Giant Spin Hall Effect in Cu–Tb Alloy Thin Films

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ABSTRACT: We report the giant spin current generation in CuTb alloys arising from the spin Hall effect. The maximum spin Hall angle from our CuTb-based magnetic heterostructures was found to be $-0.35 \pm 0.02$ for Cu$_{0.35}$Tb$_{0.65}$. We find that the contribution of skew scattering is larger than the side jump for lower Tb concentrations (<14.9%), while the converse is true for higher Tb concentrations. Additionally, we also studied the Gilbert damping parameter, spin diffusion length, and spin-mixing conductance. Interfacial spin transparency was found to be $0.55 \pm 0.03$ for the CoFeB/Cu$_{0.53}$Tb$_{0.47}$ interface. The spin diffusion length and spin-mixing conductance of the Cu$_{0.53}$Tb$_{0.47}$ alloy are $\lambda_\parallel = 2.5 \pm 0.3$ nm and $G^\parallel = (24.2 \pm 1.0) \times 10^{15}$ cm$^{-2}$, respectively. Our results pave a way for rare-earth metals to be used as a spin Hall material in highly efficient SOT devices.

KEYWORDS: spin Hall effect, spin–orbit coupling, spin–orbit torque, spin–orbit torque ferromagnetic resonance, rare-earth metal, spin Hall angle

1. INTRODUCTION

Spintronic devices such as spin–orbit torque magnetic random access memories and spin Hall nano-oscillators are key components of next-generation magnetic storage and logic devices because of their high response speed and low power consumption.1–3 Among them, the spin Hall effect (SHE) derived from the spin–orbit coupling (SOC) is a topic of immense interest because it can generate a longitudinal spin current when applying a lateral charge current, which is an efficient route for magnetization manipulation.4–9 The spin Hall angle represents the conversion efficiency of the charge and spin current. A large spin Hall angle enables efficient generation of the transfer torque and reduces the energy loss. Moreover, the SHE plays an important role in exciting or detecting the spin Seebeck effect.10–13 The large spin Hall angle can also lead to a high spin Seebeck coefficient in the magnon-driven spin Seebeck effect.14–16 Therefore, the search for large spin Hall angles has long been a key focus in the field of spintronics.17–20 To date, most studies have focused on heavy metals (HMs) such as Pt,1,21–23 β-Ta,24 and β-W,24 diluted alloys of CuBi,25 CuPt,26,27 AuW,28 WH,28 PdW,26 AuPt,27 and PtBi,28–30 and even antiferromagnetic materials.31–33 The future development of high-performance devices hinges on the discovery of new types of large spin Hall angle materials.

Rare-earth metals have recently received considerable attention because of their large SOC that arises from their large spins and orbital angular momentum in f-electron atoms and are expected to be a potential SOT source for spintronic applications.34–36 Several recent studies have reported that the spin Hall efficiency can be enhanced using rare-earth metals: Wong et al. have determined a large spin–orbit torque efficiency of $-0.480$ in Pt/[Co/Ni]$_3$/Co/Tb multilayers.36 Ueda et al. conducted a study on Pt/Co/Gd multilayers and explained that the large spin Hall efficiency stemmed from Gd.33 However, because of the strong spin-pumping effect caused by the strong SOC in heavy rare-earth materials, it is difficult to extract the spin Hall angle by using spin–torque ferromagnetic resonance. Reynolds et al. reported the lower bounds for the spin Hall torque ratio for Gd, Dy, Ho, and Lu by using a thin Hf spacer layer between the rare-earth and the FM to reduce the magnetic damping.34 Therefore, it would be particularly useful to be able to control not only the spin Hall angle but also the magnetic damping.

In this work, we studied the giant spin Hall angle and moderately low effective magnetic damping in CoFeB/Cu$_{1-x}$Tb$_x$ bilayers. This material system was chosen for several reasons. First of all, Cu-based alloys have been widely reported for studying the effect of alloying on SHE.15,19,25,26 Second, the Cu–Tb alloy can form a solid solution across a wide composition range from the phase diagram,37 making it a

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suitable system to understand the mechanism of extrinsic SHE. Furthermore, an amorphous Cu–Tb alloy thin film has been reported by McGuire and Gambino, which can be beneficial for pinning-free skyrmion dynamics because of the absence of grains. By conducting spin–orbit torque ferromagnetic resonance (ST-FMR) measurements, we show that the maximum spin Hall angle of our CuTb-based magnetic heterostructures was $-0.35 \pm 0.02$ for Cu$_{0.35}$Tb$_{0.65}$. From the extrinsic spin Hall resistivity induced by Tb, we found that the skew-scattering contribution dominates the extrinsic SHE at low Tb concentrations ($x < 14.9\%$), whereas the side jump contribution is dominant when $x > 14.9\%$. By varying the damping with CuTb thickness, we have determined the spin-mixing conductance increases with increasing Tb concentration. Meanwhile, the spin diffusion length is found to increase with decreasing Tb concentrations. Furthermore, using the drift-diffusion model, we have estimated the interfacial spin transparency. These results provide important information on the large SHE induced by extrinsic SHE mechanisms, opening a way for rare-earth metals to be used as a spin Hall material in highly efficient SOT devices.

2. EXPERIMENT AND METHOD

Thin film stacks of Co$_{0.9}$Fe$_{0.1}$B$_{20}$ (20 nm)/Cu$_{1-x}$Tb$_x$ (10 nm) were deposited on thermally oxidized Si(001) substrates by magnetron sputtering. The growth was carried out at room temperature with a base pressure below $5 \times 10^{-8}$ mTorr and a deposition pressure of 2 mTorr. Cu$_{1-x}$Tb$_x$ was deposited by cosputtering Cu and Tb targets. The sputtering power of the two targets was calibrated to tune the Tb concentration ($x$) in the Cu$_{1-x}$Tb$_x$ alloy. Subsequently, the stacks were patterned into strips (of length 50 $\mu$m and width 10 $\mu$m) 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 DC magnetron sputtering. The crystalline structure was examined by X-ray diffraction (XRD) and was found to be amorphous (refer to the Supporting Information, S1 XRD). The saturation magnetization was evaluated by using the vibrating sample magnetometer. The composition of alloy films was determined by energy-dispersive X-ray spectroscopy (EDX). A signal generator (Keysight N5183B) and a network analyzer were used to provide the radio frequency (RF) current for the ST-FMR and conventional FMR measurements, respectively. For ST-FMR studies, the DC voltage was measured with a Keithley 2000 multimeter. All the measurements were performed at room temperature.

3. RESULTS AND DISCUSSION

Figure 1a–d shows the CoFeB thickness $t$ dependence of the magnetic moment per unit area at saturation $m_s$ for $x = 0.05$, 0.13, 0.22, and 0.47, respectively. As the $m_s$ is expressed as $M_S(t - t_d)$, where $M_S$ is the saturation magnetization and $t_d$ is the magnetic dead layer thickness in CoFeB, we evaluate $M_S$ from the slope and $t_d$ from the horizontal intercept by fitting with a linear function. The effective saturation magnetization of CoFeB in the CoFeB/Cu$_{0.95}$Tb$_{0.05}$ system is found to be $M_S = 1049.8$ emu/cc with a magnetic dead layer $t_d = 1.55$ nm. The thickness of the dead layer $t_d$, saturation magnetization $M_S$, and coercivity $H_c$ are summarized in Figure 1e–g. As the concentration of Tb increases, $H_c$ decreases, while both $M_S$ and $t_d$ decrease. The thickness of the magnetic dead layer and saturation magnetization were found to be 0.35 nm and 932.9 emu/cc, respectively, for Cu$_{0.53}$Tb$_{0.47}$. The reduction in the $M_S$ value can be potentially due to compensation from the FM properties of Tb as the Tb atoms near the adjacent CoFeB atoms induces a non-zero magnetic moment. At the interface, the magnetic moments of Tb align antiparallel with the magnetic moments of CoFeB, leading to an antiferromagnetic coupling between the FM layer and Tb. Additionally, the antiferromagnetic coupling gives rise to enhanced $H_c$. The magnetic dead layer could be due to the diffusion and intermixing at the CoFeB/Cu$_{1-x}$Tb$_x$ interface, which indicates a better interface in the Tb-rich alloy.

The schematic diagram of the ST-FMR measurement setup is illustrated in Figure 2. A RF current $I_{RF}$ is applied to the structure through the RF port of a bias tee to generate a microwave frequency SOT on the ferromagnetic layers, which induces magnetization precession. This leads to an oscillation of the longitudinal resistance due to the spin Hall magnetoresistance effect. The rectified voltage $V_{mic}$ due to the mixing of RF current and the time-varying resistance is detected by using a lock-in amplifier. The ST-FMR spectra were measured for microwave frequencies from 6 to 25 GHz for all samples without an external DC bias. An in-plane magnetic field was applied at an angle of $45^\circ$ to maximize the ST-FMR signal and swept from 0 to 5000 Oe while keeping the microwave frequency constant for each measured frequency. The input microwave power was varied from 10 to 20 dBm, and the measured DC voltage was proportional to the applied power, suggesting that the induced precession was in the small angle range.
Figure 2. Schematic circuit for the ST-FMR measurement. M and H are the FM magnetization and applied magnetic field vectors, respectively. The $\tau_{\text{el}} + \tau_{\text{M}}$ term denotes the sum of the field-like torque and the Oersted field torque, while $\tau_{\text{el}}$ is the damping-like torque. $J_C$ and $J_S$ are the charge current density and the spin current density, respectively. Top left shows the optical image of the device with contact pads.

Figure 3a–e illustrates the normalized voltages ($V_{\text{mix}}$) measured at 13 GHz for $x = 0.05, 0.13, 0.22, 0.47,$ and $0.61$. The data were normalized to the maximum value of $V_{\text{mix}}$. The gray dashed line indicates the corresponding maximum of $V_{\text{mix}}$. The symmetric and antisymmetric Lorentzian components are presented in Figure 3a–e as the red and blue curves, respectively. The ST-FMR curves are fitted with symmetric and antisymmetric Lorentzian functions, according to

$$V_{\text{mix}} = \frac{V_S}{\pi} \frac{(\Delta H)^2}{(\Delta H)^2 + (H - H_{\text{res}})^2} + \frac{V_A}{\pi} \frac{\Delta H (H - H_{\text{res}})}{\Delta H (H - H_{\text{res}})^2 + (H - H_{\text{res}})^2}$$

(1)

where $\Delta H$ is the full width at half-maximum and $H_{\text{res}}$ is the resonance magnetic field. The symmetric and antisymmetric parameters $V_S$ and $V_A$ are the amplitudes of the symmetric and antisymmetric components of the mixing voltage, respectively. Here, the symmetric component is proportional to the damping-like torque, and the antisymmetric component is due to the sum of the Oersted field torque and the field-like effective torque. The damping-like torque and the field-like torque can be expressed as $m \times (\hat{\sigma} \times \mathbf{m})$ and $m \times \hat{\sigma}$, respectively, where $m$ is the magnetization unit vector and $\hat{\sigma}$ denotes the direction of the injected spin moment. The damping-like torque is generated by the spin current converted from the charge current flowing in the HM layer because of the SHE. As shown in Figure 3f, with increasing concentration of Tb, the magnitude of $V_S/V_A$ increases, which suggests that the spin Hall angle increases with Tb concentration.

According to the ST-FMR theory, the spin Hall angle ($\theta_{\text{SH}}$) is the ratio of the spin current density to the RF current density. The ratio can be obtained from the line shape of the ST-FMR spectra for a qualitative dependence of the $\theta_{\text{SH}}$ by assuming that the field-like torque contribution is negligibly small. For the self-calibrated method, the $\theta_{\text{SH}}$ is given by

$$\theta_{\text{SH}} = \frac{eI_k}{f_{\text{res}}} = \frac{V_S}{V_A} \frac{e\mu_m M_d d}{h} \left(1 + \frac{4\pi M_{\text{eff}}}{H_{\text{res}}} \right)^{1/2}$$

(2)

where $J_S$ is the spin current density generated within the HM, $J_C$ is the applied charge current density, $t$ is the FM layer thickness, $d$ is the HM layer thickness, and $M_{\text{eff}}$ is the effective magnetization, which can be extracted by fitting the resonance frequency $f_{\text{res}}$ as a function of $H_{\text{res}}$ using the Kittel formula,

$$f_{\text{res}} = \frac{e\mu_m M_d d}{\pi h} \left( \frac{\gamma}{2} \right) \left( \frac{H_{\text{res}} + 4\pi M_{\text{eff}}} {H_{\text{res}}} \right)^{1/2}$$

The calculated $\theta_{\text{SH}}$ are shown in Figure 4a. $\theta_{\text{SH}}$ was found to increase with increasing Tb concentration. This is expected, as the SHE arises from SOC and Tb is known to have a much larger SOC compared to Cu. Furthermore, we observed that Cu$_{1-x}$Tb$_x$ with 61% Tb can give rise to $\theta_{\text{SH}}$ of $-0.35 \pm 0.02$, which is larger than the reported value of $-0.30$ found in $\beta$-W. With a further increase in the Tb concentration, the voltage signals become weak, leading to large noise of the spin Hall angle.

To determine the contribution of the extrinsic SHA due to Tb impurities, the extrinsic spin Hall resistivity induced by Tb was calculated and fitted using eq 3. As the intrinsic SHE originates from the effect of SOC on the electronic band structure of hosts when in the dilute regime according to previous reports, it can be assumed that the intrinsic spin Hall resistivity of CuTb is constant at low Tb concentrations. The electrical resistivity $\rho$ of the Cu$_{1-x}$Tb$_x$ layer increases with increasing Tb concentration, as shown in the inset of Figure 4b. The $\rho_{\text{eff}}$ was calculated from

Figure 3. Normalized ST-FMR spectra of CoFeB/Cu$_{1-x}$Tb$_x$ with $x = 0.05$ (a), 0.13 (b), 0.22 (c), 0.47 (d), and 0.61 (e). (f) $V_S/V_A$ as a function of $x$. 

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Figure 4. (a) $\theta_{\text{SH}}$ for different Tb concentrations extracted from ST-FMR spectra. (b) $|\rho_{\text{SH}}|\text{i}$ as a function of resistivity induced by Tb impurities. The inset shows the electrical resistivity of Cu$_{1-x}$Tb$_x$.

Figure 5. (a) FMR linewidths as a function of microwave frequency for different Tb concentrations. Solid lines were obtained by linear fits to the data points. (b) Effective Gilbert damping factor ($\alpha_{\text{eff}}$) extracted from the ST-FMR linewidth as a function of different Tb concentrations.

\[
|1 - \rho_{\text{SH}}^{\text{imp}}| = \rho_{\text{CuTb}}^{-\theta_{\text{SH}}} - (\theta_{\text{SH,Cu}}/\rho_{\text{Cu}})\rho_{\text{CuTb}}^{-2}
= \theta_{\text{SH}}^{-\rho_{\text{imp}}} + \sigma_{\text{SJ}}^{-\rho_{\text{imp}}} \approx \rho_{\text{imp}}^{-\theta_{\text{SH}}}
\]

where $\theta_{\text{SH,Cu}}$ is the spin Hall angle of pure Cu, $\rho_{\text{imp}} = \rho_{\text{CuTb}} - \rho_{\text{Cu}}$ is the resistance induced by Tb impurities, $\theta_{\text{SH}}$ is the contributions of the skew scattering, and $\sigma_{\text{SJ}}$ is the contributions of the side jump.

The extrinsic spin Hall resistivity as a function of $\rho_{\text{imp}}$ is shown in Figure 4b from the fitting, we extract the following values: $\theta_{\text{SH}} = 0.1059$ and $\sigma_{\text{SJ}} = 0.0016 \, \mu \Omega^{-1} \text{cm}^{-1}$. When $\rho_{\text{imp}} = \theta_{\text{SH}}^{-\rho_{\text{imp}}} + \sigma_{\text{SJ}}^{-\rho_{\text{imp}}}$ is the fixed CoFeB thickness of 20 nm and a varied Cu$_{1-x}$Tb$_x$ space. The frequency dependence of $\Delta H$ is summarized in Figure 5a. As Tb concentration increases, significant enhancement was observed in the slope of $\Delta H$. The frequency dependence of $\Delta H$ for all the samples is linear, suggesting a negligible contribution from the two-magnon-scattering mechanisms that would induce a nonlinear trend.

The damping parameter was calculated using eq 5 for different Tb alloy concentrations, as shown in Figure 5b. The $\alpha_{\text{eff}}$ values are in the range from 0.0045 ± 0.0002 to 0.0170 ± 0.0008, which are lower than previous reports. Increasing the Tb concentration leads to significant enhancement of the Gilbert damping parameter, which can be attributed to spin pumping. A spin current is generated from the ferromagnetic layer and diffuses into the nonmagnetic metallic layer when the magnetization precesses. In this process, the ferromagnetic layer loses the spin angular momentum, which results in additional damping. In our study, the damping contribution from the magnetic proximity effect is assumed to be negligibly small as the interfacial effect would be minimal in our relatively thick ferromagnetic layer.

To determine the spin diffusion length ($\lambda_{\text{sd}}$) and the spin-mixing conductance ($G^{(1)}$), a series of samples were fabricated with a fixed CoFeB thickness of 20 nm and a varied Cu$_{1-x}$Tb$_x$ thickness $d$. According to the spin pumping theory, the dependence of the damping on adjacent HM layer thickness can be described by $\Delta H = \Delta H_{\text{inh}} + \Delta H_{\text{Gil}} + \Delta H_{\text{2mag}}$.
Figure 6. (a) Gilbert damping in CoFeB/Cu_{x-Tb} as a function of Cu_{x-Tb} thickness for x = 0.05, 0.13, 0.22, and 0.47. Solid lines are theoretical fit by the spin-pumping theory. (b) Spin diffusion length (λ_{sd}) and spin-mixing conductance (G_{eff}) dependence on Tb concentration in CoFeB/Cu_{x-Tb} films.

\[ \alpha_{\text{eff}} = \alpha_{\text{PM}} + \frac{G_{P}^{1}}{4\pi M_{s}(t - t_{d})} G_{x}^{1} (1 - e^{-2d/\lambda_{sd}}) \]  

(6)

Figure 6a summarizes the damping due to spin pumping with varying thickness. The solid lines are fits for each composition. Damping was found to increase sharply in the lower thickness regime before saturating at a higher thickness. However, this exponential behavior for the enhanced damping as a function of d is inconsistent with the magnetic proximity effect-induced additional damping, which is a quasilinear thickness dependence of damping.\(^{53,54}\) This indicates that there is no evidence that the magnetic proximity effect affects \(\alpha_{\text{eff}}.\) The spin diffusion length deteriorates with increasing Tb concentration because of the strong SOC in Tb impurities that result in spin relaxation.

From the fit for thickness dependence of each Tb concentration, we calculated the diffusion length \(\lambda_{sd}\) as plotted in Figure 6b. The obtained \(\lambda_{sd}\) decreases from 16.8 ± 1.7 nm for Cu_{0.95-Tb} to 2.5 ± 0.3 nm for Cu_{0.53-Tb} with the increase of Tb concentration. The spin diffusion length of Cu_{0.53-Tb} is comparable to that of Pt (\(\lambda_{sd} = 2.8\) nm).\(^{53}\) However, it is much smaller than what was reported by Yue et al.,\(^{41}\) which may be due to the strong scattering caused by Tb impurities in our system, and this is consistent with other reports on HM alloys such as CuIr\(^{56}\) and Au_{1-x}Pt.\(^{55}\) The spin-mixing conductance for the CoFeB/Cu_{0.53-Tb} interface is \((2.4 \pm 1.0) \times 10^{15} \text{ cm}^{-2}\), which is of the same order of magnitude as the Pt/Tb (\(G_{P}^{1}/Tb = 68 \times 10^{15} \text{ cm}^{-2}\)) interface.\(^{41}\) It is to be noted that the voltage signal from the spin-pumping effect (\(V_{SP}\)) has a negligible effect on the symmetric component of the voltage signal because of a large anisotropic magnetoresistance (refer to the Supporting Information).\(^{57}\) Furthermore, a large interface spin transparency (\(T\)) is required for low-energy consumption applications of spin current in multilayered devices.\(^{58,59}\) The interfacial spin transparency can be estimated from the drift-diffusion model\(^{57}\)

\[ T = \frac{G_{P}^{1} \tanh \left( \frac{d}{2\lambda_{sd}} \right)}{G_{x}^{1} \coth \left( \frac{d}{\lambda_{sd}} \right) + \frac{\hbar}{2\pi e G_{x}^{1} C_{eff}}} \]  

(7)

The interface spin transparency of CoFeB/Cu_{0.53-Tb} was estimated to be 0.55 ± 0.03, which is larger than that in CoFeB/β-Ta (0.50\(^{60}\)) and comparable with the value of Co/Pt interfaces (0.3–0.67).\(^{55}\)

4. CONCLUSIONS

In summary, we demonstrated the enhancement of the spin Hall angle by tuning the composition of CuTb alloy thin films. The giant spin Hall angle in the CuTb alloy is due to the strong spin–orbit interaction from the Tb impurities. We found that the contribution of skew scattering dominates side jump at lower Tb concentrations (<14.9%), while the side jump contributions play a significant role at higher Tb concentrations. We also obtained the spin diffusion length and spin-mixing conductance for the CuTb alloy. Furthermore, we extracted the interfacial spin transparency of CoFeB/Cu_{0.53-Tb} as 0.55 ± 0.03, which is comparable with various studied HM/FM interfaces. The giant \(\theta_{SH}\) and high effective spin-mixing conductance with moderate interfacial spin transparency of CoFeB/Cu_{x-Tb} make it a key spin Hall material in highly efficient SOT devices.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsami.0c07441.

XRD patterns; M–H loops; input RF power dependence; inhomogeneous linewidth broadening; spurious voltage signal due to spin pumping; and influence of interfacial antiferromagnetic coupling (PDF)

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Z.X. conceived the idea and designed this work. G.D.H.W. and J.T. assisted in the development of the experimental setup. E.L. and W.L.G. made scientific comments on the results. W.S.L. and F.X. coordinated and supervised the entire work. All authors contributed to the discussion and manuscript writing.

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