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Stacking order dependence of inverse spin Hall effect and anomalous Hall effect in spin pumping experiments

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I. INTRODUCTION

The injection of pure spin current $J_s$ into adjacent non-magnetic materials (NMs) has demonstrated strong potential for the development of applied spintronic devices. The $J_s$ injection processes are commonly elucidated using three general methods such as thermal spin injection, electrical spin injection, and ferromagnetic resonance (FMR). In particular, spin pumping, which is the generation of $J_s$ from FMR, in a NM/ferromagnet (FM) bilayer enables efficient spin injection into metals and semiconductors free from the impedance mismatch problem. The inverse spin Hall effect (ISHE) in the NM/FM bilayers is commonly used for detecting $J_s$ via the ISHE as

$$J_{ISHE} = \frac{2e}{h} J_s \times \sigma,$$

where $\theta_{ISH}$, $e$, $h$, and $\sigma$ denote the spin Hall angle, elementary charge, Dirac constant, and spin-polarization vector of $J_s$, respectively. Here, $\theta_{ISH}$ represents the efficiency of the spin-charge conversion in NM, which is measured for several metals and alloys. It is well known that $\theta_{ISH}$ can be obtained from the experimental ISHE voltage $V_{ISHE}$ using a phenomenological model of spin pumping.

Moreover, the strength of spin-orbit coupling (SOC) contributes to the amplitude of $V_{ISHE}$. Generally, the interaction between spin and orbital angular momentum generates the anomalous Hall effect (AHE), ISHE, and other spin-related phenomena in magnetic systems. For example, in the resonant cavity, the in-plane component of a microwave electric field $E_{MW}$ in the FM layer may induce an AHE current $J_{AHE}$ via the AHE in conjunction with FMR. Then, $J_{AHE}$ can be expressed as

$$J_{AHE} \propto E_{MW} \times m,$$

where $m$ is the $M$ component perpendicular to $E_{MW}$. As a result, $V_{ISHE}$ at the NM layer should overlap with the AHE voltage $V_{AHE}$ at the FM layer due to originating SOC in the NM/FM bilayer systems. Therefore, the values of $\theta_{ISH}$ estimated to $V_{ISHE}$ largely vary in accordance with the ratio of $V_{AHE}/(V_{ISHE} + V_{AHE})$, which is consistent with a simple circuit model for the FM/NM bilayer.

Thus, far, to attain more accurate $\theta_{ISH}$, many studies have been decreased the AHE terms. When the $V_{ISHE}$ measurement is performed, the bilayer sample was placed at the center of a resonant cavity where the magnetic field was maximized, while $E_{MW}$ was minimized, which in turn minimizes the AHE. On the other hand, the selection of microwave modes is important despite the optimized centering. The AHE contribution of the TE011 mode is larger than that of the TE102 mode. In case of the TE011 mode, the bilayer sample is easily exposed to $E_{MW}$ in all directions. However, these previous results remain unclear on the origin or correlation of the ISHE and AHE in bilayer structures.

In this paper, the dependence of NMs on measuring the DC voltage $V$ under FMR conditions is experimentally investigated to examine the CoFeB/NM and NM/CoFeB bilayers, where NM = Pt, Pd, Ta, and Ti. The sign of the directional $V_{ISHE}$ depends on the stacking order of layers between CoFeB and NM, which are designed as CoFeB/NM or NM/CoFeB. However, the $V_{AHE}$ sign is not associated with the order of the layer. Therefore, the pure $V_{ISHE}$ and $V_{AHE}$ can be separated utilizing the method of addition and subtraction in the bilayer structure. Then, the estimated $\theta_{ISH}$ from the pure $V_{ISHE}$ is compared with the reported values for different NMs. Both Ta and Ti materials show a broad deviation in $\theta_{ISH}$, which originates from the enormous AHE in
across with the high resistivity of NMs. Therefore, the NM characteristics should be simultaneously satisfied with large $\theta_{\text{ISH}}$ and low resistivity to obtain reasonable and pure $V_{\text{ISH}}$ values in a bilayer system.

II. EXPERIMENT

Figures 1(a) and 1(b) show schematic illustrations of the CoFeB/NM and NM/CoFeB (NM = Pt, Pd, Ta, and Ti) bilayers used in these experiments, respectively. The samples are bilayer systems comprised of a 10-nm-thick ferromagnetic CoFeB layer and a 10-nm-thick NM layer sputtered on thermally oxidized Si substrates. All bilayer samples have a size of 1 mm (width) $\times$ 2 mm (length). Two electrodes are connected to the ends of the sample. The present experiments minimized the effect of $E_{\text{MW}}$ by optimizing the device under test (DUT) centering by placing the DUT near the center of a TE$_{011}$ cylindrical cavity. The external magnetic field $H$ is applied perpendicular to the direction across the electrodes. Figure 1(a) shows that $V$ measured for the CoFeB/NM bilayer is described using Eqs. (1) and (2) as

$$V_{\text{CoFeB/NM}} = V_{\text{ISH}}('/ / J_0 \times \sigma) + V_{\text{AHE}}('/ / E_{\text{MW}} \times m).$$

On the other hand, $V$ measured for the NM/CoFeB bilayer is also expected to be

$$V_{\text{NM/CoFeB}} = -V_{\text{ISH}}('/ / -J_0 \times \sigma) + V_{\text{AHE}}('/ / E_{\text{MW}} \times m).$$

Ultimately, the $J_0$-sign governed by the directional spin pumping is changed by inverting the orders of the layers, as shown in Fig. 1(b), which looks like the bilayer sample is rotated. For example, the sign of $V_{\text{ISH}}$ is reversed by reversing the $H$ or $J_0$-directions. However, $V_{\text{ISH}}$ delicately contributes to the variation of the $E_{\text{MW}}$ profile at the center of the cavity resonator. Moreover, it is difficult to separate these two effects because the sign of the effects can have the same directions, depending on the rotation of the sample. Therefore, structurally exchanging FM and NM allows us to obtain the pure $V_{\text{ISH}}$ and $V_{\text{AHE}}$ without changing the microwave profile of cavity inside.

In the FMR measurements, the microwave frequency is 9.46 GHz and the power is 50 mW. The resonance field $H_0$ is around 857 Oe. Simultaneously, the value of $V$ for the various NMs is measured under the FMR condition.

III. RESULTS AND DISCUSSION

Figure 1(c) shows the FMR spectra for the NM/CoFeB (NM = Pt, Pd, Ta, and Ti) bilayer and a CoFeB sample. Here, $I$ denotes the microwave absorption intensity. The half width at half maximum of $I(H)$, $W = 4\pi f_0 \delta / \sqrt{3} \gamma$, shown in the inset, for the CoFeB sample is clearly enhanced by attaching the NMs layer, where $f_0$, $\gamma$, and $\delta$ are the resonance frequency, the Gilbert damping constant, and the gyro-magnetic ratio, respectively. These results experimentally demonstrate the evidence of the direct ISHE contribution due to the spin-pumping method. The value of $x$ for the CoFeB sample is estimated to be 0.0077 from the FMR spectral width, which agrees reasonably well with the reported $x = 0.015$ for a 5-nm CoFeB sample. It is two times larger than that of the 10-nm CoFeB sample, which is given by $x \propto 1/(\text{thickness of FM layer})$.

Figures 2(a) and 2(b) show the $H$ dependence of $V$ measured for the NM/CoFeB and CoFeB/NM bilayers. The measured $V$ signals under the FMR condition are well fitted using the following function:

$$V(H) = V_{\text{ISH}}(H) + V_{\text{AHE}}(H) = V_{\text{ISH}} \left( \frac{\Gamma^2}{(H - H_0)^2 + \Gamma^2} + V_{\text{AHE}} \frac{-2\Gamma(H - H_0)}{(H - H_0)^2 + \Gamma^2} \right),$$

where the first and second terms indicate the ISHE ($V_{\text{ISH}}$ indicates the magnitude of the first term) and AHE ($V_{\text{AHE}}$ indicates the magnitude of the second term) contributions, respectively, and $\Gamma$ is the damping constant. Here, $V_{\text{ISH}}$ and $V_{\text{AHE}}$ are proportional to the inverse spin Hall coefficient in NMs and the anomalous Hall coefficient in CoFeB, respectively. The signs of the $V$ signals using the NM layer of Pt and Pd are distinctly reversed between the NM/CoFeB (Fig. 2(a)) and CoFeB/NM (Fig. 2(b)) bilayers. Furthermore, the spectra shapes using Pt and Pd are well reproduced using simple Lorentzian functions. These results indicate that the ISHE contribution dominates the measured $V$ signal, which is estimated as $V_{\text{ISH}}/V_{\text{AHE}} \approx 23$ (for Pt/CoFeB) and 27 (for Pd/CoFeB).

However, in case of the NM layer of Ta and Ti, despite the inverted layer order, the $V$ signal shows that it is difficult to understand the reversed sign of $V_{\text{ISH}}$ under the primary

![FIG. 1](image-url)  
(a) Schematic illustration of the bilayer sample (CoFeB/NM) used in the present study, where $V_{\text{ISH}}$ and $V_{\text{AHE}}$ have the same sign. (b) Schematic illustration of the bilayer sample (NM/CoFeB) used in the present study. Here, $V_{\text{ISH}}$ and $V_{\text{AHE}}$ have opposite signs. (c) The FMR spectra for the CoFeB single layer and NM/CoFeB (NM = Pt, Pd, Ta, and Ti) bilayer in the TE$_{011}$ resonant cavity.

![FIG. 2](image-url)  
The $H$ dependence of $V$ for (a) NM/CoFeB and (b) CoFeB/NM bilayer samples, where $NM =$ Pt, Pd, Ta, and Ti.
ISHE contribution. These results indicate that the analysis of the \( V \) signal for the case of Ta and Ti-NM layer precisely consider not only the ISHE but also the AHE contribution in bilayer structures, which are estimated as \( V_{\text{ISH}}/V_{\text{AH}} \approx 7 \) (for Ta/CoFeB) and 1.8 (for Ti/CoFeB).

Figure 3 shows the \( H \) dependence of pure \( V_{\text{ISH}} \) in the bilayer structures. To properly separate the pure spin-pumping contribution, the pure \( V_{\text{ISH}} \) is described utilizing the subtraction method from Eqs. (3) and (4) as

\[
V_{\text{NM/CoFeB}} - V_{\text{CoFeB/NM}} = 2V_{\text{ISH}}(H). \tag{6}
\]

Compared with previous experimental results (Fig. 2), note that the \( V_{\text{ISH}} \) signal is well defined by the Lorentzian functions in all NMs, which excludes the AHE contribution from the measured \( V \) signals. In particular, the Ta and Ti-NMs cases can be used to consider only the ISHE contribution in spite of the large AHE. The bilayer structure utilizing the Ta-NM show the opposite sign of \( \theta_{\text{ISH}} \) compared to the other NMs.

Here, \( V_{\text{ISH}} \) is designated as the peak height of the Lorentzian function in the \( V_{\text{ISH}} \) spectrum (see Fig. 3). For the bilayer systems, \( V_{\text{ISH}} \) is given by

\[
V_{\text{ISH}} \approx \lambda_{\text{NM}} \tanh\left(\frac{d_{\text{NM}}}{2\lambda_{\text{NM}}}\right) \left(\frac{R_{\text{NM}}R_{\text{CoFeB}}}{R_{\text{NM}} + R_{\text{CoFeB}}}\right) \times \theta_{\text{ISH}}(W_{\text{NM/CoFeB}} - W_{\text{CoFeB}}), \tag{7}
\]

where \( \lambda_{\text{NM}} \) is the spin diffusion length in NM, \( d_{\text{NM}} \) is the thickness of NM, \( R_{\text{NM}} \) is the resistance of NM layer, \( R_{\text{CoFeB}} \) is the resistance of CoFeB layer, \( W_{\text{NM/CoFeB}} \) is the FMR spectral width of NM/CoFeB bilayer, and \( W_{\text{CoFeB}} \) is the FMR spectral width of CoFeB layer, respectively. The inset in Fig. 3 shows the \( R_{\text{NM}} \) for Pt, Pd, Ta, and Ti and \( R_{\text{CoFeB}} \), which is measured for a single layer. To obtain \( \theta_{\text{ISH}} \) for various NMs, we used Eq. (7) with the parameters \( \lambda_{\text{NM}} = 10\,\text{nm} \) (Pt), 9 nm (Pd), 1.8 nm (Ta), and 300 nm (Ti), the difference between \( W_{\text{NM/CoFeB}} \) and \( W_{\text{CoFeB}} \) (see Fig. 1(c)), and the \( V_{\text{ISH}} \) values. We obtained \( \theta_{\text{ISH}} \) of Pt, Pd, Ta, and Ti as \( \theta_{\text{ISH}} = 0.03 \) (Pt), 0.01 (Pd), –0.02 (Ta), and 0.0021 (Ti), respectively, which are consistent with the literature values.\(^{7–9}\) Finally, the bilayer samples with the Ta and Ti-NM layer (related to large AHE) exhibit different \( V_{\text{ISH}} \) values (\( V_{\text{ISH}} = -42\,\mu\text{V} \) (or 11.2 \( \mu \text{V} \)) for Ta/CoFeB (or CoFeB/Ta); \( V_{\text{ISH}} = -20\,\mu\text{V} \) (or –29.1 \( \mu\text{V} \)) for Ti/CoFeB (or CoFeB/Ti)) according to the order of the layers (see Figs. 2(a) and 2(b)), the \( E_{\text{MW}} \) profile of the cavity inside, possibility of oxidation problem depending on an air exposure, and so on, which may result in controversial and deviated \( \theta_{\text{ISH}} \) values. On the other hand, the bilayer samples with the Pt and Pd-NM layer (related to little AHE) show almost same \( V_{\text{ISH}} \) values (\( V_{\text{ISH}} = 30\,\mu\text{V} \) (or –28 \( \mu\text{V} \)) for Pt/CoFeB (or CoFeB/Pt); \( V_{\text{ISH}} = 17.6\,\mu\text{V} \) (or –16.6 \( \mu\text{V} \)) for Pd/CoFeB (or CoFeB/Pd)) despite changing the order of the layers.

Figure 4 shows the \( H \) dependence of pure \( V_{\text{AH}} \) in the bilayer structures. Against the aforementioned analysis, the pure \( V_{\text{AH}} \) utilizing the addition method is described using Eqs. (3) and (4) as

\[
V_{\text{NM/CoFeB}} + V_{\text{CoFeB/NM}} = 2V_{\text{AH}}(H). \tag{8}
\]

The shape of the pure \( V_{\text{AH}} \) spectra is consistent with the above-mentioned prediction. Here, \( V_{\text{AH}} \) is designated as half of the peak-to-peak height of the \( V_{\text{AH}} \) spectrum. The \( V_{\text{AH}} \) values of the Pt and Pd NM cases are also smaller than that of the Ta and Ti cases in the NM/FM bilayers. To analyze the contribution of AHE in the bilayers, we assume that the NM/FM bilayers are two independent resistors (\( R_{\text{FM}} \) or \( R_{\text{NM}} \)) in parallel (see the inset to Fig. 4), which is known as the two-channels model. In this model, it is also assumed that the Hall voltage is only generated by the FM layer. Then, the total current \( I_t \) and voltage \( V_t \) along the sample can be expressed as \( I_t = I_{\text{FM}} + I_{\text{NM}} \) and \( V_t = R_{\text{FM}} I_{\text{FM}} = R_{\text{NM}} I_{\text{NM}} \), where the FM and NM subscripts indicate the FM and NM layer, respectively. Consequently, \( V_{\text{AH}} \) from the FM (= CoFeB) layer can be given by

\[
V_{\text{AH}} = \left(\frac{\rho_{\text{AH}}}{d_{\text{CoFeB}}}\right) \left(\frac{I_t}{(R_{\text{CoFeB}}/R_{\text{NM}}) + 1}\right) \approx \frac{1}{(R_{\text{CoFeB}}/R_{\text{NM}}) + 1}, \tag{9}
\]

where \( d_{\text{CoFeB}} \) and \( \rho_{\text{AH}} \) are the thickness of CoFeB layer and the anomalous Hall resistivity in bilayer, respectively. Based on Eq. (9), the \( V_{\text{AH}} \) values of the Pt and Pd NM case are

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**Fig. 3.** The \( H \) dependence of \( V_{\text{ISH}} \) based on Eq. (6). The inset shows the resistance of the single layer \( (1\,\text{mm} \times 2\,\text{mm}) \) for Pt, Pd, CoFeB, Ta, and Ti.

**Fig. 4.** The \( H \) dependence of \( V_{\text{AH}} \) based on Eq. (8). The inset shows an equivalent circuit model for the AHE contribution.
smaller than that of Ta and Ti NM case. That is, the AHE contribution is significantly decreased for the $R_{\text{NM}}/R_{\text{FM}} > 1$ condition such as $R_{\text{CoFeB}}/R_{\text{Pt}} = 7.6$ and $R_{\text{CoFeB}}/R_{\text{Pd}} = 5.8$. On the contrary, the AHE contribution is fairly dominant for the $R_{\text{FM}}/R_{\text{NM}} < 1$ condition such as $R_{\text{CoFeB}}/R_{\text{Ta}} = 0.84$ and $R_{\text{CoFeB}}/R_{\text{Ti}} = 0.58$. In the bilayer system, it is important that the relative contributions between the ISHE and AHE depend on the $R$-ratio between the FM and NM layer. When only the ISHE term is considered in the bilayer, $V_{\text{ISHE}}$ was previously written as a simple circuit model ($V_{\text{ISHE}} \propto R_{\text{FM}}R_{\text{NM}}/(R_{\text{FM}} + R_{\text{NM}})$). Highly resistive NMs were employed in the NM layer to obtain the large $V_{\text{ISHE}}$ value. However, according to the above results, it is unavoidable that NMs with large $R$ values exhibit the unwanted AHE in bilayer samples.

IV. CONCLUSIONS

The dependence of the measured voltage on the NM in bilayer systems has been investigated under FMR conditions in the TE011 resonant cavity. The directional change of $V_{\text{ISHE}}$ in the bilayer, $V_{\text{ISHE}}$, was previously written as a simple circuit model ($V_{\text{ISHE}} \propto R_{\text{FM}}R_{\text{NM}}/(R_{\text{FM}} + R_{\text{NM}})$). Highly resistive NMs were employed in the NM layer to obtain the large $V_{\text{ISHE}}$ value. However, according to the above results, it is unavoidable that NMs with large $R$ values exhibit the unwanted AHE in bilayer samples.

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