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ABSTRACT

We report on the enhancement of spin Hall angle from the CoFeB/Pt interface by introducing nitrogen into the Pt thin film. Spin-torque ferromagnetic resonance measurements on the effective spin Hall angle ($\theta_{\text{SH}}$) reveal a non-monotonic variation as a function of the amount of nitrogen gas introduced, $Q$ in the film deposition, which peaks at $\theta_{\text{SH}} = 0.16$ when $Q$ is 8%. Our analysis shows that the $\theta_{\text{SH}}$ enhancement is mainly attributed to the increase in spin-dependent scattering at the interface. The effective magnetic damping decreases with increasing $Q$ due to the reduced spin–orbit coupling. The interfacial spin transparency is also observed to show improvement after the introduction of nitrogen. Moreover, the additional damping-like torque from the interface may also lead to the enhancement of the linewidth modulation.

Spin–orbit torque (SOT) has been widely studied due to its application in current-induced magnetization switching or in oscillators.1–3 Composed of spin-transfer torque magnetic random access memory, SOT-based magnetic memory is faster and has higher energy efficiency.4–9 In the ferromagnetic/ heavy metal (FM/HM) bilayer, SOT originates from the spin Hall effect (SHE) in the HM and/or the Rashba–Edelstein effect at the interface.10–13 In the case of SHE, the spin current generated from the HM due to spin–orbit coupling (SOC) is transmitted to the FM, and the spin-transfer torque is applied to the magnetic moment. Effective spin Hall angle (ESHA, $\theta_{\text{SH}}$) is the ratio of spin current to charge current, which is used to quantify the impact due to SOT. In order to find materials with effective large spin Hall angles, many efforts have been made.4–9 To date, large ESHA has been observed in HM,14–16,18,19 diluted alloys such as CuPt,20 PtBi,15 AuPt,21 topological insulators,20–22 and antiferromagnet materials.23

Recently, a significant influence on the generation of spin torque in metal oxides has been reported.25–27 An et al.27 reported a significant enhancement of the spin-torque generation by the natural oxidation of Cu. A higher spin-torque generation was also observed in oxygen incorporated tungsten thin film, which originated from the interface.28 An et al.27 also reported a giant spin-torque generation by incorporating oxygen into Pt and they concluded that the enhancement originates from the interface. Gao et al.28 conducted a study on the intrinsic damping-like SOT arising from Berry curvature in the Ni$_81$Fe$_{19}$/CuO$_x$ bilayer film, which is an order of magnitude larger than a field-like SOT. Ding et al.28 reported a large enhancement of the SOT efficiency in the TmIG/Pt system by capping with a CuO$_x$ layer. Engineering spin Hall source materials by oxidation may result in efficient SOT manipulation of adjacent FM layers. Chen et al.29 demonstrated that the damping-like SOT can be enhanced by engineering buffer layer via nitrogen doping in TaN/FM/MgO. However, there are a few studies on FM/metal nitride interfaces. In this work, we report the enhancement of the spin Hall angle by introducing nitrogen into Pt thin film. We find that the PtN$_x$ films exhibit improved interfacial spin transparency and reduced effective magnetic damping. The spin-dependent scattering at the interface was found to be the main mechanism for the non-monotonic behavior of $\theta_{\text{SH}}$ as a function of the amount of nitrogen gas in mixture $Q$. Moreover, the additional damping like torque from the interface may also lead to the enhancement of the linewidth modulation. Our findings open up a means of improving the spin–orbit torque efficiency in SOT-based devices.

The Co$_{40}$Fe$_{40}$B$_{20}$ (5 nm)/PtN$_x$ (5 nm) films were deposited on thermally oxidized Si(001) substrates by a magnetron sputtering...
system at room temperature. Before the deposition, the base pressure in the main chamber was better than 5 × 10⁻⁸ mTorr and the deposition pressure is 3 mTorr. The CoFeB films were deposited by applying pure argon gas with a flow of 20 SCCM (standard cubic centimeters per minute). For PtNx deposition, argon and nitrogen gases were introduced into the chamber, and the amount of nitrogen gas in the mixture, Q, was varied from 0 to 20% to change the nitrogen content of the PtNx films. The film thickness was controlled by the deposition time with a pre-calibrated deposition rate. For the fabrication of the devices used in the ST-FMR, the stacks were patterned into microstrips (length of 50 μm and width of 10 μm) using a combination of electron beam lithography and Ar ion milling techniques. Ta (5 nm)/Cu (200 nm)/Pt (3 nm) electrodes were also fabricated using electron beam lithography and liftoff following DC magnetron sputtering. The M–H loops were evaluated with a vibrating sample magnetometer (VSM). Single PtN blanket layer films were fabricated for the surface roughness measurement by atomic force microscopy (AFM) and the crystalline structure by X-ray diffraction (XRD). The input microwave power of ST-FMR was varied from 10 to 20 dBm and the measured θul were independent of the applied RF power, suggesting that measurement involves no significant microwave heating effect (see the supplementary material). All measurements were performed at an RF power of 18 dBm. The ST-FMR spectra were measured for microwave frequencies from 8 to 17 GHz for all samples. All the measurements were conducted at room temperature.

Figure 1(a) shows the X-ray diffraction pattern of the PtNx film for Q = 0% and Q = 8%. When no nitrogen is incorporated, a strong (111) peak is observed, indicating a highly (111)-oriented texture in the Pt film. By increasing Q to 8%, the Pt (111) peak is shifted to lower angles compared with that of pure Pt, indicating that nitrogen is incorporated into the Pt. The surface root mean square roughness \( R_{\text{rms}} \) in all the films with different Q are lower than 1 nm, as shown in Fig. 1(b), revealing a flat surface morphology of the PtNx films in the range of Q from 0% to 20%. Figure 1(c) shows the typical in-plane and out-of-plane magnetic hysteresis loops for Q = 8% sample. It exhibits a small magnetic coercivity, suggesting that the CoFeB layer is magnetically soft with in-plane magnetic anisotropy.

Figure 1(d) shows the schematic of the setup with an optical image of the patterned structure for the ST-FMR measurement. An RF current \( I_{\text{rf}} \) was injected along the longitudinal direction, and an in-plane external magnetic field \( H \) with an angle of 45° from the longitudinal direction of the device was applied and swept from 0 to 5000 Oe, with the microwave frequency fixed during each sweep. The \( I_{\text{rf}} \) generates a microwave-frequency SOT on the ferromagnetic layers, which induces magnetization precession. The magnetization precession then gives rise to an oscillation of the resistance due to anisotropic magnetoresistance (AMR). The rectified voltage \( V_{\text{mix}} \) due to the mixing of RF current and the oscillating resistance is measured by using a bias tee. The measured mixing dc voltage \( V_{\text{mix}} \) is expressed as

\[
V_{\text{mix}} = V_5 \frac{(\Delta H)^2}{(\Delta H)^2 + (H - H_{\text{res}})^2} + V_\Delta \frac{\Delta H(H - H_{\text{res}})}{(\Delta H)^2 + (H - H_{\text{res}})^2},
\]

where \( \Delta H, H_{\text{res}}, V_5 \) and \( V_\Delta \) are the resonance linewidth, the resonance field, the amplitudes of the symmetric and antisymmetric components of the mixing voltage, respectively. In the ST-FMR signal, the symmetric component is proportional to the damping-like effective torque, and the antisymmetric component is due to the sum of the Oersted field torque and the field-like effective torque.

The voltage signal from the spin pumping effect (\( V_{\text{sp}} \)) has a negligible effect on the symmetric component of the voltage (see the supplementary material), according to previous studies.

Figure 2(a) shows the ST-FMR spectra \( V_{\text{mix}} \) for CoFeB/PtNx devices with Q = 8% measured at a frequency range from 8 to 16 GHz. The resonance peak changes its sign by reversing the direction of the external magnetic field \( H \), suggesting that the damping-like torque is dominant compared to the Oersted field, which is consistent with the

![Materials characterization](image)

![ST-FMR measurements](image)
The effective damping constant ($\alpha_{\text{eff}}$) of $Q = 8\%$ sample is evaluated by fitting the $\Delta H$ vs $f$ data using the equation $\Delta H = \Delta H_{\text{inh}} + 2\pi f \alpha_{\text{eff}} / \gamma$, as shown in Fig. 2(d), where $\Delta H_{\text{inh}}$ is the frequency independent linewidth contribution from inhomogeneity in the magnetic film.\(^{10,27}\) From the fitting, the effective damping constant was found to be $(9.9 \pm 0.5) \times 10^{-3}$, which is lower than the value of CoFeB/Pt ($\sim 11.0 \times 10^{-3}$).\(^{40,41}\) The inhomogeneous linewidth is found to be $11.89 \pm 0.59$ Oe, which indicates a smooth interface and a high quality of the CoFeB/Pt\(_N\) heterostructure. Furthermore, the $\Delta H$ vs $f$ response in Fig. 2(d) shows a linear behavior over the entire frequency range, suggesting a negligible contribution from the non-linear two-magnon scattering mechanisms in the CoFeB/Pt\(_N\) film.\(^{40,42}\)

The effective spin Hall angle is the ratio of the spin current density to the RF current density.\(^{10,27}\) The ratio can be obtained from the line shape of the ST-FMR spectra without considering the spin current induced field-like torque.\(^{27}\) The $\theta_{\text{SH}}$ can be calculated as

$$\theta_{\text{SH}} = \frac{c f_S}{I_C} = \frac{V_S e \mu_0 M_{\text{std}}}{V_A h} \left( 1 + \frac{4\pi M_{\text{eff}}}{H_{\text{res}}} \right)^{-\frac{1}{2}},$$

where $I_C$ is the spin current density generated within the heavy metal, $f_S$ is the applied charge current density, $t$ is the CoFeB layer thickness, and $d$ is the heavy metal layer thickness. $\theta_{\text{SH}}$ can be calculated as a function of $f$, as shown in Fig. 3(a). For $8.0 < f < 16.0$ GHz, $\theta_{\text{SH}}$ can be obtained to be $0.12 \pm 0.01$ and $0.16 \pm 0.01$ for $Q = 0\%$ and $Q = 8\%$, respectively. The value of $\theta_{\text{SH}}$ in CoFeB/Pt is consistent with previous reports.\(^{40}\) The $\theta_{\text{SH}}$ increases non-monotonically with increasing $Q$, with a maximum at $Q = 8\%$, as shown in Fig. 3(d). Therefore, by incorporating nitrogen into the Pt layer, a giant enhancement of $\theta_{\text{SH}}$ by $33.3\%$ can be achieved.

The dependence of the effective damping constant on $Q$ is given in Fig. 3(c). As $Q$ increases from $0\%$ to $20\%$, the value of $\alpha_{\text{eff}}$ decreases from $(11.0 \pm 0.5) \times 10^{-3}$ to $(8.9 \pm 0.4) \times 10^{-3}$ ($\Delta H/\gamma \sim 20.4\%$ for $Q = 20\%$). However, the effective damping in the CoFeB/Pt\(_N\) bilayer is still larger than the intrinsic damping constant in the amorphous CoFeB thin film ($\sim 4.0 \times 10^{-3}$).\(^{43,44}\) The Co\(_{40}\)Fe\(_{40}\)B\(_{20}\) layers for different $Q$ were deposited by direct current magnetron sputtering with the same deposition condition. The intrinsic Gilbert damping should be almost the same. The additional extrinsic damping can be attributed to the loss of the angular momentum induced by the spin pumping effect, which originates from spin–orbit coupling.\(^{45}\) Therefore, the decreasing damping indicates that the strength of the SOC at the CoFeB/Pt\(_N\) interface is reduced by increasing $Q$. In contrast, a large damping enhancement ($\Delta H/\gamma \sim 33.3\%$ for $Q = 20\%$) was found in the Py/Pt(O) film due to the spin absorption at the interface.\(^{27}\) This suggests that the interface may play a different role in our CoFeB/Pt\(_N\) film.

For SOT-MRAM and SHNO applications, the energy efficiency is limited by the critical currents required for current-induced magnetization switching or oscillations.\(^{4,5}\) The critical switching current density for in-plane magnetization switching using SOT is given by

$$J_\text{cr} = \frac{2 e}{h} \frac{a}{\theta_{\text{SH}}} \left( \frac{4\pi M_{\text{eff}}}{M_{\text{std}}} \right).$$

Using Eq. (3), we calculated $J_\text{cr}$ with the data in Figs. 3(b) and 3(c) and plotted the results in Fig. 3(d). At $Q = 10\%$, $J_\text{cr}$ is at a minimum of $7.0 \pm 0.3$ MA/cm\(^2\), which is approximately $50\%$ less than that in pure Pt due to the large enhancement of $\theta_{\text{SH}}$ and low effective damping factor. Thus, the nitrogen-incorporated Pt films demonstrate great potential for applications as a spin Hall material for SOT-MRAM.\(^{45,46}\)
The dependence of the enhanced damping on PtN$_x$ thickness is given in Fig. 4. As the thickness increases, the value of $\Delta x$ increases in the lower thickness regime and saturates at higher thicknesses. This non-linear behavior also rules out the possible mechanism of extrinsic damping induced by the magnetic proximity effect, which has a quasi-linear thickness dependence.\textsuperscript{46,47} According to the spin pumping theory, the thickness dependence of the damping can be described by\textsuperscript{48}

$$\alpha = \alpha_{id} + \frac{g \mu_B \alpha}{4\pi M_0 t} G(t \alpha),$$

(4)

From the fitting, spin mixing conductance $G$ and spin diffusion length $\lambda_{ad}$ are found to be $(36.0 \pm 2.0) \times 10^{14}$ cm$^{-2}$, $2.8 \pm 0.1$ nm for $Q = 0\%$ and $(29.8 \pm 1.8) \times 10^{14}$ cm$^{-2}$, $1.5 \pm 0.2$ nm for $Q = 8\%$. The obtained $G$ for $Q = 0\%$ agrees with the previous report.\textsuperscript{37,40} The obtained $\lambda_{ad}$ for $Q = 0\%$ is close to the value of 2.4 nm reported by Liu et al.\textsuperscript{11} Since the resistivity is increased by nitrogen incorporation, the reduced $\lambda_{ad}$ for $Q = 8\%$ is consistent with the Elliott–Yafet spin-flip scattering model, in which the $\lambda_{ad}$ is approximately inversely proportional to $\rho$.\textsuperscript{37,44} According to the Elliott–Yafet model, a shorter spin diffusion length for $Q = 8\%$ than that for $Q = 0\%$ can be understood as a result of the spin scattering and/or the interface SOC.\textsuperscript{37} Furthermore, according to the drift-diffusion model, the interfacial spin transparency can be estimated by\textsuperscript{37,53,54}

$$T = \frac{G(t \alpha) \tanh \left( \frac{d}{2\lambda_{ad}} \right)}{G(t \alpha) \coth \left( \frac{d}{2\lambda_{ad}} \right) + \frac{h}{2\lambda_{ad}^2 \rho}},$$

(5)

where $0 \leq T \leq 1$, $T = 1$ when all of the injected spin current exerts a spin torque on the CoFeB layer through the interface, and $T = 0$ when it is dissipated before reaching the CoFeB layer. Using $G$ and $\lambda_{ad}$ obtained from the fitting, the interfacial spin transparency $T$ is calculated to be $0.58 \pm 0.02$ and $0.71 \pm 0.02$ for $Q = 0\%$ and $Q = 8\%$, respectively, which is in agreement with values reported in Co/Pt (0.3–0.65)\textsuperscript{42,55} and CoFeB/Pt (0.63).\textsuperscript{37} This indicates a better band matching at the interface for $Q = 8\%$ than for $Q = 0\%$.\textsuperscript{37} Moreover, a large interfacial spin transparency is benefit for energy efficient application of spin current in multilayered devices.\textsuperscript{53,54} However, the improvement in interfacial spin transparency can only partially explain the enhanced $\theta_{SE}$.\textsuperscript{37,40}

As an alternative approach to determine the value of the effective spin-torque efficiency, the linewidth modulation method was used by applying a dc-current during the ST-FMR measurement to modulate the linewidth.\textsuperscript{16,26} In this method, the contributions from spin pumping due to the inverse spin Hall effect and the field-like torque are insignificant.\textsuperscript{27,40}

Due to the SHE from the Pt layer, the spin accumulation induced by the DC at the interface generates a torque on the FM magnetization, which leads to a change of the linewidth.\textsuperscript{16} The dependence of the change of the linewidth on the applied dc current is given by\textsuperscript{16,40}

$$\Delta H(I_{DC}) - \Delta H(I_{DC} = 0) = \frac{2\pi f_s}{\gamma} \left( \frac{\sin \varphi}{H_{res}} + \frac{h}{2\pi M_s f_s} \right) I_s,$$

(6)

where $f_s = \frac{I_{dc} \rho_{ac}}{A_{c} R_{FM} + R_{NN}}$ is the spin current density in the PtN$_x$ layer, $A_{c}$ is the cross-sectional area of the PtN$_x$ layer, and $R_{FM}$ and $R_{NN}$ are the resistance of the ferromagnetic layer and PtN$_x$ layer, respectively. $\varphi$ is the angle between the magnetization and the applied field, which is 45° in our case. To rule out the heating induced modulation of the effective damping, $\Delta H^* = \frac{\Delta H(I_{DC}) - \Delta H(I_{DC} = 0)}{\Delta H(I_{DC} = 0)}$ as a function of $I_{DC}$ is plotted in the inset of Fig. 5(a). $\Delta H^*$ shows a linear behavior with $I_{DC}$ suggesting that heat-related effects are negligible in our measurements.\textsuperscript{36} Figs. 5(a) and 5(b) show the change of the linewidth as a function of dc-current for $Q = 0\%$ and $Q = 8\%$, respectively. The slopes of change of the linewidth vs $I_{DC}$ curves for the two field directions are almost equal, which confirms that the damping-like torque that acts on the magnetization in our CoFeB/PtN$_x$ bilayer is due to the SHE-generated spin current.\textsuperscript{40} The slope of the change in $\Delta H$ with DC-current ($\Delta H(I_{DC}) - \Delta H(I_{DC} = 0)$) for positive applied fields is 2.12 Oe/mA and 3.47 Oe/mA for $Q = 0\%$ and 8%, respectively.
enhanced spin Hall angle. Moreover, the interfacial spin–orbit coupling may play a minor role in our case due to the reduced SOC, which is different from observations of the interfacial Rashba spin–orbit torque affected by oxygen incorporation. The small change in $\theta_{3\text{SH}}$ despite significant changes in the bulk properties, namely, the resistivity, the effective magnetic damping, indicates that the mechanism responsible for the enhanced spin Hall angle is likely to originate at the PtN$_x$/CoFeB interface. The interfacial spin-dependent scattering may be a possible origin of the enhanced spin Hall angle. Moreover, the interfacial spin–orbit scattering generates a spin current owing to the interface SOC, which can diffuse from the interface to the ferromagnetic layer. It can also explain the non-monotonic variation of the $\theta_{3\text{SH}}$ with $Q$ as being due to a competition between the interfacial spin–orbit scattering that increases as a function of increasing $Q$ and the strength of SOC, which decreases as $Q$ increases. For lower $Q$, the enhancement of spin–orbit scattering is larger than the decrease in the SOC, which results in the enhancement of $\theta_{3\text{SH}}$. While for higher nitrogen content, the reduction of the SOC strength is larger than the increase in scattering, which leads to the reduction in $\theta_{3\text{SH}}$. A sizable damping-like SOT generation was also observed in the Ni$_8$Fe$_{19}$/CuO$_x$ bilayer film, which originates from the Berry curvature at the interface. Recently, Behera et al. also reported that the FM/TiN interface generates damping-like torque, which might originate from the Berry curvature. However, to determine the origin of enhanced linewidth modulation, detailed theoretical analysis and further experimental are required. Furthermore, the effect of the inhomogeneous broadening on the $\theta_{3\text{SH}}$ was examined, which shows different dependence with $Q$ from the $\theta_{3\text{SH}}$ (see the supplementary material).

In summary, we demonstrated the enhancement of the spin Hall angle by introducing nitrogen into the Pt thin film. We find the non-monotonic behavior of $\theta_{3\text{SH}}$ as a function of $Q$. The effective damping is found to decrease with increasing $Q$. Compared with other studies in metal oxide, we found that spin-dependent scattering at the interface is the main mechanism to the enhancement of the spin Hall angle. The additional damping-like torque from the interface also contributes to the improvement of the linewidth modulation. Our findings provide an encouraging route for the development of low energy consumption spintronic devices.

See the supplementary material for the detailed five parts, including input RF power dependence, angular dependence, inhomogeneous broadening, linewidth modulation of ST-FMR spectra, and $Q$-dependent resistivity.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.
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