Anomalous Hall conductivity and switch of antiferromagnetic net magnetic moment in exchange-biased bilayers

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(Dated: March 11, 2014)

In this work, IrMn/Y₃Fe₅O₁₂ exchange bias bilayers are studied. The behavior of antiferromagnetic net magnetic moment ∆m_AF is directly probed by anomalous and planar Hall effects. Anomalous Hall conductivity is observed in this noncollinear antiferromagnet. The exchange bias (EB) and rotational hysteresis are demonstrated to be induced by the irreversible switching of ∆m_AF. In the training effects, the ∆m_AF changes continuously. This work highlights the fundamental role of ∆m_AF in the exchange bias and facilitate manipulation of AFM spintronic devices.

Exchange bias (EB) phenomena in ferromagnetic (FM)/antiferromagnetic (AFM) systems have attracted lots of attention because of its intriguing physics and technological importance in spin valve based magnetic devices [1–4]. In EB system, after the FM/AFM bilayers are cooled under an external magnetic field from high temperatures to temperatures below the Néel temperature of AFM layers, the hysteresis loops are simultaneously shifted and broadened [5, 6]. FM/AFM bilayers are now commonly integrated in spintronic devices [7]. Nevertheless manipulation and characterization of antiferromagnetic net magnetic moment are important to understand and control EB.

Indeed, rotatable and frozen AFM spins are thought to be responsible for the coercivity enhancement and shift of the FM hysteresis loops [8–12]. The FM and AFM spins are found to be correlated under an external magnetic field (H) [8]. Ohldag et al found that a nonzero AFM net magnetic moment ∆m_AF is necessary to establish the EB [13]. However, Wu et al thought that the EB can be established even without frozen AFM spins [9]. Therefore, the behavior of AFM spins is still under debate. Moreover, the EB training effect has been studied extensively and is attributed to the shrinking ∆m_AF during consecutive hysteresis loops [14–21]. For exchange-biased FM/AFM bilayers, the rotational hysteresis loss still sustains even at H larger than the saturation magnetic field and is correlated to the irreversible switching of AFM spins during clock wise (CW) and counter clock wise (CCW) rotations [22–25]. Since there is still lack of the direct experimental evidence, it is necessary to elucidate the fundamental mechanism of the AFM spins in the FM/AFM bilayers in experiments.

In most of studies, the information of AFM spins is indirectly explored from the hysteresis loops of the FM layers with micromagnetic simulations and Monte Carlo calculations. In sharp contrast, very few methods can be implemented to directly probe the AFM spins due to almost zero net magnetic moment of the AFM layers. However, different measurements, combining x-ray...
magnetic circular dichroism and x-ray magnetic linear dichroism can detect FM and AFM spins due to their element-specific advantage. Only very recently, tunneling anisotropic magnetoresistance (TAMR) effect, which was initially proposed for tunneling device consisting of a single FM electrode and a nonmagnetic electrode, was used to probe the $\Delta m_{AFM}$. Since the TAMR depends on the tunneling density of states and the orientation of the AFM magnetization in a complex way, direct characterization of the $\Delta m_{AFM}$ becomes impeded with conventional electric measurements. In this paper, we demonstrate effects of the $\Delta m_{AFM}$ on the EB, the training effect, and the asymmetry because $\rho_{xy}$ is two orders of magnitude smaller than $\rho_{xx}$.

FIG. 2: AHE loops at 20 K (a), 30 K (b), and 55 K (c) with $H$ along the film normal direction. (d) AHC as a function of temperature. Schematic spin configuration near positive and negative saturations, and points C and D, the longitudinal resistance versus $H$ at 30 K, and points C and D, the longitudinal resistance versus $H$ at 30 K, and Mn spin structure of A and B phases in IrMn in the insets of (a), (b), and (d), respectively.

IrMn (5 nm)/YIG (20 nm) bilayers are fabricated by pulse laser deposition (PLD) and subsequent magnetron sputtering in ultrahigh vacuum on (111)-oriented, single crystalline Gd$_3$Ga$_5$O$_{12}$ (GGG) substrates. The IrMn layer is deposited on YIG thin film at room temperature to avoid interfacial diffusion. The thicknesses of the YIG and IrMn layers are determined by x-ray reflection, as shown in Fig. (1a). The x-ray diffraction peaks are found at $2\theta = 54$ and 119 degrees corresponding to (444) and (888) orientations for GGG substrate and YIG film in Fig. (1b). Figures (1c) and (1d) show pole figures with $\Phi$ and $\Psi$ scan at $2\theta$ fixed for the (008) reflection of GGG substrate and YIG film. These results confirm the epitaxial growth of the YIG film. In-plane magnetization hysteresis loops of the YIG films were measured at room temperature using vibrating sample magnetometer (inset of Fig. (1a)). The measured magnetization (134 emu/cm$^3$) is close to the theoretical value of 131 emu/cm$^3$. The small coercivity clearly indicates high quality epitaxial YIG films.

Before measurements, the films are patterned into normal Hall bar and then cooled from room temperature to 5 K under $H = 30$ kOe along the film normal direction. The Hall resistivity $\rho_{xy}$ is measured as a function of the out-of-plane magnetic field for various temperatures, and then the anomalous Hall resistivity $\rho_{AH}$ is extrapolated from the linear dependence of $\rho_{xy}$ at large $H$. Figure (2d) shows the anomalous Hall conductivity $\sigma_{AH}$ decreases with increasing $T$ and vanishes near 55 K, where $\sigma_{AH}=\rho_{AH}/(\rho_{xx}^2+\rho_{AH}\cdot\rho_{xx})=\rho_{AH}/\rho_{xx}^2$ because $\rho_{AH}$ is two orders of magnitude smaller than the $\rho_{xx}$. In particularly, the Hall loop is asymmetric at 20 K, and the descent and ascent branches exhibit broad and sharp peaks, respectively. The asymmetry can also be seen from the $R_{xx}$ versus $H$ curve in the inset of Fig. (2b). The asymmetry becomes weak at high temperatures in Fig. (2c). The asymmetric reversal of the AFM spins explains the asymmetric hysteresis loops of the FM magnetization. It is interesting that the AHC, the exchange field, and the asymmetry all vanish near the same temperature of 55 K. The similar temperature dependence indicates that the AHC is strongly related to the perpendicular EB establishment and therefore exclusively caused by the $\Delta m_{AFM}$. All other physical reasons such as so-called spin Hall magnetoresistance can be excluded in the explanations of the AHC. AFM and FM spins are both aligned upwards and downwards along the film normal direction at positive and negative saturation fields, and are tilted with the film plane near points C and D, due to the combined effects of the demagnetization and the pinning effect from the AFM layer, as shown in the insets of Fig. (2a). To our knowledge, the AHC has for the first time been observed in the AFM IrMn with noncollinear spin structure on the kagome lattice although it has
FIG. 3: PHE loops of cycles $n = 1$, 2, and 7 at 5 K (a). Schematic pictures of the sensing current $i$ and cooling field $H_{FC}$ (b), and direction of $\Delta m_{AFM}$ vector at stages A(c), B(d), C(e), D(f), and E(g) of the $n = 1$ hysteresis loop in (a). The inset in (a) shows typical Hysteresis loops at low temperatures measured by magnetometry.

Figures 4(a)- 4(c) show the PHE loops at different temperatures. The PHE loop is shifted from zero field at low temperatures and centered $H = 0$ at 60 K. Meanwhile, the AFM spins at 5 K are far from saturation within the magnetic field of -200 Oe and easily saturated within -100 Oe at high temperatures. The observed results agree with the general trend of the exchange field and coercivity with temperature \cite{1}. The blocking temperature $T_B$ is about 60 K, where the exchange field approaches zero. Moreover, the magnitude change of the PHE decreases sharply with temperatures. Therefore, the temperature dependence of the PHE magnitude is strongly related to the EB. PHE loops were also measured at different $\theta_H$, as shown in Figs. 4(d)- 4(f). The angular dependent PHE loop reproduces the main feature of the angular dependence of the EB that the exchange field and the coercivity decrease when the $H$ deviates from the cooling field direction \cite{41}.

FIG. 4: PHE loops along the cooling field at 5 K (a), 20 K (b), 60 K (c), and at $\theta_H = 0$(d), 45(e), and 90(f) (degrees) at 5 K.

already been theoretically predicted in the IrMn \cite{59}.

After the sample is cooled from room temperature to 5 K under the cooling field parallel to the sensing current $i$(in Fig. 3b)), the transverse resistivity is measured with the in-plane field $H$ along the cooling magnetic field during measurements of the PHE loops. The PHE signal is proportional to the product of $\Delta m_{AFM,x}$ and $\Delta m_{AFM,y}$, which are components of $\Delta m_{AFM}$ parallel and perpendicular to both the current $i$ and the field $H$. Figure 3(a) shows the PHE loop with consecutive cycles. At point A, the $m_{AFM,x}$ is expected to be equal to the saturated value of the $\Delta m_{AFM}$ after the field cooling procedure. When the $H$ sweeps from points B to C, the $\Delta m_{AFM}$ is sharply switched irreversibly, followed by slow and irreversible rotations from C to D. Here, the AFM layer is far from saturation at $H = -600$ Oe. Figures 3(c)- 3(f) schematically show the angular rotation of the $\Delta m_{AFM}$ vector at typical stages in the descent branch of the first reversal. During the second reversal from points D to E, the $\Delta m_{AFM}$ is rotated in CCW sense. The irreversible switching and rotation of the $\Delta m_{AFM}$ are clearly demonstrated and explain the physical reason for the coercivity enhancement in exchange-biased FM/AFM bilayers \cite{23}.

For the cycle number $n = 1$, 2, and 7, the descent branch shifts significantly whereas the ascent branch almost does not change. The coercive field of descent branch is equal to -119, -88, and -82 (Oe). In particular, the PHE loop of $n = 1$ is still open and the PHE signal at the starting point A is larger than that of the ending point E. As schematically shown in Figs. 3(c) and 3(g), the $\Delta m_{AFM}$ vector has different orientations. As Hoffmann pointed out, the athermal training effect from $n = 1$ to $n = 2$ is due to the switching of AFM spins among easy axes \cite{14}. The reduced PHE signal at saturation corresponds to the decrease of the exchange field, which confirms the theoretical predictions \cite{15, 17–19}. With weak training effect between $n = 2$ and 7, the PHE signal at $H = 300$ Oe does not change much. Apparently, the $\Delta m_{AFM}$ vector experiences different trajectories during continuous reversals of the FM magnetization, which explains the physics behind the measured magnetization hysteresis loops of the YIG layer in the inset of Fig. 3(a) and previously reported results \cite{14, 17, 20, 21, 25, 40}.

Figures 3(a)- 3(c) show the PHE loops at different temperatures. The PHE loop is shifted from zero field at low temperatures and centered $H = 0$ at 60 K. Meanwhile, the AFM spins at 5 K are far from saturation within the magnetic field of -200 Oe and easily saturated within -100 Oe at high temperatures. The observed results agree with the general trend of the exchange field and coercivity with temperature \cite{1}. The blocking temperature $T_B$ is about 60 K, where the exchange field approaches zero. Moreover, the magnitude change of the PHE decreases sharply with temperatures. Therefore, the temperature dependence of the PHE magnitude is strongly related to the EB. PHE loops were also measured at different $\theta_H$, as shown in Figs. 4(d)- 4(f). The angular dependent PHE loop reproduces the main feature of the angular dependence of the EB that the exchange field and the coercivity decrease when the $H$ deviates from the cooling field direction \cite{41}.

Figures 3(a)- 3(d) show the PHE signal as a function
of \( \theta_H \) with CW and CCW under different magnitudes of \( H \). At \( H = 50 \) Oe, the CW and CCW curves overlap and the FM and AFM magnetic moments are expected to rotate reversibly within a small angular region. For higher \( H \), the hysteretic behavior begins to occur and becomes strong for \( H = 300 \) and \( 500 \) (Oe). This hysteretic behavior starts to decrease for \( H = 1.0 \) kOe but still persists at \( H = 20 \) kOe. Figures (e)-(h) show the angular dependence of the PHE signal with CW and CCW rotations under \( H = 1.0 \) kOe at different temperatures. It is interesting that the hysteretic effect becomes weak at enhanced temperatures. The CW and CCW curves almost overlap near 45 K, exhibiting reversible rotation of the \( \Delta m_{AFM} \). The hysteretic effect of the PHE curves behaves in a way similar to the rotational hysteresis loss of the FM layers [1, 24]. Since the measured results can be fitted well with \( \sin(2\theta_H) \), this means that \( \Delta m_{AFM} \) follows both the FM magnetization and \( H \). In contrast, the \( \Delta m_{AFM} \) rotates irreversibly during CW and CCW rotations at low temperatures. The irreversible switching of the \( \Delta m_{AFM} \) reveals the nature of the rotational hysteresis loss in most FM/AFM bilayers [1, 24]. The PHE magnitude change decreases significantly at enhanced temperatures, consistent with the AHC results in Fig. 2(d). The present angular dependence of the PHE provides a more direct approach to deduce the anisotropy constant in metallic AFM materials, compared to the TAMR because the latter is also strongly related to the interfacial effect at the AMF/insulating barrier layer [33].

Since the heavy Ir atom has large atomic spin-orbit coupling and each Mn atom has a magnetic moment of 2.91 \( \mu_B \), the IrMn alloys are expected to have large AHC due to broken mirror symmetry and spin-orbit coupling [39, 42, 43]. However, there exist two energetically equivalent phases in the inset of Fig. 2(d) and their AHC contributions are opposite to each other. When the IrMn layer experiences the field cooling procedure, the Mn spins make a very small angle with (111) plane, and IrMn layer experiences the field cooling procedure, the AHC due to broken mirror symmetry and spin-orbit coupling [39, 42, 43]. However, there exist two energetically equivalent phases in the inset of Fig. 2(d) and their AHC contributions are opposite to each other. When the IrMn layer experiences the field cooling procedure, the Mn spins make a very small angle with (111) plane, and the PHE are reduced and finally vanish near \( T_B \), as observed in the high-temperature data. Moreover, since more AFM grains become "suparamagnetism" at higher temperatures, thus AHC and the PHE are reduced and finally vanish near \( T_B \), as observed in Figs. 2 and 4. In conclusion, we clearly demonstrate that EB is correlated to the nonzero AFM net magnetic moment. Moreover, the Meiklejohn-Bean model is not suitable for the present results [4, 5]. In this model, AFM spins are fixed during the reversal of the FM magnetization, which is in contradiction with the present system. AHE and PHE measurements allow to probe the entire IrMn layer and not only the interface as in the reported TAMR measurements [31, 32]. The measured results in the present work better reflect the entire behavior of the IrMn layer.

In this work, the AFM net magnetic moment is
directly characterized in IrMn/YIG bilayers by AHE and PHE. The AHC has for the first time been observed in this metallic AFM materials. $\Delta m_{AFM}$ is proved to be necessary to observe EB. The irreversible switching of the AFM spins is directly observed and address the physical source for the coercivity enhancement and the rotational hysteresis loss. The direction of the $\Delta m_{AFM}$ is found to continuously change during the EB training effect. The present work permits a better understanding of EB and related phenomena in FM/AFM bilayers. It demonstrate that anomolous Hall measurement allows to probe the behavior of the AFM layer and consequently is powerful tool to understand FM/AFM system. This technique should be useful in the field of spintronics.

This work was supported by the National Science Foundation of China Grant Nos.51171129 and 51201114, and the State Key Project of Fundamental Research Grant No.2002CB613504, Shanghai Nanotechnology Program Center (No. 0252nm04).

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