Interfacial spin transport in (W, Ta)/epitaxial-Co_{60}Fe_{40}/TiN heterostructures: enhancement of switching efficiency

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The giant spin Hall effect reported in industrially viable W and Ta nonmagnetic materials has an immense potential to realize energy efficient spin orbit torque based spintronic devices. However, their use in spintronic heterostructures is subjected to interfacial compatibility with the ferromagnetic layer, requiring low spin memory loss (SML) and high interfacial spin mixing conductance. To shed light on this issue we have performed spin pumping and spin transfer torque measurements on W and Ta interfaced with TiN buffered epitaxial Co_{60}Fe_{40} thin films with varying thickness of the nonmagnetic and magnetic layers. These thin films evident low SML and high interfacial spin-mixing conductance. The significantly high modulation of the effective Gilbert damping of 22.2% for W and 4.4% for Ta interfaced Co_{60}Fe_{40}(t_{CoFe})/TiN structures has been achieved only at an applied dc current density of $J_C = 1 \times 10^9 \frac{A}{m^2}$, which is a result from anti-damping torques from both W(Ta) and TiN interfaces.
INTRODUCTION

Pure spin current based spintronic devices have advantages over conventional microelectronic devices owing to low energy dissipation, fast switching, and high-speed data processing etc., and can be integrated with microelectronic semiconductor devices for better functionality [1-4]. These new spintronic devices mainly work on the principles of the spin manipulation employing the spin Hall effect (SHE) and the Rashba Edelstein effect (REE). The basic building block of spin devices is comprised of ferromagnetic (FM)/ non-magnetic (NM) bilayers, and the NM layers and their interfaces should possess strong relativistic spin-orbit interaction (SOI). The relativistic SOI can be bulk as well as of interfacial nature, and it generates spin orbit torques (SOTs), i.e. damping-like (DL) and field-like (FL) SOTs [4-8]. A charge current applied to SHE and REE based devices generates a transverse spin current and therefore a spin orbit torque at the FM/NM interface, which can be used to manipulate the FM state [4-7]. In contrast, devices based on the inverse effects, the inverse SHE (ISHE) and inverse REE (IREE) convert a spin current generated by spin pumping into a charge current in the NM layer by the ISHE and at the FM/NM interface by the IREE [6-12]. Bulk SOI of the NM layer is responsible for the SHE and ISHE mechanisms [2, 4, 5, 8], while interfacial SOI of the FM/NM interface is responsible for the REE and IREE mechanisms [6, 7, 9, 10]. Recently, a strong DL SOT in bare epitaxial ferromagnetic semiconductor (Ga,Mn)As thin films has been reported, the origin of which lies in the crystal inversion asymmetry induced Berry curvature [7]. Therefore, spin manipulation in the bulk or at the interface plays a decisive role in building the next generation of spintronics devices, viz. spin torque magnetic random access memories (ST-MRAMs), spin logic devices, ST-transistors and ST-nano-oscillators [4]. However, to optimize the bilayer stacks for devices, one needs to control properties like spin backflow, spin memory
loss and magnetic proximity effects that may arise at/near the interface in FM/NM bilayer structures, properties that demean the overall spin transport in the structures [3, 13-19]. Spin backflow arises from non-equilibrium spin accumulation at/near the interface, which depends on the nature of the FM/NM interface and the band mismatch between the FM/NM layers [13-19]. More quantitatively it critically depends on the ratio between the spin-conserved and spin-flip relaxation times $\epsilon$ [3, 13-19]. When $\epsilon \leq 0.001$, which is the case for weak SOI systems, e.g. Ag, Au and $\alpha$-Ta, the NM layer interfaced with a FM layer generates significant spin backflow in spin pumping measurements due to poor spin sink properties. Contrarily, for $\epsilon \geq 0.1$, in case of strong SOI systems, e.g. Pt, the NM layer acts as a perfect spin sink, which results in zero spin backflow [13-19]. However, when $0.001 \leq \epsilon \leq 0.1$, which is likely to be the case for $\beta$-Ta and $\beta$-W, the interface with the FM layers can also create a non-equilibrium spin density accumulation and hence spin backflow during spin pumping [13-19]. It should be noted that the spin current will not dissipate spin angular momentum in the NM layer on length scales shorter than the spin diffusion length ($\lambda_{SD}$) [18]. The SOI values of $\beta$-W and $\beta$-Ta are 0.027 Ry and 0.023 Ry, respectively, comparable to that of Pt (0.030 Ry) [20-21]. However, the reported values of the spin Hall angle, SHA, ($\lambda_{SD}$) vary in the range of 0.12-0.40 (2-4 nm) and 0.006-0.160 (1-7 nm) for $\beta$-W and $\beta$-Ta, respectively [22-31]. It is known that the spin current at the interface also exhibits spin memory loss, in conjunction with spin backflow, in the presence of interfacial SOI, by creating a parallel relaxation path for the spin current [3, 32-35]. Based on the above discussions, it is imperative to investigate how spin backflow and spin memory loss affect the spin current when $\beta$-W and $\beta$-Ta are interfaced with epitaxial FM layers.

In this work, spin pumping measurements were performed on $\beta$-W($\beta$-Ta)/epi-Co$_{60}$Fe$_{40}$/TiN heterostructures to estimate precisely the interfacial spin mixing conductance
Spin memory loss SML, spin diffusion length $\lambda_{SD}$ of $\beta$-W ($\beta$-Ta) layer structures. Strong interfacial damping like torques are evidenced in W (Ta) interfaced epi-CoFe(10nm)/TiN/Si structures, the origin of which lies at both the TiN and W(Ta) interfaces, which have been determined by performing spin transfer torque ferromagnetic resonance (ST-FMR) measurements.

RESULTS AND DISCUSSION

Structural characterization

Co$_{60}$Fe$_{40}$ layers of different thicknesses were deposited on TiN buffered Si(100). Figures 1(a) and (b) show the RHEED pattern recorded along the [001] direction for the TiN(200) film grown on a 2 × 1 reconstructed Si(100) substrate. The evolution of elongated and sharp streaks confirms the 2D-epitaxial quality of the CoFe layers, giving evidence of a CoFe(200)[001]/TiN(200)[001]/Si(400)[001] orientation relationship of these samples. To further confirm the epitaxial growth of CoFe on the TiN buffered Si substrate, texture analyses were performed by measuring X-ray diffraction (XRD) pole figures. Figures 1(c) and (d) show the pole figure XRD patterns of the CoFe(022) plane at 2$\theta$=45.2° for W/epi-CoFe/TiN/Si and Ta/epi-CoFe/TiN/Si thin films confirming the epitaxial quality of the CoFe thin films, which grow on TiN buffered Si (100) substrates. The X-ray reflectivity (XRR) measurements determined thickness of each individual layer is in close agreement with the nominal values predicted from the growth rates; shown in supplementary materials. The interface roughness of each individual layer is in a range of $\leq$ 1 nm, and these values also closely match with previous reports [36, 37]

In-plane FMR
Spin pumping measurements on the (Ta,W)/epi-CoFe series thin films were carried out by performing in-plane FMR measurements, where the magnetic field was applied along the hard axis of magnetization. Figures 2(a) and (b), respectively, show the in-plane FMR spectra of the W(3nm)/epi-CoFe(10nm)/TiN/Si and Ta(5nm)/epi-CoFe(10nm)/TiN/Si thin films at different constant frequencies. From the recorded in-plane FMR spectra, the resonance field $H_r$ and linewidth $\Delta H$ were obtained after fitting the observed spectra with the derivative of Lorentzian functions as shown by solid lines in the figures. Figures 2(c) and (d), respectively, show the observed in-plane $f$ vs. $\mu_0H_r$ data of the W(4nm)/epi-CoFe(10nm)/TiN/Si and Ta(5nm)/epi-CoFe(10nm)/TiN/Si thin films together with fits according to Kittel’s equation (solid lines) [38],

$$f = \frac{\mu_0\gamma}{2\pi} \left[ (H_r + H_a) (H_r + H_b + M_{eff}) \right]^{\frac{1}{2}}.$$  \hspace{1cm} (1)

Here $\gamma=1.80\times10^{11}$ (Hz/T) is the gyromagnetic ratio, $\mu_0M_{eff}$ is the effective magnetization, and $H_a$ and $H_b$ are the in-plane anisotropy fields. Applying the external field along the easy (hard) axis of the film $H_a = H_b$ ($H_b = 0$). The saturation magnetization $M_s$ was determined by fitting the $M_{eff}$ vs. $1/t_{CoFe}$ data using the expression $M_{eff} = M_s - H_s \left( = \frac{2K_s}{\mu_0M_s t_{CoFe}} \right)$, where $H_s$ is the surface/interface induced anisotropy field [39-40]. Figure 2(e) shows the $1/t_{CoFe}$ dependence of $M_{eff}$ for both systems, where $t_{CoFe}$ varies from 3-17 nm. From these fits, the values of $K_s$, and $M_s$ were determined: $K_s = 5.84(\pm0.01)\text{mJ/m}^2$, $\mu_0M_s = 2.46(\pm0.03)\text{T}$, and $K_s = 2.66(\pm0.01)\text{mJ/m}^2$, $\mu_0M_s = 2.40(\pm0.02)\text{T}$ for the W(4nm)/epi-CoFe($t_{CoFe}$)/TiN/Si and Ta(5nm)/epi-CoFe($t_{CoFe}$)/TiN/Si systems, respectively. The slight difference in effective magnetization between the two systems is due to different surface/interface induced anisotropy when using Ta and W as top-layer. The observed value of $\mu_0M_s$ obtained in this way corresponds well with values obtained from magnetometry. However, the possibility of a magnetically dead
layer at the interface, affecting the extracted saturation magnetization, cannot be ignored [35, 41-42]. To investigate the possible existence of a magnetically dead layer, $M$ vs. $H$ measurements were performed on the Ta(5nm)/epi-CoFe($t_{CoFe}$)/TiN/Si and W(4nm)/epi-CoFe($t_{CoFe}$)/TiN/Si series of samples; results of $M_s \times t_{CoFe}$ vs. $t_{CoFe}$ are shown in Fig. 2(f). From the linear fits, the zero intercept of $M_s \times t_{CoFe}$ vs. $t_{CoFe}$ affirms the absence of a dead-layer in these samples [41-42]. Figures 3(a) and (b), respectively, show $\Delta H$ vs. $f$ for the Ta(5nm)/epi-CoFe($t_{CoFe}$)/TiN/Si and W(4nm)/epi-CoFe($t_{CoFe}$)/TiN/Si series of thin film samples. The $\Delta H$ vs. $f$ plots reveal that $\Delta H$ increases linearly with $f$. This confirms that the damping of the magnetization precession is mostly governed by intrinsic Gilbert damping and enhanced damping due to spin pumping. The effective damping constant $\alpha_{eff}$ can be estimated by using the following equation [43]:

$$\mu_0 \Delta H = \mu_0 \Delta H_0 + \frac{4\pi \alpha_{eff} f}{\gamma},$$

(2)

where $\Delta H_0$ represents the magnetic inhomogeneity contribution to damping. In the present case, the observed value of $\Delta H_0$ for all samples fall in the range from a few tenth of mT to ~5 mT (cf. Figs. 3(a) and (b)). The $\alpha_{eff}$ values are plotted in Figs. 3(c) and 3(d) as a function of $1/t_{CoFe}$ and $t_{NM}(NM = W, Ta)$ for the W(4nm),Ta(5nm)/epi-CoFe($t_{CoFe}$)/TiN/Si and W($t_W$),Ta($t_{Ta}$)/epi-CoFe(10nm)/TiN/Si series of thin film samples. In Fig. 3(c), the $\alpha_{eff}$ vs. $1/t_{CoFe}$ plot clearly reveals a linear dependence. The $1/t_{CoFe}$ dependence of $\alpha_{eff}$ in both of these cases evidently signifies the contribution from spin pumping to the damping. An increase of $\alpha_{eff}$ with decreasing $t_{CoFe}$ follows an expression [44-45]:

$$\alpha_{eff}(t_{CoFe}) = \alpha_0 + \frac{\alpha_S(t_{NM})}{t_{CoFe}},$$

(3)
where $\alpha_0$ corresponds to the intrinsic Gilbert damping constant and

$$\alpha_s(t_{NM}) = \left(\frac{\gamma g\mu_B}{4\pi M_s}\right)\gamma_{NM/CoFe}^{\text{eff}}$$

is the enhancement of damping due to spin pumping, $\gamma_{NM/CoFe}^{\text{eff}}$ is the effective spin mixing conductance accounting for interfacial non-equilibrium spin accumulation and spin back flow as well as SML at the interface, $g$ is the Landé spectroscopic splitting factor and $\mu_B$ is the Bohr magneton. From the linear fit, $\alpha_0 = (4.95 \pm 0.23) \times 10^{-3}$ and $(3.54 \pm 0.12) \times 10^{-3}$; and $\alpha_s = (2.48 \pm 0.15) \times 10^{-2}$ nm and $(1.80 \pm 0.11) \times 10^{-2}$ nm, for the W(4nm)/epi-CoFe($t_{CoFe}$)/TiN/Si and Ta(5nm)/epi-CoFe($t_{CoFe}$)/TiN/Si series samples, respectively. The observed difference in the intrinsic values of damping constant of CoFe in contact with W and Ta (in the two series of samples) is attributed to the presence of different extent of two-magnon-scattering (TMS) contributions in respective cases [41]. In Fig. 3(d), the $\alpha_{eff}$ vs. $t_{NM}$ (NM = W, Ta) behavior also indicates the increase in effective damping with respect to $t_{NM}$ ($W, Ta$) in both W($t_W$), and Ta($t_Ta$) capped epi-CoFe(10nm)/TiN/Si systems. This characteristic behavior of $\alpha_{eff}$, despite having different magnitudes, clearly gives evidence of the occurrence of spin pumping in both of these systems. However, a TMS contribution to the FMR linewidth ($\Delta H$) and resonance field ($H_r$) can be expected in in-plane FMR measurements. To avoid the possibility of a TMS contribution to $\Delta H$ out-of-plane FMR measurements were performed on the W(4nm)/epi-CoFe($t_{CoFe}$)/TiN/Si and Ta(5nm)/epi-CoFe($t_{CoFe}$)/TiN/Si series of thin film samples.

**Out-of-plane FMR**

The out-of-plane FMR spectra were obtained by measuring the complex transmission scattering parameter $S_{21}$. During these measurements the VNA was utilized to record the complex transmission parameter $S_{21}$ of the microwave signal at different constant frequencies in
field sweep mode (for more details see Refs. [46, 47]). Figure 4(a) and (b), respectively, show the real and imaginary parts of the out-of-plane FMR spectra for W(4nm)/epi-CoFe(10nm)/TiN/Si and Ta(5nm)/epi-CoFe(10nm)/TiN/Si at two different frequencies. The $\Delta H$ and $H_r$ values at each frequency $f$ were determined by Lorentzian fitting of the observed spectra (red solid lines). Figures 4(c) and (d), respectively, show plots of $f$ vs. $H_r$ and $\Delta H$ vs. $f$ for the W(4nm)/epi-CoFe($t_{CoFe}$)/TiN/Si and Ta(5nm)/epi-CoFe($t_{CoFe}$)/TiN/Si series samples. In Fig. 4(c), the experimental data is fitted with Kittel’s equation for the out-of-plane configuration [38, 46, 47],

$$\frac{2\pi f}{\gamma} = \mu_0 H_r - \mu_0 M_{eff}.$$  \hspace{1cm} (4)

From the fitting, the effective saturation magnetization ($\mu_0 M_{eff}$) is obtained as a function of $t_{CoFe}$. The determined values $\mu_0 M_{eff}$ vary in the range of 1.7–2.30 T for both series of samples. Therefore, $M_s$ can be determined by fitting the $\mu_0 M_{eff}$ vs. $t_{CoFe}$ data (not shown here) using $\mu_0 M_{eff} = \mu_0 M_s - \frac{2K_s}{M_{st_{CoFe}}}$. From the fitting, one obtains $\mu_0 M_s = 2.34(\pm 0.03)$ T and 2.27(±0.01) T for the W(4nm)/epi-CoFe($t_{CoFe}$)/TiN/Si and Ta(5nm)/epi-CoFe($t_{CoFe}$)/TiN/Si series of samples, respectively. Figure 4(d) shows $\Delta H$ vs. $f$ extracted from out-of-plane FMR results and the effective Gilbert damping constant is determined by using Eq. (2). These results again indicate an increase of $\alpha_{eff}$ with decreasing $t_{CoFe}$ obeying Eq. (3). The $1/t_{CoFe}$ dependence of $\alpha_{eff}$ (see Fig. 4(e)) for both series of thin film samples clearly signifies the contribution from spin pumping to the damping of the spin dynamics. From the fits using Eq. (3), $\alpha_0 = (3.50 \pm 0.09) \times 10^{-3}$ and $(3.34 \pm 0.04) \times 10^{-3}$; and $\alpha_s = (2.45 \pm 0.08) \times 10^{-2}$nm and $(7.41 \pm 0.38) \times 10^{-3}$nm was obtained for the W(4nm)/epi-CoFe($t_{CoFe}$)/TiN/Si and Ta(5nm)/epi-CoFe($t_{CoFe}$)/TiN/Si series samples, respectively. The observed values of $\alpha_0$ for for
CoFe are quite close to each other and are in good agreement with values reported in literature [48]. It should be noted that the $\alpha_s$ contribution for W(4nm)/epi-CoFe($t_{CoFe}$)/TiN/Si is two times higher as compared to Ta(5nm)/epi-CoFe($t_{CoFe}$)/TiN/Si, suggesting that spin pumping is more effective in W.

For a detailed study of the spin pumping mechanism it is necessary to investigate how $\alpha_{eff}$ depends on the thickness of the non-magnetic layer, $t_{NM}$; NM=Ta, W. Here $\alpha_{eff}(t_{NM}) = \alpha_0 + \frac{\alpha_s(t_{NM})}{t_{CoFe}}$, where $\alpha_s(t_{NM})$ includes interfacial contributions such as spin back flow [13-19] and SML [32-35]. Figure 4(f) shows $\alpha_{eff}$ vs. $t_{NM}$ for the Ta($t_{Ta}$)/epi-CoFe(10nm)/TiN/Si and W($t_{W}$)/epi-CoFe(10nm)/TiN/Si series of samples obtained from out-of-plane FMR measurements. $\alpha_{eff}$ is relatively higher in W($t_{W}$)/epi-CoFe(10nm)/TiN/Si as compared to Ta($t_{Ta}$)/epi-CoFe(10nm)/TiN/Si. This reveals that with increasing thickness of the W (Ta) layer, $\alpha_{eff}$ increases from $3.50 \pm 0.09 \times 10^{-3}$ ($3.34 \pm 0.04 \times 10^{-3}$) to maximum interfacial value of $6.50 \pm 0.20 \times 10^{-3}$ ($4.30 \pm 0.20 \times 10^{-3}$). This increase of $\alpha_{eff}$ due to spin pumping is weak in the Ta layer as compared to W, which gives evidence that Ta is relatively less a spin sink as compared to W, which is consistent with the SOI strength of materials. The interfacial spin mixing conductance ($g_{NM/CoFe}^{11}$) can be determined as a fitting parameter from the combined fit of $\alpha_{eff}(t_{CoFe})$ vs. $t_{CoFe}$ and $\alpha_{eff}(t_{NM})$ vs. $t_{NM}$, shown in Figs. 4(e) and (f), by using the expression [25, 33, 49]

$$\alpha_{eff}(t_{NM}) = \alpha_0 + \frac{g_{NM/CoFe}^{11}(eff) \mu_B \gamma_{NM/CoFe}^{eff}}{4\pi M_s t_{CoFe}}. \quad (5)$$

It should be noted that in Eq. (5) $g_{NM/CoFe}^{11}(eff)$ at the interface is different on the NM and FM sides due to spin memory loss. For the NM side it can be written as [33, 49]
\[ g_{\text{NM/CoFe}}^{\text{eff}}(t) = (g_{\text{NM/CoFe}}^{\text{eff}}) \left( 1 - \frac{g_{\text{NM/CoFe}}^{\text{eff}}}{g_{\text{NM/CoFe}}^{\text{eff}} + \frac{2}{3} \lambda_{SD} \tanh \left( \frac{f_{\text{NM}}}{\lambda_{SD}} \right)} \right)^{(1 - \delta)}, \]  

(6)

while for the FM side the expression is

\[ g_{\text{NM/CoFe}}^{\text{eff}}(t) = (g_{\text{NM/CoFe}}^{\text{eff}}) \left[ 1 - (1 - \delta)^2 \left( \frac{g_{\text{NM/CoFe}}^{\text{eff}}}{g_{\text{NM/CoFe}}^{\text{eff}} + \frac{2}{3} \lambda_{SD} \tanh \left( \frac{f_{\text{NM}}}{\lambda_{SD}} \right)} \right)^2 \right]. \]  

(7)

In Eqs. (6) and (7) the spin back flow is given by \( \frac{g_{\text{NM/CoFe}}^{\text{eff}}}{g_{\text{NM/CoFe}}^{\text{eff}} + \frac{2}{3} \lambda_{SD} \tanh \left( \frac{f_{\text{NM}}}{\lambda_{SD}} \right)} \) and the SML by \( \delta \).

Here \( \delta = 1(0) \) refers to 100\% (0\%) SML. Self-consistent fitting of the \( \alpha_s(t_{\text{NM}}) \) vs. \( t_{\text{NM}} \) and \( \alpha_s(t_{\text{CoFe}}) \) vs. \( t_{\text{CoFe}} \) data was performed using Eqs. (5-7). The Fermi wave vector \( k_f \) value for W is 15.5 \( \text{nm}^{-1} \) (taken from Ref. [50]), and the \( k_f \) value for Ta is 11.8 \( \text{nm}^{-1} \) (taken from Refs. [24, 25]). The mean free path \( l_{mf} \) for W is 2.2nm and for Ta 3.7nm, which were determined from the thickness dependent resistivity \( \rho_{\text{NM}} \) vs. \( t_{\text{NM}} \) behaviour (not shown here) for W and Ta, respectively. From the best fit \( \delta \), \( \lambda_{SD} \) and \( g_{\text{NM/CoFe}}^{\text{eff}} \) are found to be \( \sim 9\% \) (~2\%), \( 3.20 \pm 0.90\text{nm} \) (6.50±0.75nm) and \( 3.60 \pm 0.2 \times 10^{19} \text{m}^{-2} \) (1.13±0.02×10^{19} \text{m}^{-2}) for W (Ta). Ignoring the presence of spin memory loss, putting \( \delta = 0 \) in Eqs. (6) and (7), the values of \( \lambda_{SD} \) and \( g_{\text{NM/CoFe}}^{\text{eff}} \) become \( 2.16 \pm 0.89\text{nm} \) (4.00±0.71nm), and \( 2.57 \pm 0.15 \times 10^{19} \text{m}^{-2} \) (1.24±0.07×10^{19} \text{m}^{-2}) for W (Ta), respectively, which clearly indicates the importance of including spin memory loss for accurate determination of \( \lambda_{SD} \) and \( g_{\text{NM/CoFe}}^{\text{eff}} \). The obtained values of \( g_{\text{NM/CoFe}}^{\text{eff}} \) for W and Ta, respectively, somewhat smaller than values reported for Pt interfaced FM layers, which can be linked to the presence of the spin back flow effect in W, and Ta based heterostructures [15-18].

The observed \( \lambda_{SD} \) for W is very close to the value (2.0±0.5 nm) reported by Cho et al. [31] but smaller than the value (3.30±0.3 nm) reported for the W/Co_{40}Fe_{40}B_{20} system by Hao et al. [28-29]. The extracted \( \lambda_{SD} \) for Ta is larger than the value of 2.70±0.40 nm reported by Morata et
al.[21], while it lies close to value of 5.10±0.60 nm reported by Yu et al. [25]. The observed decrease of $\alpha_{\text{eff}}$ for all $t_W > 5$ nm is due to loss of a spin coherent state at $t_W > \lambda_{SD}$ [18, 36]. These results are in agreement with the results discussed by Jiao et al., where they observed a larger ISHE signal in bilayers of Ta, Pt and Pd interfaced with Permalloy (Py) in the thickness regime $t_{NM} > \lambda_{SD}$ [18]. After considering the interfacial effects, i.e. spin back flow and SML, it is also of importance to determine the interfacial transparency ($T$) of the FM/NM interface by using the expression [1, 46]

$$T = \frac{g_{NM/CoFe}^{\text{eff}}(\alpha_{\text{eff}}) \tanh\left(\frac{t_{NM}}{\lambda_{SD}}\right)}{g_{NM/CoFe}^{\text{eff}}(\alpha_{\text{eff}}) \coth\left(\frac{t_{NM}}{\lambda_{SD}}\right) \frac{h}{\lambda_{SD} \cdot 2e^2}}$$

(8)

where $\sigma_{NM}$ is the conductivity of W ($3.5 \times 10^5 \Omega^{-1} \text{m}^{-1}$) and Ta ($5.5 \times 10^5 \Omega^{-1} \text{m}^{-1}$) layer, $h$ is the Planck’s constant, and $e$ is the electron charge. Using $g_{NM/CoFe}^{\text{eff}}$ and $\lambda_{SD}$ obtained from fitting the $\alpha_{\text{eff}}$ data to Eqs. (5-7), the calculated values of the interfacial transparency $T$ comes out to be 75% and 40% for $\beta$-W(5nm)/epi-CoFe and $\beta$-Ta(5nm)/epi-CoFe, respectively. The relatively higher value of $T$ for $\beta$-W/epi-CoFe indicates a better band matching between $\beta$-W and epi-CoFe compared to $\beta$-Ta and epi-CoFe.

A. Spin transfer ferromagnetic resonance (STFMR)

To determine the SHA of W and Ta STFMR measurements were performed on patterned (size 20×100 μm²) W(6)/epi-CoFe(10)/TiN/Si and Ta(6)/epi-CoFe(10)/TiN/Si thin films. Schematic figures of the STFMR setup and the thin film structures are shown in Fig. 5(a) (see Ref. [51] for measurement details). At resonance the time varying anisotropic magnetoresistance (AMR) of the FM layer mixes with the applied microwave current $I_{rf}$ producing a dc voltage output. In our
case, using a low-frequency (1 kHz) modulation of \( I_{rf} \) the STFMR signal is detected using a lock-in amplifier. The STFMR spectra were recorded by sweeping the external in-plane dc magnetic field \( H_{dc} \) from high to low value at an angle of 45° with respect to the long axis of micro strip as shown in Figure 5(a). The observed ST-FMR spectra exhibit a combination of symmetric and anti-symmetric Lorentzian weight factors [22-24, 52-55], shown in Figs. 5(b) and (c) along with fits. The STFMR spectrum can be expressed as

\[
V_{mix} = V_0 \left[ SF_S(H) + AF_A(H) \right],
\]

where \( F_S(H) = \left( \frac{\Delta H}{2} \right)^2 \left/ \left[ \left( \frac{\Delta H}{2} \right)^2 + (H - H_r)^2 \right] \right. \) is the symmetric and \( F_A(H) = F_S(H) \times \left( \frac{H-H_r}{\Delta H/2} \right) \) is anti-symmetric Lorentzian function. \( S \) is a symmetric Lorentzian weight factor which accounts for anti-damping SOTs, and \( A \) is an anti-symmetric Lorentzian weight factor which accounts for field-like torques due to the Oersted field and the Rashba effect. \( V_0 \) is the amplitude of the mixing voltage (for details see Ref. [52]), \( S = V_0 \hbar J_S/2e\mu_0 M_S t_{\text{CoFe}}, \) \( A = V_0 H_{rf} \sqrt{1 + \frac{M_{\text{eff}}}{H_r}}, \) \( H_{rf} = \frac{t_{\text{NM}}}{2} J_C(NM); I_{rf} \) generates Oersted \( H_{rf}, J_C(NM) \) is the rf current density in NM layer, and \( \hbar J_S/2e \) is the spin current density generated in the NM layer in units of J/m². The SHA (\( \theta_{SH}^{LS} \)) can be calculated from the expression [52-55]

\[
\theta_{SH}^{LS} = \frac{S}{A} \frac{2e\mu_0 M_S t_{\text{CoFe}}}{\hbar} \frac{t_{\text{NM}}}{2} \sqrt{1 + \frac{M_{\text{eff}}}{H_r}}.
\]

The line-shape parameters \( S, A, H_r \) and \( \Delta H \) were obtained by fitting the STFMR spectra using Eq. (9); results are shown in Fig. 6. The results of \( f \) vs. \( H_r \) and \( \Delta H \) vs. \( f \) are shown in Figs. 6(a) and 6(b), respectively, together with fits employing Eqns. (1) and (2). The fitted values of \( \mu_0 M_{\text{eff}} \) are 2.05±01T and 2.13±0.01T for W(6nm)/epi-Co_{60}Fe_{40}(10nm)/TiN/Si and
Ta(6nm)/epi-CoFe(10nm)/TiN/Si, respectively, which closely match with FMR determined values. The determined values of $\alpha_{\text{eff}}$ are found to be 0.0084±0.0002 and 0.0062±0.0001 for W(6nm)/epi-CoFe(10)/TiN/Si and Ta(6nm)/epi-CoFe(10)/TiN/Si, respectively. Since the intrinsic $\alpha_0$ values of CoFe in both samples are nearly equal (cf. Fig. 4(e)), the higher value of $\alpha_{\text{eff}}$ in W(6nm)/epi-CoFe(10nm)/TiN/Si is due to a larger spin pumping contribution, as discussed in the previous section. The inhomogeneity contribution to damping $\Delta H_0$ is found to be 5mT and 1mT for W(6nm)/epi-CoFe(10nm)/TiN/Si and Ta(6nm)/epi-CoFe(10nm)/TiN/Si, respectively. To estimate the true $\delta_{SH}^S$ values, Eq. (10) must be corrected for the spin pumping contribution, as $\delta_{SH}^S$ only accounts for spin torque contributions in the line-shape. The spin orbit torque contribution (SOT) weight factor in STFMR spectra can be expressed as $\eta = 1 / \left(1 + \frac{V_{\text{ISHE}}}{V_{\text{SYM}}^{\text{STFMR}}} \right)$, where $V_{\text{ISHE}}$ and $V_{\text{SYM}}^{\text{STFMR}}$ are the spin-pumping and STT contributions, respectively, in the STFMR spectrum to total symmetric contribution ($V_{\text{Total}} = V_{\text{ISHE}} + V_{\text{SYM}}^{\text{STFMR}}$). The frequency dependent $\eta$ values are estimated using the method presented in Ref. [51], and subsequently the spin pumping contributions ($V_{\text{ISHE}}$) are subtracted from the STFMR spectra. Spin-pumping corrected as well as with non-corrected values of $\delta_{SH}^S$ at each measured frequency are presented in Fig. 6(c), which clearly reflect the impact of spin-pumping in estimation of the SHA. The calculated values of $|\delta_{SH}^S|$ averaged over all measured frequencies without spin pumping correction is 0.340±0.010 for W and 0.083±0.001 for Ta; interfaces with epi-CoFe(10nm)/TiN/Si. After correction for the spin pumping contribution to the STFMR spectrum, the calculated value of $|\delta_{SH}^S|$ averaged over all measured frequencies is 0.182±0.006 for W and 0.045±0.001 for Ta structures. These observed SHA values are within a range of the reported values for W and Ta based heterostructures [22-31, 52-54]. In order to make a more reliable
estimation of the SHA, one can use the dc current modulation of damping (MOD) method, for which the spin pumping (ISHE) contribution is absent. In the MOD method there is only a rate of change of STFMR linewidth at a given applied frequency, therefore effective damping with respect to the applied dc charge current density in NM layer is required \([52-54]\). The dc current modulation of the effective Gilbert damping \(\alpha_{\text{eff}}(I_{dc})\) is given by \([53, 54]\)

\[
\alpha_{\text{eff}}(I_{dc}) - \alpha_{\text{eff}}(I_{dc} = 0) = \left(\frac{\sin \varphi}{(H_r + 0.5M_{eff})M_0 M_S t_{\text{CoFe}}^2 e} \right) J_S, \quad (11)
\]

where \(J_S = \frac{I_{dc}^{\text{Mod}}}{A_{\text{NM}}} \frac{R_{\text{CoFe/TiN/Si}}}{R_{\text{NM}} + R_{\text{CoFe/TiN/Si}}}\) is the spin current density in the NM layer, \(A_{\text{NM}}\) is the cross sectional area of the NM layer, \(R_{\text{NM}}\) is the resistance of the NM layer; \(R_W = 2384 \, \Omega\), \(R_{Ta} = 1524 \, \Omega\). The resistance of the CoFe(10nm)/TiN/Si stack is \(R_{\text{CoFe/TiN/Si}} = 177.6 \, \Omega\). The dc current flowing in the W (Ta) layer is about 7% (10%) of \(I_{dc}\). These values are close to the value reported by Huang et al. for the dc current flowing in the Ta (6.8% of \(I_{dc}\)) in Ta(5nm)/CoFeB(1.5-4 nm)/Pt(5nm) heterostructures \([56]\). However, Nan et al. \([30]\) reported that 34.5% of the total applied dc current was flowing through the Ta layer in Ta(6nm)/Py(4nm) structures. Since our structures comprise a TiN seed layer with conductivity \(4 \times 10^6 \, \Omega^{-1} m^{-1}\), a significant part of \(I_{dc}\) is expected to go through the CoFe/TiN bilayer. The SHA \(\theta_{SH}^{\text{MOD}}\) can be estimated by measuring the \(I_{dc}\) dependent rate of change of the effective damping; \(\frac{\partial \alpha_{\text{eff}}(I_{dc})}{\partial I_{dc}}\), hence \(\theta_{SH}^{\text{MOD}}\) can be expressed as \([54]\)

\[
\theta_{SH}^{\text{MOD}} = \left[ \frac{\partial \alpha_{\text{eff}}}{\partial I_{dc}} \frac{\sin \varphi}{(H_r + 0.5M_{eff})M_0 M_S t_{\text{CoFe}}^2 e} \right] \times \left( \frac{R_{\text{NM}} + R_{\text{CoFe/TiN/Si}}}{R_{\text{CoFe}}^A} A_C \right). \quad (12)
\]
STFMR spectra were recorded for different constant $I_{dc}$ in the range +5 mA to –5mA on W(6nm)/epi-CoFe(10nm)/TiN/Si and Ta(6nm)/epi-CoFe(6nm)/TiN/Si patterned structures. The recorded spectra were fitted using Eq. (9) to determine the lineshape parameters, and subsequently $\alpha_{eff}(I_{dc})$ was extracted for both the structures; the results are shown in Fig. 6(d). $\alpha_{eff}(I_{dc})$ varies from 0.0024±0.0002 to 0.0138±0.0002 for W(6nm)/epi-CoFe(10nm)/TiN/Si, and from 0.0049±0.0001 to 0.0073±0.0001 for Ta(6nm)/epi-CoFe(10nm)/TiN/Si as $I_{dc}$ is varied from +5 mA to 5mA. To make this data more understandable, the change in linewidth $\Delta H$ and $\alpha_{eff}$ with $I_{dc}$, i.e. $\Delta H(I_{dc}) - \Delta H(I_{dc} = 0)$ and $\alpha_{eff}(I_{dc}) - \alpha_{eff}(I_{dc} = 0)$ vs. $I_{dc}$ for positive external field direction are plotted in Figs. 6(e) and (f). A linear decrease in $\Delta H(I_{dc}) - \Delta H(I_{dc} = 0)$ with increasing $I_{dc}$ for both samples clearly depicts the absence of heating in our measurements. The observed variation in $\alpha_{eff}(I_{dc})$ is larger in W(6nm)/epi-CoFe(10nm)/TiN/Si, which is due to the larger SHA and therefore higher SOT in the W based heterostructure. The percentage modulation of $\alpha_{eff}(I_{dc})$, defined as $(\alpha_{eff}(I_{dc}=0)-\alpha_{eff}(I_{dc})) / \alpha_{eff}(I_{dc}=0) \times 100\%$, at $I_{dc} = \pm 5mA$ is 64% (23%) for the W (Ta)/epi-CoFe(10)/TiN/Si structure; here $I_{dc} = 5mA$ corresponds to $J_C = 2.88 \times 10^9 \frac{A}{m^2}$ ($5.22 \times 10^9 \frac{A}{m^2}$) in W (Ta). The observed modulation value in W/epi-CoFe/TiN/Si is significantly higher than that reported by Pai et al. [22], where a modulation of 25% in the effective damping constant at $J_C = \pm 1.4 \times 10^{11} \frac{A}{m^2}$ was reported for the W(6nm)/Co$_{40}$Fe$_{40}$B$_{20}$(5nm)/SiO$_x$/Si system. The modulation value in Ta/epi-CoFe/TiN/Si is comparable to that reported by Tiwari et al. [57], where a modulation of 30% in the effective damping constant at $J_C = \pm 6.25 \times 10^9 \frac{A}{m^2}$ was reported. Our modulation values are even higher than the Liu et al. [52] achieved modulation of 2.8% at $J_C = \pm 8.95 \times 10^{10} \frac{A}{m^2}$ in Py(4nm)/Pt(6nm) structures. In Table I, a comparison between different literature reported
values and the values of modulation reported here is presented at an applied $f_C = \pm 1 \times 10^9 \frac{A}{m^2}$, which clearly indicates the superiority of our W/epi-CoFe/TiN/Si system. In principle, the calculation of the SHA either by lineshape analysis, using Eq. (10), or $I_{dc}$ dependent damping modulation, using Eq. (12), should be same. If no extra torques are acting on the CoFe layer from the TiN/CoFe interface, then $\theta_{SHL} = \theta_{SHN}$. If $\theta_{SHL}$ is used as input in Eq. (11), the calculated value of the damping modulation is 0.7% (1%) for the W(Ta)/epi-CoFe(10)/TiN/Si structure, i.e. much smaller than the experimentally determined effective damping modulation of 64% (23%). This inconsistency in the calculated and experimentally determined values of the effective damping modulation clearly indicates the presence of additional torques in the W(Ta)/CoFe(10)/TiN/Si structure. Tiwari et al. [57] have also reported a similar inconsistency in the calculated and experimentally determined values of damping modulation in Ta/Py/TiN/Si structures. Recently, Gao et al., [10] have reported the formation of Berry curvature at the interface of CuO$_x$ with Py, which generates antidamping torque at the interface. Moreover, the external field free switching has been demonstrated in FM(4)/Ti(3)/CoFeB(1-1.4)/MgO(1.6) employing interfacial torques [58]. In these structures, the presence of non-negligible interface-generated spin currents stem from FM/Ti and Ti/CoFeB interfaces [58]. Since TiN bulk SOI is very low, its spin diffusion length is 43 nm [59]. Therefore, we anticipate that in our results, the FM/TiN interface plays a significant role in the enhancement of the effective damping modulation, which might originate from Berry curvature induced torques at the interface. However, to conclude with certainty on the origin of the enhanced damping modulation will require detailed theoretical analysis as well as further experimental studies.

In summary, we have performed spin pumping and spin transfer torque experiments on $\beta$-W/epi-CoFe(10nm)/TiN/Si and $\beta$-Ta/epi-CoFe(10nm)/TiN/Si heterostructures using...
ferromagnetic resonance and spin transfer torque ferromagnetic resonance techniques. The FMR induced spin pumping in the W interface is three times larger than the spin pumping in the Ta interface with epi-CoFe/TiN/Si. The STFMR determined spin Hall angle and dc current dependent damping modulation value of W/CoFe(10)/TiN/Si is more than twice the values of Ta/CoFe(10)/TiN/Si. Furthermore, with (without) interfacial parameter corrected values of the interfacial spin-mixing conductance, the spin diffusion length and spin Hall angle are found to be 1.13±0.02×10^{19} \text{m}^{-2}, 6.50±0.75\text{nm} (4.00±0.71\text{nm}), 0.045±0.001 (0.083±0.001) for Ta/CoFe(10)/TiN/Si, and 3.60±02×10^{19} \text{m}^{-2}, 3.20±0.90\text{nm} (2.16±0.89\text{nm}), 0.182±0.006(0.340±0.010) for W/CoFe(10)/TiN/Si. These values clearly demonstrate the superiority of W interfaced epi-CoFe/TiN/Si structures and their potential for energy efficient SOT spin device applications.

**MATERIALS AND METHODS**

Epitaxial Co_{60}Fe_{40} thin films were prepared at 300 °C on TiN(200)[100]/Si substrates by pulsed dc magnetron sputtering technique using 99.99% pure sputtering targets. The sputtering chamber was evacuated to 2×10^{-7} \text{Torr} base pressure and during film growth the Ar gas working pressure was maintained at 3×10^{-3} \text{Torr}. Here epitaxial TiN acts as buffer layer for compensating the lattice mismatch between the Si substrate and the CoFe layer [60, 61]. The growth details of TiN were reported in our previous work [41, 57, 62]. In-situ film growth was monitored by high-pressure reflection high energy electron diffraction (RHEED) (STAIB Instruments) and recorded RHEED patterns were analyzed using the KSA-400 software. After the growth of the epi-CoFe(10nm) layer on buffered TiN(10nm)/Si substrate, layers of W(\text{t}_W) and Ta(\text{t}_T) with different thickness in the range of 1-15 nm were deposited. In another set of films two series of W(4nm)/epi-Co_{60}Fe_{40} (t_{CoFe})/TiN/Si, and Ta(5nm)/epi-Co_{60}Fe_{40} (t_{CoFe})/TiN/Si were grown;
here $t_{CoFe}$ was varied in the range 3-17nm. To investigate the epitaxial quality and crystallographic orientations of the films, X-ray diffraction (XRD) measurements in $\theta$-$2\theta$ and pole figure configurations were performed on all samples using the X’Pert-Pro X-ray diffractometer with Cu-$K_\alpha$ (1.54 Å) radiation. It was also noted that the grown W and Ta layers mostly exhibit the desired $\beta$-phase (i.e. A-15 cubic for W and tetragonal for Ta) as reported in our previous work [36, 37, 41, 57, 62, 63]. The same diffractometer was also used to perform X-ray reflectivity (XRR) measurements on these multilayer thin films. Specular XRR measurements were performed to investigate structural parameters such as thickness, density and interface roughness of each individual layer after simulating the observed XRR spectra, shown in supplementary materials. The saturation magnetization and the possible existence of a magnetically dead layer at the FM/NM interface was determined by performing magnetization vs. field ($M$-vs.-$H$) measurements using a Quantum Design magnetic property measurement system (MPMS). Dynamic magnetic properties were investigated by using broadband in-plane and out-of-plane ferromagnetic resonance (FMR), and spin transfer torque FMR (ST-FMR) measurements in the frequency range of 10-30 GHz, and X-band cavity FMR measurements at the frequency of 9.8 GHz, respectively. In all FMR measurements the magnetic field was swept from high to low value while the frequency was kept constant during the measurement. The X-band cavity FMR setup is equipped with a goniometer to perform in-plane angular-dependent FMR measurements; these results are consistent the cubic anisotropy of the FM films (see supplementary materials). In the in-plane FMR configuration, the resonance field $H_r$ and linewidth $\Delta H$ of the spectrum were determined by using a lock-in-amplifier based FMR technique [46]. The out-of-plane FMR measurements were performed in a Quantum Design physical property measurement system using vector network analyzer (VNA) FMR technique;
magnetic fields up to 4T were employed [47]. To understand the interfacial spin orbit torques in W(6nm)/epi-CoFe(10nm)/TiN/Si and Ta(6nm)/epi-CoFe(10nm)TiN/Si thin films structures, STFMR measurements were performed on patterned (size: \(20 \times 100 \ \mu m^2\)) films in the frequency range of 10-18 GHz. The magnetic field was applied at an angle of 45° with respect to applied microwave current direction in the structure [see Ref. [51] for details of the method]. A dc current in a range of ±5 mA at a step of 1mA was fed through the patterned structure during STFMR measurements.

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**Author’s contributions**

A.K. and P.S. designed and supervised this project. N.B., A.K., V.B., R.B., and R.G., performed the experiments. D.P. and S.C. provided support in the samples fabrication. N.B., A.K., and P.S. analyses the data. N.B and A.K. wrote the manuscript. All authors reviewed and commented on the manuscript.

**Competing interests**

The authors declare that they have no competing interests.

**Materials & correspondence**

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

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Figure 1  The RHEED patterns along [001] of (a) TiN 10nm, and (b) Co$_{60}$Fe$_{40}$ 10nm thin films. (c) and (d) show the pole figure XRD patterns of Co$_{60}$Fe$_{40}$ (022) plane for W/epi-Co$_{60}$Fe$_{40}$/TiN/Si and Ta/epi-Co$_{60}$Fe$_{40}$/TiN/Si thin films, respectively, which confirm the epitaxial quality of the Co$_{60}$Fe$_{40}$ thin films grown on TiN buffered Si (100) substrates.
Figure 2  *In-plane* FMR spectra of (a) W(5nm)/epi-Co$_{60}$Fe$_{40}$(10nm)/TiN/Si and (b) Ta(5nm)/epi-Co$_{60}$Fe$_{40}$(10nm)/TiN/Si thin films at different constant frequencies. (c) and (d) show the *in-plane* $f$ vs. $\mu_0 H_T$ data for W(4nm)/epi-Co$_{60}$Fe$_{40}$($t_{CoFe}$)/TiN/Si and Ta(5nm)/epi-Co$_{60}$Fe$_{40}$($t_{CoFe}$)/TiN/Si, respectively.  
(e) and (f) show $\mu_0 M_{eff}$ vs. $1/t_{CoFe}$ and $\mu_0 M_S \times t_{CoFe}$ vs. $t_{CoFe}$, respectively. Symbols are experimentally observed data and red solid lines are fits to the experimental data. Images shown as insets represent the sample geometries of the thin films.
Figure 3  (a) and (b) $\mu_0 \Delta H$ vs. $f$ for Ta(5nm)/epi-Co$_{60}$Fe$_{40}$($t_{CoFe}$)/TiN/Si and W(4nm)/epi-Co$_{60}$Fe$_{40}$($t_{CoFe}$)/TiN/Si series of thin film samples; experimental results open symbols and linear fits using Eq. (2) solid lines. (c) $\alpha_{eff}$ vs. $1/t_{CoFe}$ (open symbols) and linear fits (solid lines) using Eq. (3). (d) $\alpha_{eff}$ vs. $t_{NM}$ (NM: W, Ta) for Ta($t_{Ta}$)/epi-Co$_{60}$Fe$_{40}$(10nm)/TiN/Si and W($t_{W}$)/epi-Co$_{60}$Fe$_{40}$(10nm)/TiN/Si series of thin film samples.
Figure 4 (a) Real and (b) imaginary parts of the out-of-plane FMR spectra for the W(4nm)/epi-Co_{60}Fe_{40}(10nm)/TiN/Si and Ta(5nm)/epi-Co_{60}Fe_{40}(10nm)/TiN/Si thin film samples at two different frequencies. (c) f vs. $\mu_0H_r$ and (d) $\mu_0\Delta H$ vs. f for W(4nm), Ta(5nm)/epi-Co_{60}Fe_{40}(10nm)/TiN/Si thin films. (e) $\alpha_{eff}$ vs. $1/t_{CoFe}$ and (f) $\alpha_{eff}$ vs. $t_{NM}$ for W(4nm), Ta(5nm)/epi-Co_{60}Fe_{40}(t_{CoFe})/TiN/Si and W(t_{W}), Ta(t_{Ta})/epi-Co_{60}Fe_{40}(10nm)/TiN/Si series samples. The symbols are experimentally observed data and red solid lines are fits to the experimental data as described in the main text.
Figure 5  (a) Schematic of the patterned structure and the ST-FMR setup. (b) and (c) STFMR spectra in the frequency range 10-18 GHz of W(6nm)/epi-Co$_{60}$Fe$_{40}$(10nm)/TiN/Si and Ta(6nm)/epi-Co$_{60}$Fe$_{40}$(10nm)/TiN/Si thin films. The red solid lines are fits to the experimental data using Eq. (9).
Figure 6 (a) $f$ vs. $\mu_0 H_r$, and (b) $\mu_0 \Delta H$ vs. $f$ for the W(6nm)/epi-Co$_{60}$Fe$_{40}$(10nm)/TiN/Si and Ta(6nm)/epi-Co$_{60}$Fe$_{40}$(10nm)/TiN/Si thin film samples at $I_{dc} = 0$ mA. (c) $|\theta_{SH}|$ vs. $f$ determined using Eq. (10). (d) Effective Gilbert damping constant ($\alpha_{eff}$) vs. $I_{dc}$, (e) $\Delta H(I_{dc}) - \Delta H(I_{dc} = 0)$ vs. $I_{dc}$ at 12GHz, and (f) $\alpha_{eff}(I_{dc}) - \alpha_{eff}(I_{dc} = 0)$ vs. $I_{dc}$ for positive applied fields. The red solid lines are fits to the experimental data (see main text for explanations).
| Reports           | Studied heterostructure | % modulation at $10^9$A/m$^2$ |
|------------------|-------------------------|-------------------------------|
| Kasai et al., [58] | Pt(3.5nm)/Py(1.4nm)     | 0.13                          |
| Tiwari et al., [40] | β-Ta/epi-Py(10)/TiN(8)/Si | 4.80                          |
| Liu et al., [57]  | Pt(6nm)/Py(4nm)         | ~0.03                         |
| Ganguly et al., [60] | Co$_{75}$Fe$_{25}$(3.4nm)/Pt(5.8nm) | ~0.03                         |
| Pai et al., [22]  | W(5nm)/CoFeB(6nm)       | ~0.17                         |
| Present work      | W(6)/epi-CoFe(10)/TiN/Si | 22.22                         |
| Present work      | Ta(6)/epi-CoFe(10)/TiN/Si | 4.40                          |

Table 1: Applied dc current modulation of the effective damping of W and Ta interfaced epi-CoFe(10)/TiN/Si compared with literature reported values of high SOC systems.
Supplementary materials for

Interfacial spin transport in (W, Ta)/epitaxial-Co60Fe40/TiN heterostructures: enhancement of switching efficiency

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- Table S2. In-plane angle dependent resonance field i.e. $H_r$ vs. $\phi$ fitting parameters of W,Ta/epi-CoFe/TiN thin films.
Section S1. X-Ray reflectivity (XRR) measurements on W/epi-CoFe/TiN/Si and Ta/epi-CoFe/TiN/Si thin films.

Figures S1(a) and (b) (supplementary material) show the XRR profiles along with their simulated fits for the W/epi-CoFe/TiN/Si and Ta/epi-CoFe/TiN/Si thin films. The simulations have been performed by considering interface layers in-between each of two individual layers. The density, thickness and interface roughness of each individual layer are shown in Table S1. The results reveal the formation of oxide top-layers on the heterostructures; Ta$_2$O$_5$~3nm and WO$_3$~1-2nm in case of Ta/epi-CoFe/TiN/Si and W/epi-CoFe/TiN/Si, respectively. The thickness of each individual layer is in close agreement with the nominal values predicted from the growth rates. The interface roughness of each individual layer is in a range of $\leq 1$ nm.

Section S2. Angle dependent in-plane FMR measurements W/epi-CoFe/TiN/Si and Ta/epi-CoFe/TiN/Si thin films.

In-plane angular-dependent FMR measurements were performed to investigate anisotropic behavior of W/epi-CoFe/TiN/Si and Ta/epi-CoFe/TiN/Si thin films employing the X-band cavity FMR setup which is equipped with a goniometer and able to rotate 360 deg. in both in-plane and out-of-plane direction with respect to applied magnetic field. Figures S2(a) and (b) show the in-plane angle dependent $H_r$ vs. $\phi$ behavior at $f=9.8$ GHz of W(4nm)/epi-CoFe(10nm,6nm)/TiN/Si and Ta(5nm, 6nm)/epi-CoFe(3nm, 10nm)/TiN/Si thin films. It reveals that epitaxial grown CoFe thin films exhibit cubic anisotropy nature. The $H_r(\phi)$ exhibits a variation of $\sim 65.3$ mT and 58.3 mT for W(4nm)/epi-CoFe($l_{CoFe}=10$nm, 4nm)/TiN/Si and 89.4 mT and 69.0 mT for Ta(5nm, 6nm)/epi-CoFe(3nm, 10nm)/TiN/Si thin films, respectively, with respect to easy and hard axis of magnetization. For quantifying the
presence of cubic and uniaxial anisotropies, \( H_r \) vs. \( \phi \) behavior of W/epi-CoFe/TiN/Si and Ta/epi-CoFe/TiN/Si thin films are fitted using expression [46, 62]:

\[
 f = \frac{\mu_0 \eta}{2\pi} \left[ (H_r \cos(\phi_H - \phi_M) + H_c \cos 4(\phi_M - \phi_c) + H_u \cos 2(\phi_M - \phi_u))(H_r \cos(\phi_H - \\
\phi_M) + M_{eff} + \frac{H_c}{4}(3 + \cos 4(\phi_M - \phi_c)) + H_u \cos^2(\phi_M - \phi_u)) \right]^{1/2} \tag{S1}
\]

where \( \phi_H, \phi_M, \phi_c, \phi_u \) are the applied magnetic field, the magnetization, the cubic anisotropy and uniaxial anisotropy directions with respect to Si[100] direction, respectively. \( H_u = \frac{2K_u}{\mu_0 M_s} \) and \( H_c = \frac{2K_c}{\mu_0 M_s} \), respectively, are the uniaxial and cubic anisotropy field, while \( K_u \) and \( K_c \) are the uniaxial and cubic anisotropic constant, respectively. Here \( \mu_0 M_{eff}, H_u, \) and \( H_c \) can be derived as fitting parameter by using Eq. (S1); corresponding fitting parameters values are shown in Table S2. \( H_u \) values of respective epi-CoFe thin films are almost negligible as compare to \( H_c \) values, which confirms that epi-CoFe thin films mostly grown in cubic anisotropy. The fitting determined values of \( \mu_0 M_{eff} \) are closely match with values obtained from the in-plane FMR measurements.
Figure S1 XRR spectra of (a) W/epi-Co$_{60}$Fe$_{40}$/TiN/Si and (b) Ta/epi-Co$_{60}$Fe$_{40}$/TiN/Si thin films. Symbols are experimental data and red solid lines are corresponding simulated fitting to the experimental data.
Figure S2. *In-plane* angle dependent resonance field, i.e. $\mu_0 H_r$ vs. $\varphi$ of (a) W/epi-CoFe/TiN/Si and (b) Ta/epi-CoFe/TiN/Si thin films
| Sample name                                      | Layer | Density $\text{gm/cm}^3$ | Thickness (nm) | Interface width (nm) |
|-------------------------------------------------|-------|--------------------------|----------------|---------------------|
| W(4nm)/Co$_{60}$Fe$_{40}$ (4nm)/TiN(10nm)/Si   | TiN   | 5.2                      | 9.1            | 1.4                 |
|                                                 | Co$_{60}$Fe$_{40}$ | 7.6                      | 4.3            | 0.8                 |
|                                                 | W     | 10.2                     | 2.7            | 0.5                 |
|                                                 | WO$_3$ | 1.4                      | 1.3            | 0.1                 |
| W(4nm)/Co$_{60}$Fe$_{40}$ (10nm)/TiN(10nm)/Si | TiN   | 5.0                      | 9.5            | 0.9                 |
|                                                 | Co$_{60}$Fe$_{40}$ | 7.3                      | 10.1           | 1.4                 |
|                                                 | W     | 12.2                     | 2.5            | 1.2                 |
|                                                 | WO$_3$ | 4.3                      | 1.2            | 1.7                 |
| W(4nm)/Co$_{60}$Fe$_{40}$ (17nm)/TiN(10nm)/Si | TiN   | 4.1                      | 10.7           | 1.1                 |
|                                                 | Co$_{60}$Fe$_{40}$ | 7.8                      | 17.4           | 1.0                 |
|                                                 | W     | 11.7                     | 4.6            | 1.1                 |
|                                                 | WO$_3$ | 0.23                     | 2.0            | 0.3                 |
| W(3nm)/Co$_{60}$Fe$_{40}$ (10nm)/TiN(10nm)/Si  | TiN   | 4.4                      | 10.8           | 0.5                 |
|                                                 | Co$_{60}$Fe$_{40}$ | 7.3                      | 10.1           | 0.2                 |
|                                                 | W     | 14.2                     | 2.3            | 0.9                 |
|                                                 | WO$_3$ | 0.9                      | 1.4            | 0.5                 |
| Ta(5nm)/Co$_{60}$Fe$_{40}$ (3nm)/TiN(10nm)/Si  | TiN   | 5.0                      | 9.8            | 0.8                 |
|                                                 | Co$_{60}$Fe$_{40}$ | 6.4                      | 3.1            | 0.5                 |
|                                                 | Ta    | 14.6                     | 4.5            | 0.2                 |
|                                                 | Ta$_2$O$_5$ | 6.8                      | 3.3            | 0.4                 |
| Ta(5nm)/Co$_{60}$Fe$_{40}$ (10nm)/TiN(10nm)/Si | TiN   | 5.2                      | 10.6           | 0.8                 |
|                                                 | Co$_{60}$Fe$_{40}$ | 8.7                      | 10.1           | 0.2                 |
|                                                 | Ta    | 16.4                     | 5.4            | 0.3                 |
|                                                 | Ta$_2$O$_5$ | 8.0                      | 3.0            | 0.4                 |
| Ta(5nm)/Co$_{60}$Fe$_{40}$ (13nm)/TiN(10nm)/Si | TiN   | 4.9                      | 10.1           | 0.8                 |
|                                                 | Co$_{60}$Fe$_{40}$ | 9.5                      | 13.6           | 0.2                 |
|                                                 | Ta    | 15.5                     | 5.3            | 0.3                 |
|                                                 | Ta$_2$O$_5$ | 8.0                      | 3.2            | 0.4                 |
| Ta(1nm)/Co$_{60}$Fe$_{40}$ (10nm)/TiN(10nm)/Si | TiN   | 4.0                      | 10.6           | 0.5                 |
|                                                 | Co$_{60}$Fe$_{40}$ | 8.2                      | 10.7           | 0.2                 |
|                                                 | Ta    | 15.8                     | 1.1            | 0.5                 |
|                                                 | Ta$_2$O$_5$ | 8.0                      | 3.4            | 0.5                 |

Table S1: XRR fitting parameters; atomic density, thickness and interface width of each individual layer of W,Ta/epi-CoFe/TiN thin films.
| Sample name                                      | $\mu_0M_{\text{eff}}$ (T) | $\mu_0H_c$ (T) | $\mu_0H_a$ (T) |
|-------------------------------------------------|----------------------------|----------------|----------------|
| W(4nm)/Co$_{60}$Fe$_{40}$ (4nm)/TiN(10nm)/Si    | 1.64                       | 0.0265         | $10 \times 10^{-4}$ |
| W(4nm)/Co$_{60}$Fe$_{40}$ (6nm)/TiN(10nm)/Si    | 1.88                       | 0.0327         | $1.2 \times 10^{-4}$ |
| W(4nm)/Co$_{60}$Fe$_{40}$ (10nm)/TiN(10nm)/Si  | 2.12                       | 0.0324         | $0.6 \times 10^{-4}$ |
| W(4nm)/Co$_{60}$Fe$_{40}$ (17nm)/TiN(10nm)/Si  | 2.18                       | 0.0312         | $1.1 \times 10^{-4}$ |
| Ta(5nm)/Co$_{60}$Fe$_{40}$ (3nm)/TiN(10nm)/Si  | 1.60                       | 0.0435         | $1.2 \times 10^{-4}$ |
| Ta(6nm)/Co$_{60}$Fe$_{40}$ (10nm)/TiN(10nm)/Si | 2.14                       | 0.0336         | $0.4 \times 10^{-4}$ |

Table S2 Magnetic anisotropic parameters of W/epi-CoFe/TiN/Si and Ta/epi-CoFe/TiN/Si thin films system determined from the fittings of in-plane $H_r$ vs. $\mu_0H$ using Eq. S1.