AIP Advances

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Cite as: AIP Advances **10**, 015108 (2020); https://doi.org/10.1063/1.5129564
Submitted: 30 September 2019 . Accepted: 25 November 2019 . Published Online: 06 January 2020

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Presented: 7 November 2019 • Submitted: 30 September 2019 •
Accepted: 25 November 2019 • Published Online: 6 January 2020

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Note: This paper was presented at the 64th Annual Conference on Magnetism and Magnetic Materials.
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ABSTRACT

We investigated interlayer exchange coupling (IEC) and interface magnetic anisotropy (K_i) between two ferromagnetic layers with crossed in-plane and perpendicular magnetic anisotropies separated by a non-magnetic spacer by using the anomalous Hall effect (AHE). The sample consisted of a $Co_{0.75}Fe_{2.25}O_4$ layer with perpendicular magnetic anisotropy and a Fe layer with in-plane anisotropy, separated by a MgO layer with variable thickness. Since $Co_{0.75}Fe_{2.25}O_4$ and MgO are insulators, the AHE signal only reflects the magnetization process of Fe. From this, we determined both IEC and K_i. A strong antiferromagnetic IEC was confirmed between $Co_{0.75}Fe_{2.25}O_4$ and Fe. The strongest IEC of -1.1 mJ/m^2 was observed for directly coupled Fe and $Co_{0.75}Fe_{2.25}O_4$ for which K_i was -1.1 mJ/m^2.

I. INTRODUCTION

Interlayer exchange coupling (IEC) between two ferromagnetic (FM) layers separated by a non-magnetic interlayer has been intensively studied both theoretically[1–5] and experimentally.[6–14] In fact, it is an important phenomenon that is exploited in modern spintronics.[15–17] The origin of IEC lies in the difference of spin-polarized reflection at the FM interface, as found by modelling the system as a quantum well.[4] If the insertion layer is metallic, the IEC oscillates between the ferromagnetic (FM) and the antiferromagnetic (AFM) coupling as a function of the interlayer thickness.[6–8] On the other hand, if the insertion layer is insulating, the electron Fermi wavenumber (k_F) is imaginary, and the IEC simply decays with the interlayer thickness.[9,11–13] Trilayer systems comprising FM oxide layers such as Fe/MgO/Fe_3O_4,[13,18] Fe/MgO/γ-Fe_2O_3,[13] Fe_3O_4/TiN/Fe_3O_4,[19] and $La_{2/3}Ba_{1/3}MnO_3$/$LaNiO_3$/$La_{2/3}Ba_{1/3}MnO_3$,[20] also exhibit IEC.

In most cases, IEC has been investigated in a system with individual FM layers having collinear magnetic easy axes, either perpendicular or in-plane. However, few reports are available for systems where the FM materials have non-collinear or orthogonally crossed magnetic easy axes,[21,22] and more experiments are needed to gain a deeper understanding of the IEC. Recently, Fallarino *et al.*[22] reported the observation of IEC between a CoFeB layer with in-plane magnetic anisotropy (IMA) and a Co/Ni one with perpendicular magnetic anisotropy (PMA) by using ferromagnetic resonance (FMR). The FM-AFM oscillation behavior was also observed.

In this study, we investigated both IEC and K_i in a trilayer system composed of two FM layers having orthogonal magnetic easy axes. The structure of the trilayer system is a MgO substrate/$Co_{0.75}Fe_{2.25}O_4$/MgO/Fe. An epitaxial $Co_{0.75}Fe_{2.25}O_4$ (CFO) film grown on MgO(001) exhibits large coercivity and PMA due to t_{2g}-level splitting caused by a local lattice symmetry enhancing the spin-orbit interaction.[23–26] While CFO and MgO are good insulators, Fe is a metal; thus, only the changes in magnetization of the latter can be detected by the anomalous Hall effect (AHE).[27]

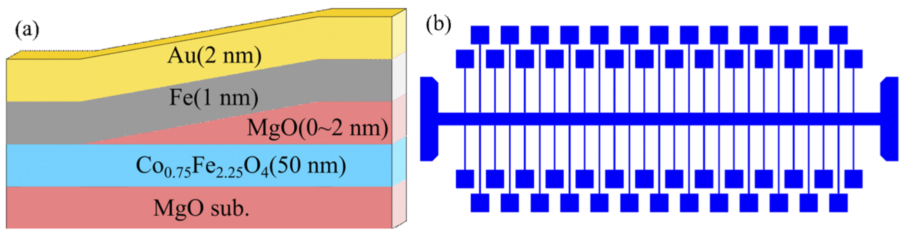

FIG. 1. (a) Sample structure. (b) Scheme of the Hall bar used to measure the IEC thickness dependence.

II. EXPERIMENTAL METHODS

All samples were grown by reactive radio-frequency magnetron sputtering (ES-250MB by Eiko Engineering Co., Ltd.).[28] The final film structure and layer thickness (in parentheses) are MgO(001) substrate/$Co_{0.75}Fe_{2.25}O_4$ (50 nm)/MgO (0-2 nm)/Fe (1 nm)/Au (2 nm), as shown in Fig. 1. Prior to film growth, the MgO(001) substrate was sequentially cleaned by ultrasonic treatment in acetone, ethanol, and de-ionized water for 5 min each. For the fabrication of $Co_{0.75}Fe_{2.25}O_4$ (CFO), we used 2-inch alloy target with the desired composition of Co: Fe = 1:3. The film was grown at a temperature of 500°C with an O_2/Ar flow ratio of approximately 0.71. After depositing CFO, a wedge-shaped MgO interlayer with continuously increasing thickness was grown at 150°C by using a moving mask and a ceramic target. Finally, the Fe and Au layers were deposited at room temperature. The multilayer structure was characterized by reflection high-energy electron diffraction (RHEED), X-ray reflectivity (XRR, by Rigaku Smart Lab, using Co $K\alpha$ radiation) and X-ray diffraction (XRD). Perpendicular magnetic hysteresis (MH) loops were measured by using a vibrating sample magnetometer (VSM), which is part of a physical property measurement system (PPMS, by Quantum Design). For Hall measurements, the films were patterned into Hall bars by photolithography (200 μm width × 8000 μm length, applying 1 mA to the Hall bar) and Ar ion milling. Then, Cr and Au electrodes were sputtered on top. The current is designed to flow parallel to the MgO thickness gradient, and the Hall voltage perpendicular (vertical) to it. The final Hall bar pattern is shown in Fig. 1. The AHE was measured by PPMS using a Keithley 6221 DC current source and a Keithley 2182 nanovoltmeter. Both VSM and AHE experiments were carried out at room temperature with an external magnetic field of up to 7 T along the MgO[001] direction.

III. RESULTS AND DISCUSSION

Figure 2 shows the MH loop of a single-layer CFO film grown on the MgO(001) substrate. The saturation magnetization (M_S) of the CFO films was approximately 330 kA/m, a value slightly smaller than its bulk counterpart (≈ 430 kA/m). This suggests the existence of a magnetic dead layer at the interface between MgO(001) and the CFO film, which can be related to the high density anti-phase boundaries in a spinel structure.[26,29] The film squareness ratio (SR), coercivity ($\mu_0 H_c$) and saturation field ($\mu_0 H_S$) are 0.85, 0.91 T and 1.5 T, respectively. Thus, because of the large $\mu_0 H_c$ and the high SR, CFO can be considered a pinned layer.

In order to evaluate size effect in M_S of the Fe layer, we fabricated a MgO sub./Fe (1 nm)/Au multilayer by depositing the metals at room temperature and measured the magnetization by VSM at 300 K.[30] The M_S at 300 K is 1360 kA/m, approximately 0.80 times that of bulk Fe.

Figure 3 shows the $\rho_{AHE}(H)$ loop of MgO sub./CFO/Fe/Au. In the magnetization loop perpendicular to the Fe layer plane, a clear hysteresis is opened even for the hard axis of the Fe layer. This means that Fe layer feels an exchange field (H_{ex}) originated from the IEC in addition to the external magnetic field (H_{ext}). Therefore, the effective magnetic field (H_{eff}) acting on the Fe layer can be described as $H_{eff} = H_{ext} + H_{ex}$. When the magnetic field goes to zero from positive high values, the remanent state $\rho_{AHE}(0)$ is negative, meaning that $H_{ex} < 0$ and that antiferromagnetic coupling ($J < 0$) exists between the Fe and CFO layers. In other words, IEC can make the preferential axis of the Fe layer perpendicular to the plane if the Fe layer is sufficiently thin, since the exchange field at the interface acts along the out of plane direction.

The IEC energy (J) and the interface magnetic anisotropy (K_i) can be determined from the equations[31–33]

$$J = \int_0^{M_{S,Fe}} \mu_0 H_{ex} t_{Fe} dM = \mu_0 H_{ex} t_{Fe} M_{S,Fe}, \tag{1}$$

$$K_i = -\int_0^{M_{S,Fe}} \mu_0 (H(M) + H_{ex}) t_{Fe} dM + \frac{1}{2}\mu_0 M_{S,Fe}^2 t_{Fe}, \tag{2}$$

where t_{Fe} and $M_{S,Fe}$ are the Fe layer thickness and saturation magnetization, respectively. Since a single Fe layer has no coercivity for

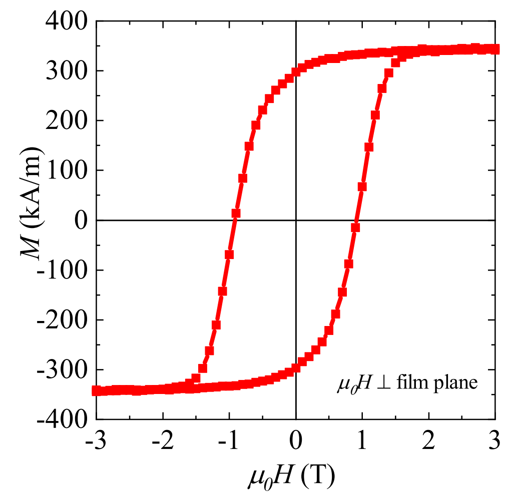

FIG. 2. Magnetic hysteresis loop of a single layer CFO film grown on the MgO(001) substrate at RT.

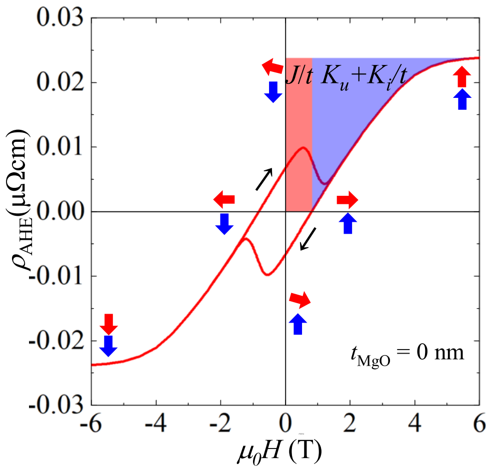

FIG. 3. $\rho_{AHE}(H)$ loop at t_{MgO} = 0 nm, where ρ_{AHE} is the Hall resistivity without the ordinary Hall component. Blue and red arrows indicate the magnetization direction of CFO and Fe, respectively. Black arrows indicate the sweep directions of the loop. The red-shaded and blue-shaded area correspond to the IEC energy (J/t) and the total magnetic anisotropy energy ($K_u + K_i/t_{Fe}$), respectively.

out-of-plane magnetization, $-\mu_0 H_{ex}$ is equal to the x-axis intersect of $\rho_{AHE}(H)$. Therefore, J corresponds to the red-shaded area of Fig. 3. Here, we assumed that the Fe layer has only the shape magnetic anisotropy as the bulk component of the magnetic anisotropy, $K_u = -\frac{1}{2}\mu_0 M_{S,Fe}^2$. Thus, K_i is determined by the difference in anisotropy energy and corresponds to the blue-shaded area of Fig. 3. Note that if FM coupling exists, $\mu_0 H_{ex}$ is positive. Note also that Eq.(1) is valid for both AFM and FM coupling.

The dependence of J and K_i (Eqs. (1) and (2)) on the thickness t_{MgO} of the MgO interlayer between the CFO and Fe layers is shown in Fig. 4 and Fig. 5, respectively. We first consider IEC. A positive J value corresponds to FM coupling, while a negative one corresponds to AFM coupling. In our system, two types of IEC can

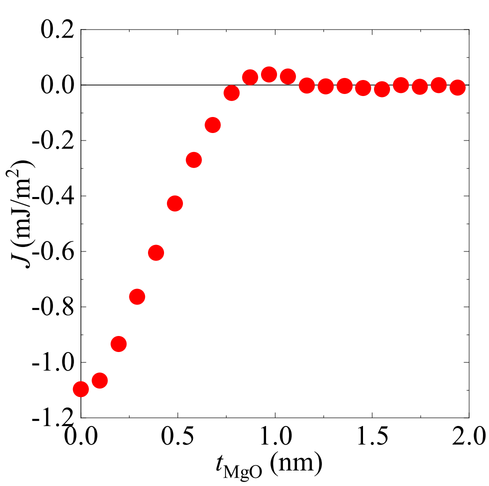

FIG. 4. Dependence of the IEC energy J on the MgO film thickness t_{MgO}.

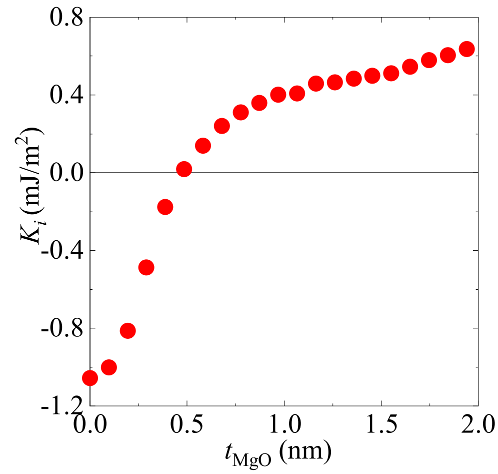

FIG. 5. Dependence of the interface magnetic anisotropy K_i on the MgO film thickness t_{MgO}.

exist: (i) direct coupling between CFO and Fe, and (ii) indirect coupling between CFO and Fe through MgO. From Fig. 4, it is seen that the coupling is AFM for almost all t_{MgO}. Furthermore, J increases linearly with t_{MgO} from its lowest value of -1.1 mJ/m^2. This indicates that both direct and indirect coupling are AFM. In the quantum well model of Ref. 4, J changes monotonically for an insulating interlayer, which is at least qualitatively consistent with our results. However, around t_{MgO} = 1 nm, we observed a weak FM coupling. A similar phenomenon has been reported by Katayama *et al.*[11] for Fe/MgO/Fe. The authors attributed this behavior to the oxygen vacancies in the MgO interlayer. Another possible origin of the weak FM coupling observed is the orange peel effect.[34] In addition, J changes almost linearly with t_{MgO}. These results indicate that the growth mode of the MgO interlayer is island growth like.

Next, we consider the interface magnetic anisotropy K_i determined by Eq. (2). As seen from Fig. 5, K_i is initially negative and changes sign as the MgO film thickness increases. This means that although the interface anisotropy between CFO and Fe is negative, it is positive between MgO and Fe. A large interface PMA has been previously reported between a Fe and MgO layer,[35] consistent with our results. In general, determining K_i between two FM layers is not straightforward, because it is difficult to measure the magnetic hysteresis of only one FM layer. However, in our system one FM layer is an insulator characterized by PMA, and the other one is a conductor characterized by IMA. The magnetic anisotropy energy of the metal layer corresponds to the area of the out-of-plane MH loop. Thus, we can determine K_i from the area difference between the two, finding that it is negative at the CFO-Fe interface, with a value of -1.1 mJ/m^2.

IV. CONCLUSIONS

In summary, we investigated the interlayer exchange coupling and the interface magnetic anisotropy in $Co_{0.75}Fe_{2.25}O_4$/MgO/Fe(001) by measuring the anomalous Hall effect. Since we can measure the MH loop only in the Fe layer by AHE, J and K_i are determined from the exchange field and the evolution of the MH loop of the Fe layer, respectively. By studying the IEC dependence on the interlayer MgO thickness, a strong AFM coupling was observed between $Co_{0.75}Fe_{2.25}O_4$ and Fe. The strongest coupling was found in the absence of the MgO interlayer, with a value of −1.1 mJ/m^2. Finally, we measured a K_i of −1.1 mJ/m^2 between the $Co_{0.75}Fe_{2.25}O_4$ and the Fe layer, being one of the few magnetic anisotropy measurements reported between two ferromagnetic layers. In the system with crossed IMA and PMA, strong exchange coupling at the interface can lead to PMA for IMA film. However, in our system, exchange coupling is not enough for the Fe layer to exhibit PMA.

ACKNOWLEDGMENTS

H.K. acknowledges the Kato foundation for promotion of science (KS-3123). This project is partly supported by Japan Science and Technology Agency (JST) under Collaborative Research Based on Industrial Demand "High Performance Magnets: Towards Innovative Development of Next Generation magnets" (grant number JPMJSK1415). H.K. thanks S. Sharmin for her help.

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