Enhancement of spin Hall magnetoresistance effect in CoFe$_2$O$_4$/Pt/CoFe$_2$O$_4$ trilayers
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The spin Hall magnetoresistance (SMR) phenomenon includes the fundamental physics of spin current, and originates from spin accumulation at an interface owing to the spin Hall effect. Although bilayers are the simplest structure exhibiting SMR, these exploit spin accumulation at only one side of a layer. Herein, trilayers of CoFe$_2$O$_4$/Pt/CoFe$_2$O$_4$ were fabricated and their spin Hall magnetoresistance was investigated. The trilayer structure featuring a thin Pt layer exhibited an SMR ratio four times that of a CoFe$_2$O$_4$/Pt bilayer. Further, the SMR ratio exhibited a dependence on Pt layer thickness that can be attributed to interference of the spin accumulations at both sides. Herein, several parameters such as spin diffusion length and mixing conductance were derived using the theory of Chen et al.

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Spin current has attracted increased attention because of novel phenomena such as the spin Seebeck effect,\textsuperscript{1} spin pumping,\textsuperscript{2,3} and spin current transport in magnetic insulators.\textsuperscript{4} In particular, spin current generation associated with the spin-orbit interaction such as the spin Hall effect\textsuperscript{5–7} or the Rashba-Edelstein effect\textsuperscript{8} is a crucial technology in spintronics research. To understand spin current, estimations of phenomenological parameters such as spin diffusion length are necessary. Spin pumping and the inverse spin hall effect is a powerful tool to investigate the generation and transport of the spin current, but high-frequency equipment is necessary to conduct these experiments.\textsuperscript{2,3} In 2013, spin Hall magnetoresistance (SMR) was discovered, which is associated with the spin Hall effect and with spin transfer at the interface of a ferromagnet insulator (FMI) and a nonmagnetic heavy metal (HM).\textsuperscript{9,10} Nakayama et al. observed a small magnetoresistance effect in yttrium iron garnet (YIG)/Pt bilayers, whereupon Chen et al. established a theory for SMR based on the spin Hall effect in the Pt layer and spin transfer at the YIG/Pt interface.\textsuperscript{11} Though SMR is a very small effect, many researchers have focused on this phenomenon because it includes fundamental physics of spin current.\textsuperscript{10,12–16}

Because SMR enables estimation of the parameters of spin current using dc-electric measurements, many studies have been conducted regarding the dependence of SMR on crystal orientation,\textsuperscript{17} material,\textsuperscript{18} temperature,\textsuperscript{19,20} and substrate.\textsuperscript{21} These experiments commonly employed bilayers comprising a magnetic insulator and a heavy metal, and sometimes all-metallic bilayers were used.\textsuperscript{22} In a bilayer, one side of the Pt layer interfaces with the FMI and the other Pt side is the surface (or interfaces with a non-magnetic capping insulator), and thus the SMR was observed to originate at one interface (Fig. 1(a)). Because the spin current in Pt accumulates in both directions, we can expect an additional effect in FMI/Pt/FMI trilayers (Fig. 1(b)). In particular, a very thin Pt layer induces an interference of spin accumulated regions at both sides (Fig. 1(c)). Chen et al. have theoretically discussed the SMR of trilayers, but few experimental studies\textsuperscript{23} have been conducted so far.

In this study, we fabricated CoFe\textsubscript{2}O\textsubscript{4}/Pt/CoFe\textsubscript{2}O\textsubscript{4} trilayers and performed magnetotransport measurements for various Pt thicknesses ($t_{\text{Pt}}$). The Pt thickness-dependence of the SMR in a trilayer was compared to that in a bilayer, where the trilayer with very thin Pt layer exhibited an SMR ratio four times greater than that in bilayers.

FIG. 1. Schematic of the spin current in a non-metallic layer adjacent to a magnetic insulator for a (a) bilayer and (b) trilayer. (c) Spin accumulation regions at the interface with a non-metallic layer. For thinner layers (lower) the accumulation region at the opposite side induces interference.

Though YIG is commonly used in conventional SMR measurements, its fabrication by physical vapor deposition is difficult except on a gadolinium gallium garnet substrate.\textsuperscript{24} Therefore, we employed CoFe\textsubscript{2}O\textsubscript{4} herein as an FMI instead of YIG.\textsuperscript{10} The CoFe\textsubscript{2}O\textsubscript{4} film was grown epitaxially on MgO or Pt\textsuperscript{25,26}, so that it is suitable for an FMI in a trilayer. The samples were grown in a molecular beam epitaxy chamber with a base pressure of $\sim 1 \times 10^{-7}$ Pa, where the film structures were

(1) MgO(001)/NiO (5 nm)/CoFe\textsubscript{2}O\textsubscript{4} (50 nm)/Pt ($t_{\text{Pt}}$)/ CoFe\textsubscript{2}O\textsubscript{4} (50 nm);
(2) MgO(001)/NiO (5 nm)/CoFe\textsubscript{2}O\textsubscript{4} (50 nm)/Pt ($t_{\text{Pt}}$);
(3) MgO(001)/Pt ($t_{\text{Pt}}$)/ CoFe\textsubscript{2}O\textsubscript{4} (50 nm).

Hereafter, samples (1), (2), and (3) will be referred to as CFO/Pt/CFO, CFO/Pt and Pt/CFO, respectively.
The MgO(001) substrates were annealed at 600 °C for 30 min and smoothed by a subsequent deposition of 20 nm MgO buffer layer grown at 400 °C at a rate of 0.3 Å·s⁻¹. An NiO layer to prevent diffusion of Mg into the CoFe₂O₄ films was then deposited at 300 °C at a rate of 0.15 Å·s⁻¹ in an oxygen pressure of 4×10⁻⁴ Pa. The CoFe₂O₄ films were deposited at 0.25 Å·s⁻¹ in an oxygen pressure of 4×10⁻⁴ Pa by co-depositing elemental Co and Fe at 300 °C. Subsequently, the CoFe₂O₄ films were annealed at 450 °C for 30 min. The Pt layer was deposited at 100 °C at a rate of 0.2 Å·s⁻¹ and was then annealed at 300 °C for 30 min. The Pt film deposition rate and substrate deposition temperature was rather low to suppress island growth and to ensure good surface coverage.²⁷

The growth and the surface morphology of the samples were confirmed by reflection high-energy electron diffraction (RHEED), while the crystal structures were investigated by X-ray diffraction (XRD). The magnetization process was measured by vibrating sample magnetometry (VSM) at room temperature (RT). Angle-dependent magnetoresistance (ADMR) measurements were performed in a cryostat, with a fixed applied field, H, of 1 T (i.e., maximum field of our magnet) and the angle was varied along the H-rotation planes whose corresponding angles were defined as α (from x, angle α = 0, towards γ), β (from z, angle β = 0, towards γ), and γ (from z, angle γ = 0, towards x), as shown in Fig. 4(b). The x-, y-, and z-directions were respectively along the current direction, in-plane and transverse to x, and out-of-plane.

The crystallinity of the films and interface quality will inevitably affect the spin current transport. Therefore, to confirm the growth mode, in situ RHEED patterns were acquired during deposition (not shown). The RHEED pattern of the CFO/Pt bilayer exhibited very clear streak pattern, while the Pt/CFO bilayer films exhibited a rather spotty pattern. This suggests that the CFO/Pt possessed a flat surface and the Pt/CFO possessed a rather rough surface. Figure 2(a) provides the XRD 0-20 profiles in logarithmic scale for the CFO/Pt/CFO, CFO/Pt, and Pt/CFO samples. The CFO on the MgO(001) substrates grew in the (001) crystal orientation, while Pt grew in the (111) direction on both CFO(001) and MgO(001) surfaces.²⁸,²⁹ Finally, CFO(111) grew on the Pt(111). The growth direction results as obtained by XRD are summarized schematically in Fig. 2(b). The fringe attributed to Pt(111) were observed in all sample profiles in the range of 30° < 2θ < 40°. The fringe attributed to Pt grown on MgO was broadened, and therefore the interface at Pt/CFO is considered rougher than that at CFO/Pt, which is consistent with the RHEED observation.

![FIG. 2. (a) XRD profiles for MgO(001)/NiO (5 nm)/CoFe₂O₄ (50 nm)/Pt (4 nm)/CoFe₂O₄ (50 nm) [upper red spectra]; MgO(001)/NiO (5 nm)/CoFe₂O₄ (50 nm)/Pt (4 nm) [center blue spectra]; and MgO(001)/Pt (4 nm)/CoFe₂O₄ (50 nm) [lower green spectra]. (b) Schematic images of the crystal orientation for the films. All substrates are MgO(001).](image-url)
The magnetic characteristic of the system is a crucial factor for SMR. Therefore, we carried out VSM measurements for all of the samples, which are given in Fig. 3. A magnetic field of 1.5 T was applied in the [001] direction (i.e., out-of-plane) and [100] and [110] directions (i.e., in-plane). All of the samples exhibited perpendicular magnetic anisotropy, which has been previously reported for CoFe²O₄ grown on an MgO substrate. The CFO/Pt bilayer exhibited a larger perpendicular anisotropy than that of the Pt/CFO bilayer, and the hysteresis of the CFO/Pt/CFO trilayer was a combination of these two bilayers. The difference between the Pt/CFO and CFO/Pt was responsible for the crystal orientation and crystallinity of the films.

![Figure 3](image_url)

**FIG. 3.** Magnetic hysteresis curves for (a) MgO(001)/NiO (5 nm)/CoFe₂O₄ (50 nm)/Pt (tₚ)/CoFe₂O₄(50 nm); (b) MgO(001)/NiO (5 nm)/CoFe₂O₄ (50 nm)/Pt (tₚ); and (c) MgO(001)/Pt (tₚ)/CoFe₂O₄ (50 nm).

Figure 4 shows the ADMR of the three samples with tₚ = 2 nm at RT in 1 T. The magnetic fields were rotated in the x–y, y–z, and z–x planes, and termed α-, β-, and γ-scans, respectively (Fig. 4(b)). Because CFO and MgO are insulators, the current flowed only in the Pt layer. All of the samples exhibited clear ADMR, consistent with previous SMR experiments. In the γ-scan, we observed a small resistance change as a function of the angle, though it should be zero according to the theory of SMR. This nonzero resistance change may be associated with a proximity effect from the Pt that touches the ferrimagnetic CFO layers. The amplitude of ADMR was strongly dependent on the layer structures; where the trilayer showed the largest ADMR (0.078%), the CFO/Pt bilayer showed the next largest ADMR (0.017%) and the Pt/CFO showed the smallest ADMR (0.003%). The difference between the ADMR of the CFO/Pt and Pt/CFO bilayers was responsible for the quality of the interface between Pt and CFO, as observed in the XRD and RHEED measurements.

Figure 5(a) plots the ADMR of the CFO/Pt/CFO trilayer for varying thicknesses of Pt (tₚ=2–12 nm), and Fig. 5(b) plots the SMR ratio as a function of tₚ. All of the samples exhibited clear attributes of SMR, whose amplitude decreased with increasing tₚ. The SMR of the trilayer was greater than that of the bilayers for all values of tₚ. It should be noted that the SMR behavior for a very thin Pt layer (tₚ =2–4 nm) were qualitatively different between the trilayer and the bilayers.

We analyzed the tₚ dependence of the SMR based on the theory of Chen et al., and estimated the phenomenological parameters of spin current. The SMR equations derived by Chen et al. are given as

\[
\frac{\Delta \rho}{\rho} = \frac{\theta_{SH}^2}{d_N} \left( \frac{2\lambda^2 G_r \tanh^2 \frac{d_N}{2\lambda}}{\sigma + 2\lambda G_r \coth \frac{d_N}{\lambda}} \right),
\]

for a bilayer and
\[
\frac{\Delta \rho}{\rho} = \frac{\theta_{SH}^2}{d_N} \frac{4\lambda^2 G_r \tanh \frac{d_N}{2\lambda}}{\sigma + 2\lambda G_r \tanh \frac{d_N}{2\lambda}},
\]

for a trilayer, where \( \theta_{SH} \) is the spin Hall angle, \( \lambda \) is the spin diffusion length in Pt, \( G_r \) is the real part of the spin mixing conductance at the CFO/Pt interface, \( d_N \) is the thickness of the Pt, and \( \sigma \) is the conductance of Pt.

FIG. 4. Angle-dependent magnetoresistance of MgO(001)/NiO (5 nm)/CoFe\(_2\)O\(_4\) (50 nm)/Pt (2 nm)/CoFe\(_2\)O\(_4\) (50 nm) (upper left); MgO(001)/NiO (5 nm)/CoFe\(_2\)O\(_4\) (50 nm)/Pt (2 nm) (upper right); and MgO(001)/Pt (2 nm)/CoFe\(_2\)O\(_4\) (50 nm) (lower left). (b) Schematic showing the angles \( \alpha \) (upper), \( \beta \) (center) and \( \gamma \) (lower) between the electric current and the magnetic field for angle-dependent magnetoresistance.

The data in Fig. 5(b) were fitted using Eqs. (1) and (2), where the value for \( \theta_{SH} \) was 0.056, as derived in a previous work regarding spin pumping whose Pt conductance was the same as herein (4.3–5.7×10\(^6\) S m\(^{-1}\)), \(^{33}\) and where experimental values were used for \( \sigma \). The parameters used in the fitting plotted in Fig. 5(b) are summarized in Table I.

To calculate the data fits, the values of \( \theta_{SH} \) and \( \sigma \) were given and thus \( \lambda \) and \( G_r \) were free parameters. The \( G_r \) parameter had a strong relationship with the maximum value of the SMR ratio, while the \( \lambda \) parameter primarily determined the damping behavior of the SMR. Because the quality of the interface and the Pt layer were different for the CFO/Pt and Pt/CFO bilayers, it is likely that the values of \( G_r \) and \( \lambda \) are different for each bilayer.\(^{14}\) Particularly, the

FIG. 5. (a) Angle-dependent magnetoresistance (ADMR) of CFO/Pt/CFO for various Pt film thicknesses, obtained at room temperature and in a 1 T magnetic field. (b) ADMR ratio as a function of \( t_{\text{Pt}} \) for Pt/CFO, CFO/Pt and CFO/Pt/CFO, where the data are fitted using Eq. (1) for CFO/Pt/CFO and Eq. (2) for Pt/CFO and CFO/Pt.
Pt/CFO bilayer has a very small SMR ratio and its XRD peaks for CFO are rather small, suggesting that the film quality of the upper CFO and the upper interface of CFO/Pt/CFO were not sufficiently smooth. We note that the formula of Chen et al. for a trilayer was derived under the assumption of identical upper and bottom interfaces.

We succeeded in identifying a parameter set that could simultaneously reproduce the $t_{\text{Pt}}$ dependence of the SMR for the CFO/Pt bilayer and the CFO/Pt/CFO trilayer, as shown in Fig. 6, although there is no necessity to have same parameter values for the fitting curves. The parameters used in Fig. 6 are summarized in Table I. For this fitting, the $\theta_{\text{SH}}$ was estimated at 0.03, which is not far from $\theta_{\text{SH}} = 0.056$ employed for the fittings in Fig. 5(b). The $G_r$ parameter in Fig. 6 has rather larger value than that in Fig. 5(b). The inset in Fig. 6 shows the ratio between the SMR of CFO/Pt and CFO/Pt/CFO, where the ratio of the trilayer/bilayer SMR values was almost 2 for the range $4 < t_{\text{Pt}} < 12$ nm and increased significantly for decreasing $t_{\text{Pt}} \leq 2$ nm. This enhancement was attributed to the interference of the spin accumulation at both sides of the Pt layer (Fig. 1(c)).

![Graph showing SMR vs. Pt thickness](image)

**FIG. 6.** Spin Hall magnetoresistance (SMR) ratio of CFO/Pt/CFO and CFO/Pt and the theoretical calculation using the parameters in Table I. (Inset) Ratio of SMR of CFO/Pt and CFO/Pt/CFO, showing the experimental data (open circles) and the calculated curve (solid line).

**TABLE I.** Fitting parameters for the $t_{\text{Pt}}$-dependent SMR ratio calculation following the theory of Chen et al., giving values for the spin Hall angle ($\theta_{\text{SH}}$), spin diffusion length ($\lambda$), conductance of Pt ($\sigma$), and the real part of the spin mixing conductance at the interface ($G_r$).

| Material       | $\theta_{\text{SH}}$ | $\lambda$ (nm) | $\sigma \times 10^6$ S m$^{-1}$ | $G_r \times 10^{15}$ |
|----------------|-----------------------|-----------------|---------------------------------|----------------------|
| CFO/Pt/CFO     | 0.056                 | 0.783           | 5.67                            | 1.87                 |
| CFO/Pt         | 0.056                 | 1.31            | 4.78                            | 0.437                |
| Pt/CFO         | 0.056                 | 1.77            | 4.32                            | 0.067                |
| Fig. 6         | 0.03                  | 1.15            | 5.00                            | 10.0                 |

We fabricated bilayers and a trilayer consisting of CoFe$_2$O$_4$ and Pt, where all of the samples exhibited clear SMR and the trilayer exhibited a much larger SMR than the bilayer. For the trilayer, the SMR ratio monotonically increased with decreasing $t_{\text{Pt}}$, while the two bilayers had a maximum SMR value at $t_{\text{Pt}} = 4$ nm. These behaviors, as qualitatively explained by the theory of Chen et al., can be considered to originate from the interference of spin accumulation at both interfaces. Further, the results indicated a method to enhance the spin-orbit torque efficiency. Because the small SMR herein arises from the small $\theta_{\text{SH}}$ of about 0.05, the SMR can be improved by using a material with a large $\theta_{\text{SH}}$, such as $\beta$-W$^{34}$ and topological materials.$^{35}$ The combination of a thin Pt layer and a large $\theta_{\text{SH}}$ material could drastically enhance the SMR.
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