Spin absorption at ferromagnetic-metal/platinum-oxide interface

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We investigate the absorption of a spin current at a ferromagnetic-metal/Pt-oxide interface by measuring current-induced ferromagnetic resonance. The spin absorption was characterized by the magnetic damping of the heterostructure. We show that the magnetic damping of a Ni₈₁Fe₁₉ film is clearly enhanced by attaching Pt-oxide on the Ni₈₁Fe₁₉ film. The damping enhancement is disappeared by inserting a ultrathin Cu layer between the Ni₈₁Fe₁₉ and Pt-oxide layers. These results demonstrate an essential role of the direct contact between the Ni₈₁Fe₁₉ and Pt-oxide to induce sizable interface spin-orbit coupling. Furthermore, the spin-absorption parameter of the Ni₈₁Fe₁₉/Pt-oxide interface is comparable to that of intensively studied heterostructures with strong spin-orbit coupling, such as an oxide interface, topological insulators, metallic junctions with Rashba spin-orbit coupling. This result illustrates strong spin-orbit coupling at the ferromagnetic-metal/Pt-oxide interface, providing an important piece of information for quantitative understanding the spin absorption and spin-charge conversion at the ferromagnetic-metal/metallic-oxide interface.

I. INTRODUCTION

An emerging direction in spintronics aims at discovering novel phenomena and functionalities originating from spin-orbit coupling (SOC). An important aspect of the SOC is the ability to convert between charge and spin currents. The charge-spin conversion results in the generation of spin-orbit torques in heterostructures with a ferromagnetic layer, enabling manipulation of magnetization in tungsten, which is attributed to the interfacial effect. The oxidation of a nonmagnetic layer in the heterostructure also offers a route to engineer the spin-orbit devices. Demasius et al. reported a significant enhancement of the spin-torque generation by incorporating oxygen into tungsten, which is attributed to the interfacial effect. The spin-torque generation efficiency was found to be significantly enhanced by manipulating the oxidation of Cu, enabling to turn the light metal into an efficient spin-torque generator, comparable to Pt. We also reported that the oxidation of Pt turns the heavy metal into an electrically insulating generator of the spin-orbit torques, which enables the electrical switching of perpendicular magnetization in a ferrimagnet sandwiched by insulating oxides. These studies have provided valuable insights into the oxide spin-orbitronics and shown a promising way to develop energy-efficient spintronics devices based on metal oxides.

The SOC in solids is responsible for the relaxation of spins, as well as the conversion between charge and spin currents. The spin relaxation due to the bulk SOC of metals and semiconductors has been studied both experimentally and theoretically. The influence of the SOC at interfaces on spin-dependent transport has also been recognized in the study of giant magnetoresistance (GMR). The GMR in Cu/Pt multilayers in the current-perpendicular-to-plane geometry indicated that there must be a significant spin-memory loss due to the SOC at the Cu/Pt interfaces. The interface SOC also plays a crucial role in recent experiments on spin pumping. The spin pumping refers to the phenomenon in which precessing magnetization emits a spin current to the surrounding nonmagnetic layer. When the pumped spin current is absorbed in the nonmagnetic layer due to the bulk SOC or the ferromagnetic/nonmagnetic interface due to the interface SOC, the magnetization damping of the ferromagnetic layer is enhanced because the spin-current absorption deprives the magnetization of the angular momentum. Although the damping enhancement induced by the spin pumping has been mainly associated with the spin absorption in the bulk of the nonmagnetic layer, recent experimental and theoretical studies have demonstrated that the spin-current absorption at interfaces also provides a dominant contribution to the damping enhancement. Since the absorption of a spin current at interfaces originates from the SOC, quantifying the damping enhancement provides an important information for fundamental understanding of the spin-orbit physics.

In this work, we investigate the absorption of a spin current at a ferromagnetic-metal/Pt-oxide interface. We show that the magnetic damping of a Ni₈₁Fe₁₉ (Py) film is clearly enhanced by attaching Pt-oxide, Pt(O), despite the absence of the absorption of the spin current in the bulk of the Pt(O) layer. The damping enhancement disappears by inserting an ultrathin Cu layer between the Py and Pt(O) layers. This result indicates that the direct contact between the ferromagnetic metal and Pt oxide is essential to induce the sizable spin-current absorption, or the interface SOC. We further show that the strength of the damping enhancement observed for the Py/Pt(O) bilayer is comparable with that reported for other systems with strong SOC, such as two-dimensional electron gas (2DEG) at an oxide interface and topological insulators.
II. EXPERIMENTAL METHODS

Three sets of samples, Au/SiO$_2$/Py, Au/SiO$_2$/Py/Pt(O) and Au/SiO$_2$/Py/Cu/Pt(O), were deposited on thermally oxidized Si substrates (SiO$_2$) by RF magnetron sputtering at room temperature. To avoid the oxidation of the Py or Cu layer, we first deposited the Pt(O) layer on the SiO$_2$ substrate in a mixed argon and oxygen atmosphere. After the Pt(O) layer was deposited from a SiO$_2$ target in the pure argon atmosphere. For the Pt(O) deposition, the amount of oxygen gas in the mixture was fixed as 30%, in which the flow rates of argon and oxygen were set as 7.0 and 3.0 standard cubic centimeters per minute (sccm), respectively. The SiO$_2$ layer was deposited from a SiO$_2$ target in the pure argon atmosphere. The film thickness was controlled by the deposition time with a precalibrated deposition rate.

We measured the magnetic damping using current-induced ferromagnetic resonance (FMR). For the fabrication of the devices used in the FMR experiment, the photolithography and lift-off technique were used to pattern the films into a 10 $\mu$m $\times$ 40 $\mu$m rectangular shape. A blanket Pt(O) film on a 1 cm $\times$ 1 cm SiO$_2$ substrate was fabricated for the composition confirmation by x-ray photoelectron spectroscopy (XPS). We also fabricated Pt(O) single layer and SiO$_2$/Py/Pt(O) multilayer films with a Hall bar shape to determine the resistivity of the Pt(O) and Py using the four-probe method. The resistivity of Pt(O) (6.3 $\times$ 10$^6$ $\mu$Ω cm) is much larger than that of Py (106 $\mu$Ω cm). Because of the semi-insulating nature of the Pt(O) layer, we neglect the injection of a spin current into the Pt(O) layer from the Py layer; only the Py/Pt(O) interface can absorb a spin current emitted from the Py layer. Transmission electron microscopy (TEM) was used to directly observe the interface and multilayer structure of the SiO$_2$/Py/Pt(O) film. All the measurements were conducted at room temperature.

III. RESULTS AND DISCUSSION

Figure 1(a) exhibits the XPS spectrum of the Pt(O) film. Previous XPS studies on Pt(O) show that binding energies of the Pt 4$f_{7/2}$ peak for Pt, PtO and PtO$_2$ are around 71.3, 72.3 and 74.0 eV, respectively. Thus, the Pt 4$f_{7/2}$ peak at 72.3 eV in our Pt(O) film indicates the formation of PtO. By further fitting the XPS spectra, we confirm that the Pt(O) film is composed of a dominant structure of PtO with a minor portion of PtO$_2$. Figure 1(b) shows the cross-sectional TEM image of the SiO$_2$(4 nm)/Py(8 nm)/Pt(O)(10 nm) film. As can be seen, continuous layer morphology with smooth and distinct interfaces is formed in the multilayer film. Although we deposited the Py layer on the Pt(O) layer to avoid the oxidation of the Py, it might still be possible that the Py layer is oxidized by the Pt(O) layer. Therefore, we measured the resistance of the Au/SiO$_2$/Py and Au/SiO$_2$/Py/Pt(O) samples used in the FMR experiment. The resistance of both samples show the same value (60 $\Omega$). Furthermore, as described in the following section, the saturation magnetization for each device was obtained by using Kittel formula (0.746 T and 0.753 T for the Au/SiO$_2$/Py and Au/SiO$_2$/Py/Pt(O), respectively). The only 1% difference indicates that the minor oxidation of the Py layer due to the presence of the Pt(O) layer can be neglected.

Next, we conduct the FMR experiment to investigate the absorption and relaxation of spin currents induced by the spin pumping. Figure 2(a) shows a schematic of the experimental setup for the current-induced FMR. We applied an RF current to the device, and an in-plane external magnetic field $\mu_0H$ was swept with an angle of 45$^\circ$ from the longitudinal direction. The RF charge current flowing in the Au layer generates an Oersted field, which drives magnetization precession in the Py layer at the FMR condition. The magnetization precession induces an oscillation of the resistance of the device due to the anisotropic magnetoresistance (AMR) of the Py layer. We measured DC voltage generated by the mixing of the RF current and the oscillating resistance using a bias tee.

Figures 2(b), 2(c) and 2(d) show the FMR spectra for the Au/SiO$_2$/Py, Au/SiO$_2$/Py/Pt(O) and Au/SiO$_2$/Py/Cu/Pt(O) films, respectively. For the FMR measurement, a small RF current power $P = 5$ mW was applied. Around $P = 5$ mW, the FMR linewidth is independent of the RF power as shown in the inset to Fig. 3(a). This confirms that the measured linewidth is unaffected by additional linewidth broadening due to nonlinear damping mechanisms and Joule heating. As shown in Fig. 2, clear FMR signals with low noise are obtained, allowing us to precisely fit the spectra and extract the magnetization damping for the three samples. Here, the mixing voltage due to the FMR, $V_{\text{mix}}$, is ex-
pressed as

\[ V_{\text{mix}} = V_{\text{sym}} \left( \frac{(\mu_0 \Delta H)^2}{(\mu_0 H - \mu_0 H_R)^2 + (\mu_0 \Delta H)^2} \right) + V_{\text{asy}} \left( \frac{\mu_0 \Delta H (\mu_0 H - \mu_0 H_R)}{\mu_0 H - \mu_0 H_R} \right)^2 + (\mu_0 \Delta H)^2, \]

where \( \mu_0 \Delta H \) and \( \mu_0 H_R \) are the magnitudes of the symmetric and antisymmetric components. The symmetric and antisymmetric components arise from the spin-orbit torques and Oersted field. In the devices used in the present study, the Oersted field created by the top Au layer dominates the RF effective fields acting on the magnetization in the Py layer [see also Fig. 2(a)]. The large Oersted field enables the electric measurement of the FMR even in the absence of the spin-orbit torques in the Au/SiO\(_2\)/Py film.

The damping constant \( \alpha \) of the Py layer in the Au/SiO\(_2\)/Py, Au/SiO\(_2\)/Py/Pt(O) and Au/SiO\(_2\)/Py/Cu/Pt(O) films can be quantified by fitting the RF current frequency \( f \) dependence of the FMR spectral width \( \mu_0 \Delta H \) using

\[ \mu_0 \Delta H = \mu_0 \Delta H_{\text{ext}} + \frac{2\pi \alpha}{\gamma} f, \]

where \( \Delta H_{\text{ext}} \) and \( \gamma \) are the inhomogeneous linewidth broadening of the extrinsic contribution and gyromagnetic ratio, respectively. Figure 3(a) shows the \( f \) dependence of the FMR linewidth \( \mu_0 \Delta H \), determined by fitting the spectra shown in Fig. 2 using Eq. (1). As shown in Fig. 3(a), the frequency dependence of the linewidth is well fitted by Eq. (2). Importantly, the slope of the \( f \) dependence of \( \mu_0 \Delta H \) for the Py/Pt(O) film is clearly larger than that for the Py and Py/Cu/Pt(O) films. This indicates larger magnetic damping in the Py/Pt(O) film. By using Eq. (2), we determined the damping constant \( \alpha \) as 0.0126, 0.0169 and 0.0124 for the Py, Py/Pt(O) and Py/Cu/Pt(O) films, respectively. The difference in \( \alpha \) between the Py and Py/Cu/Pt(O) films is vanishingly small, which is within an experimental error. In contrast, the damping of the Py/Pt(O) film is clearly larger than that of the other films, indicating an essential role of the Py/Pt(O) interface on the magnetization damping.

The larger magnetic damping in the Py/Pt(O) film demonstrates an important role of the direct contact between the Py and Pt(O) layers in the spin-current absorption. If the bottom layers influence the magnetic properties of the Py layer, the difference in the magnetic properties can also result in the different magnetic damping in the Au/SiO\(_2\)/Py, Au/SiO\(_2\)/Py/Pt(O) and Au/SiO\(_2\)/Py/Cu/Pt(O) films. However, we have confirmed that the difference in the magnetic damping is not caused by different magnetic properties of the Py layer. In Fig. 3(b), we plot the RF current frequency \( f \) dependence of the resonance field \( \mu_0 H_R \). As can be seen, the \( f \) dependence of \( \mu_0 H_R \) is almost identical for the different samples, indicating the minor change of the magnetic properties of the Py layer due to the different bottom layers. In fact, by fitting the experimental data using Kittel formula, \( 2\pi f / \gamma = \sqrt{\mu_0 H_R (\mu_0 H_R + \mu_0 M_s)} \), the saturation magnetization is obtained to be \( \mu_0 M_s = 0.746, 0.753 \) and 0.777 T for the Py, Py/Pt(O) and Py/Cu/Pt(O) films, respectively. The minor difference (< 5%) in the saturation magnetization indicates that the larger damping of the Py/Pt(O) film cannot be attributed to possible different magnetic properties of the Py layer. Thus, the larger magnetic damping of the Py/Pt(O) film can only be attributed to the efficient absorption of the spin current at the interface. Notable is that the additional damping due to the spin-current absorption disappears by inserting the 3.6 nm-thick Cu layer between the Py and Pt(O) layers. Here, the thickness of the Cu layer is much thinner than its spin-diffusion length (~ 500 nm), allowing us to neglect the relaxation of the spin current in the Cu layer. This indicates that the direct contact between the Py and Pt(O) layers is essential for the absorption of the spin current at the interface, or the interface SOC.
Here, $g$ and $\Gamma$ are the g-factor and damping constant, respectively. The inset shows the frequency $f$ dependence of the resonance field $H$ for the Py film at $f = 7$ GHz. (b) The RF current power $P$ dependence of $\mu_0H_R$ for the three samples. The solid curves are the fitting result using the Kittel formula.

FIG. 3. (a) The RF current frequency $f$ dependence of the half-width at half-maximum $\mu_0\Delta H$ for the Py, Py/Pt(O) and Py/Cu/Pt(O) samples. The solid lines are the linear fit to the experimental data. The inset shows RF current power $P$ dependence of $\mu_0\Delta H$ for the Py film at $f = 7$ GHz. (b) The RF current power $f$ dependence of the resonance field $\mu_0H_R$ for the three samples. The solid curves are the fitting result using the Kittel formula.

| Heterostructure | $\Delta\alpha$ | $\Gamma_0\eta$ [1/m$^2$] | Ref. |
|-----------------|-----------------|--------------------------|------|
| Bi/Ag/Ni$_{50}$Fe$_{20}$ | 0.015 | $8.7 \times 10^{-18}$ | [25] |
| Bi$_2$O$_3$/Cu/Ni$_{50}$Fe$_{20}$ | 0.0045 | $1.5 \times 10^{-18}$ | [26] |
| SrTiO$_3$/LaAlO$_3$/Ni$_{81}$Fe$_{19}$ | 0.0013 | $2.3 \times 10^{-18}$ | [27] |
| Pt(O)/Ni$_{81}$Fe$_{19}$ | 0.0044 | $2.3 \times 10^{-18}$ | This work |
| $\alpha$-Sn/Ag/Fe | 0.022 | $1.2 \times 10^{-19}$ | [28] |
| Sn$_{0.02}$Bi$_{0.88}$Sb$_{0.9}$Te$_2$/Ni$_{81}$Fe$_{19}$ | 0.013 | $1.4 \times 10^{-19}$ | [29] |
| Bi$_2$Se$_3$/Ni$_{81}$Fe$_{19}$ | 0.0013 | $2.5 \times 10^{-18}$ | [30] |

Table I. The summarized spin-absorption parameter $\Gamma_0\eta$ in different material systems. In order to directly compare this work with previous works, we used International System of Units. We used the magnetic permeability in vacuum $\mu_0 = 4\pi \times 10^{-7}$ H/m. $\Delta\alpha$ and $\Gamma_0\eta$ for the Sn$_{0.02}$Bi$_{0.88}$Sb$_{0.9}$Te$_2$/Ni$_{81}$Fe$_{19}$ is the values at $T < 100$ K.

To quantitatively discuss the spin absorption at the Py/Pt(O) interface and compare with other material systems, we calculate the spin absorption parameters. In a model of the spin pumping where the interface SOC is taken into account, the additional damping constant is expressed as

$$\Delta\alpha = \frac{g\mu_B\Gamma_0}{\mu_0M_s d} \left( \frac{1 + 6\eta\xi}{1 + \xi} + \frac{\eta}{2(1 + \xi)^2} \right).$$

Here, $g = 2.11$ is the g-factor, $\mu_B = 9.27 \times 10^{-24}$ Am$^2$ is the Bohr magneton, $d$ is the thickness of the Py layer, and $\Gamma_0$ is the mixing conductance at the interface. $\xi$ is the back flow factor; no backflow refers to $\xi = 0$ and $\xi = \infty$ indicates that the entire spin current pumped into the bulk flows back across the interface. $\eta$ is the parameter that characterizes the interface SOC. For the Py/Pt(O) film, $\xi$ can be approximated to be $\infty$ because of the spin pumping into the bulk of the semi-insulating Pt(O) layer can be neglected. Thus, Eq. (3) can be simplified as

$$\Delta\alpha = \frac{6g\mu_B\Gamma_0\eta}{\mu_0M_s d}.$$

Here, $6\Gamma_0\eta$ corresponds to the effective spin mixing conductance $g_{\text{eff}}^\uparrow$. From the enhancement of magnetic damping $\Delta\alpha$, we obtain $\Gamma_0\eta = 2.3 \times 10^{18}$ m$^{-2}$ for the Py/Pt(O) film. We further compared this value with $\Gamma_0\eta$ for other systems where efficient interface charge-spin conversion has been reported. As shown in Table I, the spin-absorption parameter $\Gamma_0\eta$ of the Py/Pt(O) film is comparable with that of the heterostructures with the strong SOC, such as the 2DEG at an oxide interface, topological insulators, as well as metal/oxide and metallic junctions with the Rashba SOC. This result therefore demonstrates the strong SOC at the Py/Pt(O) interface.

IV. CONCLUSIONS

In summary, we have investigated the spin-current absorption and relaxation at the ferromagnetic-metal/Pt-oxide interface. By measuring the magnetic damping for the Py, Py/Pt(O) and Py/Cu/Pt(O) structures, we show that the direct contact between Py and Pt(O) is essential for the absorption of the spin current, or the sizable interface SOC. Furthermore, we found that the strength of the spin-absorption parameter at the Py/Pt(O) interface is comparable to the value for intensively studied heterostructures with strong SOC, such as 2DEG at an oxide interface, topological insulators, metallic junction with Rashba SOC. The comparable value with these material systems illustrates the strong SOC at the ferromagnetic-metal/Pt-oxide interface. This indicates that the oxidation of heavy metals provides a novel approach for the development of the energy-efficient spintronics devices based the SOC.

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