfO_2(2)$ and at the Co/Hf interface for sample $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/Hf(0.6)/HfO_2(2)$ (in nm) can be obtained without Ar ion etching. The calibration of binding energy scale was performed to remove the charge effect with the C 1s line (284.6 eV).

## 3. Results and discussion

The plot of $\rho_{AH}$ as a function of $t_{Hf}$ for perpendicular $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/Hf(t_{Hf})/HfO_2(2)$ ($0 \le t_{Hf} \le 1$ nm) (in nm) multilayered films at 300 K, as seen in Fig. 1, shows that the $\rho_{AH}$ value increases monotonically with $t_{Hf}$ followed by a fall above 0.6 nm. The inset of Fig. 1 displays Hall loops for samples $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/HfO_2(2)$ and $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/Hf(0.6)/HfO_2(2)$ (in nm), and it can be observed that: the $\rho_{AH}$ value in Co/Pt multilayers without Hf insertion is 3.02 μW cm, but it increases to 4.42 μΩ cm after introducing a 0.6-nm-thick Hf metallic layer at the Co/$HfO_2$ interface, which is 46% larger than that in the film without Hf insertion.

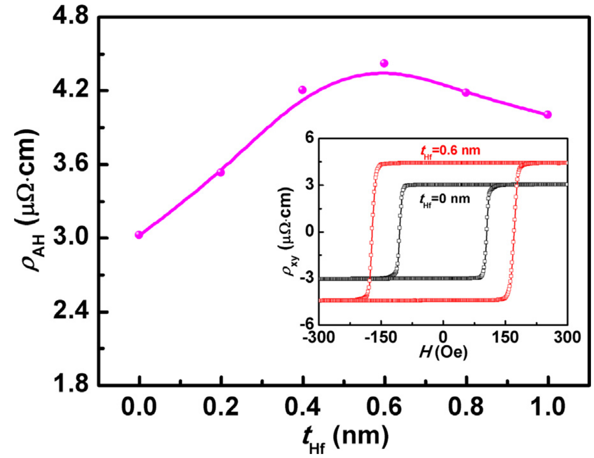


**Fig. 1.** Dependence of $\rho_{AH}$ on $t_{Hf}$ in perpendicular $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/Hf(t_{Hf})/HfO_2(2)$ ($0 \le t_{Hf} \le 1$ nm) (in nm) multilayers at 300 K. Inset: Hall loops for samples $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/HfO_2(2)$ (black square) and $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/Hf(0.6)/HfO_2(2)$ (red square) (in nm) at 300 K (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Furthermore, the $\rho_{xx}$ values for the two samples are 116.15 and 108.65 μΩ cm, respectively. As a result, the Hall angle ($\rho_{xy}/\rho_{xx}$), a key parameter for AHE-based device applications, reaches 4.1% in the film with the Hf insertion layer of 0.6 nm, and is about 58% larger than that in the sample without Hf insertion (only 2.6%).

Fig. 2 shows the magnetic hysteresis loops for samples $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/HfO_2(2)$ and $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/Hf(0.6)/HfO_2(2)$ (in nm) at 300 K. Obviously, it demonstrates that the value of the saturation magnetization ($M_S$) is about 700 emu/cm$^3$ for sample $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/HfO_2(2)$, while it increases to 960 emu/cm$^3$ for sample $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/Hf(0.6)/HfO_2(2)$ (in nm), which is 37% larger than that in Co/Pt multilayers without the Hf metallic layer.

The thermal stability is one of the key parameters for AHE-based device applications. Since the AHE in Co/Pt multilayers with the Co-$HfO_2$ interfaces shows good thermal stability [22,23], it is of great interests to investigate the variation of thermal stability of the AHE after inserting Hf at the Co/$HfO_2$ interface. Fig. 3 shows the dependence of $\rho_{AH}$ on annealing temperature ($T_a$) for perpendicular $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/Hf(0.6)/HfO_2(2)$ (in nm) multilayers. Evidently, the $\rho_{AH}$ value increases with increasing $T_a$ and then decreases, demonstrating the maximum value of 5.08 μΩ cm at $T_a$ = 250 °C, which is about 15% larger than that in the as-grown film (4.42 μΩ cm). Moreover, the $\rho_{AH}$ value is

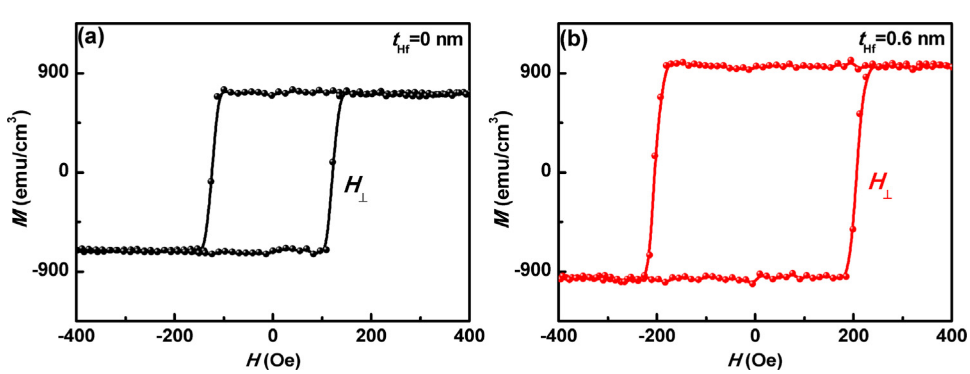


**Fig. 2.** The representative out-of-plane magnetic hysteresis loops for samples $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/HfO_2(2)$ (a) and $HfO_2(2)/[Co(0.5)/Pt(1)]_2/Co(0.5)/Hf(0.6)/HfO_2(2)$ (b) (in nm) at 300 K.