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Enhanced spin-orbit torques in Pt/Co/Ta heterostructures

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Spin-orbit torques (SOTs) are studied in perpendicularly magnetized ultrathin Co films sandwiched between two heavy metals, Pt and Ta. A significant enhancement of the Slonczewski-like torque is achieved by placing dissimilar metals with opposite spin Hall angles on opposite sides of the ferromagnet. SOTs were characterized through harmonic measurements and the contribution by the Ta overlayer was isolated by systematically varying its thickness. An effective spin Hall angle of up to 34% is observed, along with a sizable field-like torque that increases with increasing Ta layer thickness. Current-induced switching measurements reveal a corresponding increase in switching efficiency, suggesting that by engineering both interfaces in trilayer structures, the SOTs can be significantly improved. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4902529]

Current-induced torques in ultrathin ferromagnets sandwiched by a heavy metal and an oxide have been of significant recent interest for highly efficient magnetization switching and domain wall motion. In these systems, strong spin-orbit torques (SOTs) can arise through the spin Hall effect (SHE) and the Rashba effect at the heavy-metal/ferromagnet interface, which can be exploited for low-power operation of spintronic memory and logic devices. These effects produce both a Slonczewski-like torque and a field-like torque whose effective field lies parallel to the interface and orthogonal to the current flow direction. The Slonczewski-like torque is similar to the spin-transfer torque in conventional current-perpendicular-to-plane geometries, and is conveniently parameterized by an effective spin Hall angle \( \theta_{\text{SH}} \), which refers to the ratio of out-of-plane spin current to in-plane charge current. Since the efficiency of SOT switching relates directly to the magnitude of \( \theta_{\text{SH}} \), much effort is currently directed at identifying materials and interfaces for which \( \theta_{\text{SH}} \) is large.

Pt and Ta have been the most widely examined spin Hall metals due to their relatively high spin Hall angles and domain wall motion. In these systems, strong spin-orbit torques (SOTs) can arise through the spin Hall effect (SHE) and the Rashba effect at the heavy-metal/ferromagnet interface, which can be exploited for low-power operation of spintronic memory and logic devices. These effects produce both a Slonczewski-like torque and a field-like torque whose effective field lies parallel to the interface and orthogonal to the current flow direction. The Slonczewski-like torque is similar to the spin-transfer torque in conventional current-perpendicular-to-plane geometries, and is conveniently parameterized by an effective spin Hall angle \( \theta_{\text{SH}} \), which refers to the ratio of out-of-plane spin current to in-plane charge current. Since the efficiency of SOT switching relates directly to the magnitude of \( \theta_{\text{SH}} \), much effort is currently directed at identifying materials and interfaces for which \( \theta_{\text{SH}} \) is large.

Pt and Ta have been the most widely examined spin Hall metals due to their relatively high spin Hall angles, \( \sim 0.07 \) for Pt (Refs. 2, 13, and 17) and \( \sim 0.12 \) to \( \sim 0.15 \) for \( \beta \)-Ta (Ref. 3) and their simultaneous utility as underlayers that promote perpendicular magnetic anisotropy (PMA) in thin Co and CoFe(B) films. Recent work has aimed to increase the SHE efficiency by seeking other heavy metals and alloys with larger \( \theta_{\text{SH}} \), with a record of \( +0.3 \) having been reported for beta phase W.\(^4\) To date, most work has focused on heavy-metal/ferromagnet/oxide trilayer structures, where the oxide layer plays the role of breaking inversion symmetry, without actively contributing to the SOTs. One could therefore anticipate that by engineering SOTs at both interfaces, the efficiency of current-induced torques could be further increased using already-known spin Hall materials.

Here, we examine SOTs in ultrathin Co films sandwiched between two heavy metals, Pt and Ta, whose spin Hall angles are of opposite sign. In this case, the spin Hall effect at the top and bottom interfaces are expected to work in concert to enhance the total Slonczewski-like torque. We perform harmonic measurements of the current-induced effective fields and characterize current-induced switching in a series of Pt/Co/Ta stacks with PMA. We find that the effective fields and switching efficiency increase significantly with increasing Ta overlayer thickness, yielding an effective spin Hall angle of up to 0.34, which is the largest value observed to date in a metallic system.

Figure 1 shows the stack structure and corresponding hysteresis loops measured using vibrating sample magnetometry. A series of Ta(4 nm)/Pt(3 nm)/Co(0.9 nm)/Ta(t) tri-layer structures capped by 1.5 nm of TaOx was prepared by dc-magnetron sputtering onto thermally oxidized Si. The nominal thickness \( t_{\text{Ta}} \) of the Ta metal top layer varied from 0.5 nm to 4 nm (Fig. 1(a)). Sputter deposition was performed at room temperature under 3 mTorr Ar at a background pressure of \( \sim 2 \times 10^{-7} \) Torr for the metal layers. The TaOx overlayer was then deposited by reactive sputtering in an oxygen partial pressure of \( \sim 5 \times 10^{-5} \) Torr. These films all exhibited PMA in the as-deposited state (Figs. 1(b) and 1(c)), with saturation magnetization \( M_S \) of 780, 750, 660, and 530 emu/cm\(^3\) for \( t_{\text{Ta}} = 0.5, 1, 2, \) and 4 nm, respectively. The measured \( M_s \) is roughly half the bulk value for Co, which we attribute to Ta intermixing at the Co/Ta interface as discussed below.

X-ray photoelectron spectroscopy (XPS) sputter-depth profiling was performed to extract the depth-dependent material compositions. Here, a thicker Pt layer was used (15 nm) to minimize the contribution of the bottom Ta adhesion layer to the XPS spectra. Figure 2 shows the normalized intensity of Co, O, Pt, and Ta signals extracted by integrating the relative areas of the Ta 4f, Co 2p, Pt 4f, and O 1s peaks in XPS spectra acquired periodically during sputter etching (see supplementary material for spectra and analysis details). All four samples show significant overlap of Co and Ta signals indicating intermixing between Ta and Co, consistent with the reduced \( M_s \). In Fig. 2(a), with \( t_{\text{Ta}} = 0.5 \) nm, the normalized O signal is at \( \sim 20\% \) of its maximum value when the Co signal begins to appear (at \( t = 240 \) s), and the O and Co signals coexist until Co peak disappears, suggesting partial oxidation at the Co/TaOx interface. With increasing \( t_{\text{Ta}} \), a clear separation...
exists between the oxide overlayer and the metallic Co layer, indicating direct contact between Co and metallic Ta for thicker tTa.

We quantified SOTs by detecting current-driven magnetization tilting using harmonic Hall voltage measurements. For these measurements, Hall bar structures were fabricated using electron beam lithography and lift-off, followed by a second lithographic step to deposit Ta(5 nm)/Cu(135 nm) contact pads. In this measurement scheme, the variation of the first and second harmonics of the anomalous Hall voltage with in-plane fields are used to quantify the longitudinal and transverse induced effective fields generated, respectively, by the Slonczewski-like (HSL) and field-like (HFL) SOTs. Figs. 3(a) and 3(b) show schematics of the measurement geometry in each case. An AC injected current generates a periodic torque on the uniformly magnetized Co film, causing the z-component of the magnetization, Mz, to vary at the driving frequency, ω.

The first and second harmonics of the anomalous Hall voltage, Vx and V2x, are then measured, while sweeping either a longitudinal field HL (Fig. 3(a)) or transverse field HT (Fig. 3(b)), to yield HSL and HFL, respectively, through

$$H_{SL(FL)} = \frac{2\left(\frac{dV_{2x}}{dH_L(T)}\right)}{\left(\frac{d^2V_{x}}{dH^2_L(T)}\right)}.$$

Here, we define the effective fields in terms of the direction of electron flow, not the direction of conventional current flow, so that the sign differs from Ref. 28.

Measurements were performed at an excitation frequency of 20 Hz, with the in-plane field swept quasi-statically between ±600 Oe. A small out-of-plane bias field (~50 Oe) was applied during measurement to prevent magnetization switching and domain nucleation. Figs. 3(c) and 3(d) show exemplary Vx and V2x curves versus HL at a current density JAC = 1.8 × 1011 A/m2 for tTa = 0.5 nm, measured both for Mz > 0 and Mz < 0. Figs. 3(e) and 3(f) show

FIG. 1. (a) Schematic Pt/Co/Ta stack structure capped by 1.5 nm of TaOx. The nominal thickness of the Ta top layer varies between 0.5 nm and 4 nm. Normalized out-of-plane (b) and in-plane (c) hysteresis loops were obtained using vibrating sample magnetometry.

FIG. 2. The normalized intensity of Ta, Co, Pt, and O contributions to XPS spectra versus sputter time for tantalum layer thicknesses tTa of (a) 0.5 nm, (b) 1 nm, (c) 2 nm, and (d) 4 nm.

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corresponding data for $H_T$. $V_{2\omega}$ varies linearly with $H_{L(T)}$ in this range, and the slope changes sign with $M_s$ for measurements under $H_T$, but not for $H_L$, as expected for field-like and Slonczewski-like torques, respectively.\textsuperscript{28}

The effective fields $H_{SL}$ and $H_{FL}$, extracted using Eq. (1), are plotted versus $j_{AC}$ in Figs. 3(g) and 3(h) for $t_{Ta} = 0.5$ nm. The effective fields vary linearly with $j_{AC}$, indicating that Joule heating or other artefacts that would cause deviations from linearity are negligible within this current range. Similar results were obtained for samples with other $t_{Ta}$; Fig. 4 shows $H_{SL}$ and $H_{FL}$ per unit current density as a function of $t_{Ta}$, extracted from the slope of $H_{SL(FL)}$ versus $j_{AC}$. We find a relatively large $H_{SL}$ that increases from $\sim 30$ Oe to $\sim 100$ Oe per $10^{12}$ A/m$^2$ as $t_{Ta}$ increases from 0.5 nm to 4 nm. $H_{FL}$, by contrast, is very small at $\sim 5$ Oe per $10^{12}$ A/m$^2$, and is independent of $t_{Ta}$.

Since the measured Hall voltage, in general, contains contributions from both the anomalous Hall effect (AHE) and the planar Hall effect (PHE)\textsuperscript{29–31} Eq. (1) must be amended if these two contributions are comparable\textsuperscript{29–31} since this leads to a mixing of $H_{FL}$ and $H_{SL}$ terms in Eq. (1). We measured both the AHE and PHE resistances ($\Delta R_A$ and $\Delta R_P$; see supplementary material for details about AHE measurements\textsuperscript{32}), and find a ratio $\xi = \Delta R_P/\Delta R_A \sim 0.3$ for all samples. In this case, the PHE correction is not negligible and should be considered.

The results for $H_{FL}$ and $H_{SL}$ after correcting Eq. (1) for the PHE contribution using the expression given in Ref. 30 (see supplementary material for details regarding PHE correction\textsuperscript{32}) are plotted in Fig. 4. We find that the corrected $H_{SL}$ is $\sim 2$ times larger than that extracted without taking the PHE into account, and the corrected $H_{FL}$ is comparable to $H_{SL}$. Intriguingly, we find that the ratio of $H_{SL}$ to $H_{SL}$ is $H_{FL}/H_{SL} \approx 2\xi = 2\Delta R_P/\Delta R_A$, since $H_{FL}$ itself arises almost entirely through the Hall voltage mixing term proportional to $\xi$ in the PHE correction to Eq. (1). This result suggests a relation between SOTs and spin-dependent transport in this system.

Using the PHE-corrected $H_{SL}$, we computed an effective spin Hall angle to compare the strength of the Slonczewski-like torque in the present system with that in systems based on a single spin Hall-active interface. The effective $\theta_{SH}$ was computed from $H_{SL}$ using (Ref. 26) $H_{SL} = \hbar |\theta_{SH}|/e(M_s t_F)$, with $t_F$ is the ferromagnet film thickness, $e$ is the electron charge, and $\hbar$ is the Planck constant. The effective $\theta_{SH}$, plotted in Fig. 4, increases from $+0.09$ to $+0.34$ and begins to saturate, as $t_{Ta}$ increases from 0.5 nm to 4 nm, exceeding the record value of 0.30 for W.\textsuperscript{4} The value of $\theta_{SH}$ for $t_{Ta} = 0.5$ nm is close to that reported for Pt, which is reasonable considering that little if any metallic Ta is likely to remain as a continuous layer at the top interface. As $t_{Ta}$ increases, the effective $\theta_{SH}$ also increases. Since the device resistance did not decrease significantly upon addition of $t_{Ta}$, this suggests that the Ta overlayers include an appreciable fraction of high-resistivity $\beta$-phase Ta, which is known to have a large spin Hall angle.\textsuperscript{3} However, for the thickest $t_{Ta}$ examined, where Fig. 2(d) shows a significant amount of metallic Ta adjacent to Co, the effective $\theta_{SH}$ is considerably larger, exceeding even the sum of $|\theta_{SH}|$ for Pt and $\beta$-Ta, which is expected to be $\sim 0.2$.\textsuperscript{2,3,15,17} We speculate that the presence of Ta within the Co layer and the compositionally graded Co/Ta interface may increase asymmetric spin scattering within the Co layer and/or enhance the spin injection efficiency from the Ta to Co due to the diffuse nature of the interface. However, these points remain to be understood.

Finally, we characterize current-induced switching and extract a measure of the switching efficiency to compare with the effective fields obtained from harmonic SOT measurements. Switching measurements were performed using Hall cross devices, which were 10 $\mu$m long and 2 $\mu$m wide. Here, a longitudinal bias field $H_L$ was applied to tilt the magnetization along the current axis, so that the Slonczewski effective field\textsuperscript{2} $H_{SL} \propto M \times (\mathbf{z} \times \mathbf{j})$ has a projection along the out-of-plane easy axis that can deterministically switch the magnetization.

Switching measurements were performed by first saturating the magnetization along the $-\mathbf{z}$ direction, and then applying a 100 $\mu$s current pulse, after which the magnetization state was measured via the AHE voltage with a small ac sense current.

Fig. 5 shows switching phase diagrams in which the mean normalized $M_s$ after current pulse injection was determined for each pair ($H_{x}$, $j_{pulse}$) from ten measurement cycles. The boundary between non-switching and switching is broad due to current-induced nucleation of a metastable nonuniform magnetization texture as described in detail in Ref. 6.

To estimate the switching efficiency, we note that the Slonczewski effective field has an easy-axis ($\mathbf{z}$-axis) component $H_{z} = H_{SL} \sin \phi$, where $\phi$ denotes the angle between $\mathbf{M}$ and the film plane. The equilibrium angle $\phi$ for a given $H_L$ is given by $\tan \phi = H_L/H_z$, where $H_z$ is the perpendicular anisotropy field. We consider that switching should occur at a current density $j_{crit}$ such that $H_{SL} = H_L(j_{crit})$, where $H_L(j_{crit})$ denotes the coercivity measured in the presence of $H_L$. Defining $H_{FL} = j_{FL}$, we arrive at $\chi = H_L(j_{FL})/j_{crit} \sin \phi$ as a measure of the switching efficiency. $\chi$ was determined for each $t_{Ta}$ by measuring $H_L$ and $j_{crit}$ at $H_L = 1000$ Oe. Due to the broad switching boundary, $j_{crit}$ was taken as the constant density for which the mean $M_s = 0$ after current pulse injection. $H_L$ was determined separately for each device by measuring $V_{AC}(\propto M_z)$ versus $H_L$ and fitting the data to the Stoner-Wolchfarth model in order to compute $\phi$ (see supplementary material for more details\textsuperscript{32}).
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large field-like torque that exceeds 120 Oe per $10^{11}$ A/m$^2$. The large enhancement in the Slonczewski-like torque significantly increases the current-induced switching efficiency. Further improvement could likely be obtained by using spin Hall metal pairs with larger spin Hall angles, e.g., by replacing Ta with W. These results point to significant opportunities to engineer the interfaces of ultrathin transition ferromagnets to enhance SOTs for spintronic device applications.

This work was supported in part by the National Science Foundation under NSF-ECCS-1408172, by C-SPIN, one of the six SRC STARNet Centers, sponsored by MARCO and DARPA, and through the SGMI Program. Technical support from David Bono and Elisabeth Shaw was gratefully acknowledged. Work was performed using instruments in the MIT Nanostructures Laboratory, the Scanning Electron-Beam Lithography facility at the Research Laboratory of Electronics, and the Center for Materials Science and Engineering at MIT.

Figure 5(e) shows $\chi$ versus $t_{Ta}$, which exhibits the same qualitative behavior as $H_{SL}$ in Fig. 4. The switching efficiency increases significantly with the addition of a metallic Ta overlayer, by about a factor of 2 over the range of $t_{Ta}$ examined. Notably, $\chi$ is consistently less than $H_{SL}$ extracted from harmonic measurements in Fig. 4, which increased by a factor of ~4 for the thickest $t_{Ta}$. However, it should be emphasized here that this estimate of $\chi$ is based on equating field-induced and current-induced switching thresholds. Field-driven reversal can occur by domain nucleation anywhere in the Hall bar, whereas current-induced switching must take place within the Hall cross region, so $H_c$ represents a lower bound on the nucleation threshold corresponding to current-induced switching. Moreover, current-induced nucleation occurs at a current lower than $i_{CRM}$, due to the broad switching boundary. Finally, our analysis of the switching efficiency omits the effects of field-like torque and Dzyaloshinskii-Moriya interaction, which can significantly influence the switching process. Nevertheless, these data show that the $H_{SL}$ enhancement through addition of a Ta overlayer leads to a significant qualitative increase in the current-induced switching efficiency.

We have shown that by sandwiching an ultrathin Co film between heavy metals whose spin Hall angle is of opposite sign, the SOTs from each interface work in concert to enhance the total effective torque. In Pt/Co-Ta, an effective spin Hall angle of 0.34 is achieved, together with a large field-like torque that exceeds 120 Oe per $10^{11}$ A/m$^2$. The large enhancement in the Slonczewski-like torque significantly increases the current-induced switching efficiency. Further improvement could likely be obtained by using spin Hall metal pairs with larger spin Hall angles, e.g., by replacing Ta with W. These results point to significant opportunities to engineer the interfaces of ultrathin transition ferromagnets to enhance SOTs for spintronic device applications.

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32See supplementary material at http://dx.doi.org/10.1063/1.4902529 for details on the XPS measurements and analysis, planar Hall effect correction, and switching efficiency analysis.