Anomalous spin orbit torques with large Rashba spin orbit coupling in epitaxial Pt/Co bilayers

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Abstract

The Rashba-Edelstein effect (REE), which characterizes the generation of a transverse spin polarization at interfaces with a longitudinal external electrical field, provides new opportunities to the efficient spin current generation and the manipulation of spin orbit torques (SOTs) in magnetic hetero-structures. Here in this work, we report an exceptionally large interfacial charge-to-spin conversion efficiency (i.e. the inverse Edelstein length \( \lambda_{\text{IEE}} \) of a few nanometers) in epitaxial (epi-) Pt/Co bilayers from a REE-induced magnetoresistance analysis using a new drift-diffusion model that incorporates both REE and bulk spin Hall effect. From the spin torque ferromagnetic resonance analysis, a tenfold enhancement in the field-like (FL) SOT efficiency is demonstrated compared to polycrystalline samples, showing quantitative consistency with the large \( \lambda_{\text{IEE}} \). Additionally, a twofold increase in the damping-like (DL) SOT efficiency is observed in epi-samples, with Pt films having lower resistivity. Our study demonstrates that both DL- and FL-SOTs can be significantly modulated by the Rashba SO coupling with hetero-interface modification, providing new perspectives to the engineering of SOTs in spintronic devices.
The generation of a transverse spin current from a longitudinal charge current by taking advantage of the giant spin Hall effect (SHE)\(^1\)–\(^3\) in materials with strong spin orbit coupling (SOC) facilitates the manipulation of local magnetizations via electrical means.\(^4\) Sufficiently large spin orbit torques (SOTs) derived from the SHE are key to realizing energy-efficient applications such as magnetization switching\(^4\),\(^5\) and fast domain wall motion.\(^6\),\(^7\) The giant SHE in heavy metals originates from their predicted large spin Hall conductivity through first-principle calculations,\(^8\),\(^9\) and allows the transfer of spin angular momentum to neighboring magnetizations in spin-orbit/ferromagnetic (SO/FM) bilayers, enabling phenomena such as the spin-torque ferromagnetic resonance (ST-FMR)\(^10\) and spin Hall magnetoresistance (SMR).\(^11\) While the SHE converts charge current to spin current within the bulk of the SO materials, the Rashba-Edelstein effect (REE)\(^12\),\(^13\) realizes it in reduced dimensions, that is, at interfaces. In particular, the REE describes a lift of the spin degeneracy at interfaces due to the structural inversion asymmetry,\(^12\) and consequently contributing to the generation of nonzero interfacial spin accumulation induced by the SO interactions with an external charge current.\(^13\) Recently, the inverse REE has been experimentally demonstrated by means of spin pumping in 2-dimentional electron systems (2DES) of metallic hetero-structures\(^14\) and gate-controllable\(^15\) oxide interfaces.\(^16\),\(^17\)

Although the mechanism of REE is intrinsically different from that of SHE, the ultimate goal of these two effects, \textit{i.e.} to exert SOTs on the neighboring ferromagnetic (FM) layer via spin angular momentum transfer, appears to be quite similar. In terms of physical interpretation, the origin of the SOTs can be understood by the Laudau-Lifshitz-Gilbert equation for the SOT-induced magnetization dynamics:

\[
\frac{d\bar{m}}{dt} = -\gamma \bar{m} \times \vec{H}_{\text{eff}} + \alpha \bar{m} \times \frac{d\bar{m}}{dt} + \gamma \frac{\hbar J_c}{2e\mu_0 M_s t_F} \bar{m} \times (\xi_{DL}\vec{y} \times \bar{m} + \xi_{FL}\vec{y}) - \gamma \bar{m} \times \vec{H}_{\text{rf}}
\]

where \(\gamma\) is the gyromagnetic ratio, \(\bar{m}\) is the magnetic moment vector, \(\vec{H}_{\text{eff}}\) is the effective magnetic field that contains external and demagnetization field, \(\alpha\) is the effective damping constant, \(J_c\) is the applied charge current density in Pt, \(e\) is the electron charge, \(\mu_0 M_s\) and \(t_F\) are
the saturation magnetization and thickness of the ferromagnetic material, respectively, \( \vec{y} \) is the unit vector which is perpendicular to both current flow and thin film normal, \( \xi_{DL(FL)} \) is the damping-like (field-like) SOT efficiency, respectively, and \( \vec{H}_{rf} \) is the current-induced Oersted field. Based on the above equation, the DL- and FL-SOT vectors are orthogonal to each other.

In recent years, intense efforts have been made to quantitatively determine \( \xi_{DL} \), partly owing to practical implications of DL-SOTs on deterministic magnetic switching.\(^4\)\(^,\)\(^5\)\(^,\)\(^18\)\(^,\)\(^19\) Generally, the DL-SOTs are mainly considered to result from the bulk SHE, therefore in the SHE scenario \( \xi_{DL} \) is expected to scale with the spin Hall angle \( \theta_{SH} \) of SO materials. Recent reports have found an enhancement in \( \xi_{DL} \) by increasing the resistivity of the SO materials (\( \rho_N \)) through either oxygen doping\(^20\) or incorporation of scattering sites by defects or disorder.\(^21\)\(^,\)\(^22\) This can be well understood by an intrinsic or a side-jump contribution to the bulk SHE.\(^3\) On the other hand, interfacial SOC such as the REE could also be an additional source for the DL-SOTs, as proposed in recent theories\(^23\)\(^,\)\(^24\) and suggested by experiments.\(^25\)\(^,\)\(^20\)\(^,\)\(^26\) However, explicit experimental evidence for a significant control of the SOTs induced by the REE, is lacking. Here in this work, we report large DL- and FL-SOTs stemming solely from the large REE in epitaxial (epi-) Pt/Co bilayers: a large inverse Edelstein length, \( i.e. \lambda_{IEE} \),\(^14\)\(^,\)\(^16\) of a few nanometers is obtained from a temperature (\( T \)) dependent REE-induced magnetoresistance with a drift-diffusion model developed in this work that includes both REE and SHE. Such an anomalously large \( \lambda_{IEE} \) is quantitatively consistent with a tenfold increase in \( \xi_{FL} \) in epi-samples compared to that of conventional polycrystalline (poly-) ones; additionally, a twofold increase in \( \xi_{DL} \) is found in epi-samples, with thin films having lower resistivity, suggesting that such \( \xi_{DL} \) enhancement is mainly due to the large Rashba SOC at Pt/Co interface. Our study has shown that the Rashba SOC can profoundly manipulate the SOTs in magnetic hetero-structures, providing new strategies for the engineering of SOTs in spintronic devices.
Sample preparations

Multilayer thin films of Pt($t_N$/Co($t_F$/Al-O(2)) with thicknesses $t_N = 0.2$-10 nm and $t_F = 2.5$-10.2 nm were deposited onto Al$_2$O$_3$ (0001) and Si/SiO$_2$ substrates at room temperature (RT) using magnetron sputtering facility with base pressure better than $2 \times 10^{-7}$ Pa to fabricate epi- and poly- samples, respectively. Poly-samples are mainly used as control samples to compare with previous SOT studies. Reflection high-energy electron diffraction (RHEED) patterns and X-ray diffraction (XRD) results confirm epitaxial growth of Pt/Co bilayers upon Al$_2$O$_3$ (0001) substrate (Fig. 1 (a) and (b)). The average roughness ($R_a$) of the Pt/Co interface for epi-samples is determined to be below 0.1 nm from the X-ray reflectivity measurement (Fig. 1 (c)), demonstrating interfacial flatness at the atomic scale. In contrast, $R_a$ for poly-Pt/Co interface is above 0.3 nm. More details about thin film deposition and device fabrication are described in Methods.

Spin-Hall Rashba-Edelstein magneto-resistance in epitaxial Pt/Co bilayers

As an electrical current flows through the SO/FM bilayers, the longitudinal resistance ($R_{xx}$) has been found to change with respect to the FM layer magnetization orientation. Such a resistance change, known as the spin Hall magneto-resistance (SMR),$^{11,27,28}$ is related to spin transport across the interface, i.e. transmission and/or reflection of spin current generated in the SO layer. The magnitude of the SMR ratio scales with the square of the $\theta_{SH}$ and depends on the ratio of thickness to the spin diffusion length of the SO material. In this study, the definition of the coordinate system is shown in Fig. 2 (a). Figures 2 (c) and (d) show the magnetic field dependence of $R_{xx}$ for poly- and epi-samples with magnetic field applied along the $x$, $y$ and $z$ directions at RT. The SMR ratio is defined as

$$\frac{\Delta R_{xx}^{SMR}}{R_{xx}^0} = \frac{R_{xx}(H_y) - R_{xx}(H_z)}{R_{xx}(H_z)}$$
where \( R_x(H_x) \) and \( R_x(H_z) \) refers to the longitudinal resistance when the magnetization is saturated with a large field that is applied in the \( y \) and \( z \) directions. Using a parallel channel model, we subtract parasitic MR coming from the Co layer induced by the geometrical size effect,\(^{29}\) by performing additional MR measurement for a single Co layer (see supplementary materials). Note that by comparing Fig. 2 (c) and (d), the resistance difference between \( R_{xx}(H_x) \) and \( R_{xx}(H_z) \) is much larger in poly-samples than that in epi-samples. This is likely due to a larger anisotropic magnetoresistance\(^{30}\) in poly-Co layer. As shown in Fig. 2 (f) and (g), the plot of SMR ratio as a function of \( t_N \) is used to estimate \( \theta_{SH} \) and the effective spin diffusion length \( \lambda_{sf} \) of Pt via the following equation:\(^{11,27,28}\)

\[
\frac{\Delta R_{xx}^{SMR}}{R_{xx}^0} = -\frac{\lambda_{sf}}{t_N} \frac{\lambda_{sf}}{\lambda_{sf}} \tanh\left(\frac{t_N}{2\lambda_{sf}}\right) \left[ 1 - \frac{1}{\cosh\left(\frac{t_N}{\lambda_{sf}}\right)} \right] \tag{1}
\]

where \( \eta = \rho_N t_F / \rho_F t_N \) is the current shunting coefficient. Here we assume a transparent Pt/Co interface and neglect the longitudinal spin absorption because \( \lambda_{sf} \) of Co is much longer compared to \( t_F \) in this work\(^{31}\). In Fig. 2 (f), we fit the \( t_N \) dependence of the MR ratio with Eq. (1) and obtain \( \theta_{SH} = 0.19 \) and \( \lambda_{sf} = 0.93 \) nm for the poly-samples, in good agreement with earlier reports (e.g. \( \theta_{SH} = 0.12-0.16 \) and \( \lambda_{sf} = 1.0-3.0 \) nm),\(^{32-34}\) suggesting that the SMR model is applicable to spin transport in poly-samples. For epi-samples, surprisingly, a large \( \theta_{SH} \) of 0.33 and an extremely short \( \lambda_{sf} \) of 0.35 nm is obtained from the fitting of Fig. 2 (g). Such an abnormally short \( \lambda_{sf} \) strongly suggest that the SMR model, which originates from spin current generation in bulk, is no longer valid for analyzing the MR results of epi-samples: firstly, \( \rho_N \) for epi-Pt is lower compared to poly-films (Fig. 2 (h)). It follows from the Elliot-Yafet (EY) spin relaxation mechanism\(^{35,36}\) that \( \lambda_{sf} \) scales with \( 1/\rho_N \) in metallic films such as Pt.\(^{22}\) Therefore, epi-Pt is expected to have longer \( \lambda_{sf} \) and a SMR peak position at larger \( t_N \) (e.g. > 2.5 nm in Fig. 2 (f)) compared to the poly-samples in the SMR model. However, this is not the case for the results of epi-samples. Consequently, one has to consider the D’yakonov-Perel’ (DP) mechanism\(^1\) rather than the EY mechanism (SMR model) that results in an extremely small \( \lambda_{sf} \). Such a small \( \lambda_{sf} \) is
not consistent with any reports so far\textsuperscript{10,22,33} and is therefore very unlikely. Note that an earlier work\textsuperscript{37} in our group has demonstrated that the DP mechanism is dominant in epi-Pt grown on MgO (111) substrates and its origin lies in the interface Rashba SOC. Secondly, the $T$ dependence of the MR (Fig. 2 (g)) shows almost no peak shift in the whole $T$ range, though $\rho_N$ decreases by half as $T$ decreases from 300 K to 4.2 K. According to the EY mechanism, there should be a 2-time difference in the values of $\lambda_{sf}$ between 300 K and 4.2 K, which causes a shift in the SMR peak (e.g. more than 1 nm) for epi-samples in the SMR model. That, again, is contradictory to what we observe in Fig. 2 (g). We therefore conclude that SHE due to the EY mechanism in Pt is not a dominant mechanism for spin current generation in our epi-samples. Instead, we consider the REE and its inverse effect,\textsuperscript{26,38} which accomplishes the charge-to-spin current inter-conversion at Pt/Co interfaces,\textsuperscript{39} to be the dominant origin for the MR in epi-samples, which we term it REMR hereafter (following Ref. 26).

In order to analyze the $t_N$ dependence of the REMR, a drift-diffusion model is developed by incorporating an Edelstein contribution $e(\lambda_{IEE} \hat{z} \times \mathbf{E})$ of spin accumulation\textsuperscript{40,41} into the SMR theory\textsuperscript{11,27,28}. The hybrid MR including both REE and SHE reads:

$$\frac{\Delta R_{xx}}{R_0} \approx - \frac{\lambda_{sf}/\rho_N}{t_N/\rho_N + t_F/\rho_F} \text{Re} \left[ \frac{2\rho_N \lambda_{sf} G_{\uparrow \downarrow}}{1 + 2\rho_N \lambda_{sf} G_{\uparrow \downarrow} \coth(t_N/\lambda_{sf})} \right]$$

$$\times \left\{ \left[ \theta_{SH} \tanh \frac{t_N}{2 \lambda_{sf}} + \frac{\lambda_{IEE}}{2 \lambda_{sf}} \right]^2 \right. $$

$$- 2 \left( \frac{\lambda_{IEE}}{2 \lambda_{sf}} \right) \left[ \theta_{SH} \tanh \frac{t_N}{2 \lambda_{sf}} + \frac{\lambda_{IEE}}{2 \lambda_{sf}} \right] \frac{\sinh[(t_N - d_z)/\lambda_{sf}]}{\sinh(t_N/\lambda_{sf})}$$

$$+ \left( \frac{\lambda_{IEE}}{2 \lambda_{sf}} \right)^2 \frac{\sinh^2[(t_N - d_z)/\lambda_{sf}]}{\sinh^2(t_N/\lambda_{sf})} \right\}$$

(2)

where $\lambda_{IEE}$ has a unit of length that characterizes the distance of the spin-momentum locking,\textsuperscript{14,16} $d_z$ is the thickness of the RE region, $G_{\uparrow \downarrow}$ is the spin mixing conductance at the Pt/Co interface,\textsuperscript{42} and $\langle \cdot \rangle$ denotes the average with respect to $d_z$ weighted by $\exp(-d_z/d_R)$, where $d_R$ is the effective thickness of the RE region (see supplementary materials for details). To extract $\lambda_{IEE}$ and
from the experimental data, we first predefine $\theta_{SH}$ and $\lambda_{sf}$ for bulk epi-Pt. Assuming the EY mechanism and intrinsic/side-jump contribution to the SHE in epi-Pt, $\lambda_{sf}\rho_N$ is set to be in the range of $(0.36-0.69)\times10^{-15}$ $\Omega m^2$ (e.g. a value of $\sim0.6\times10^{-15}$ $\Omega m^2$ is suggested for Pt from both theoretical calculation$^{43}$ and experimental analysis$^{22}$) with a fixed $\lambda_{sf}\theta_{SH} = 0.2$ nm,$^{44,45,16}$ which corresponds to a $\theta_{SH}$ ($\lambda_{sf}$) ranging from 0.06 to 0.12 (1.7 to 3.3 nm), consistent with previous SHE studies for Pt.$^{22}$ The $G_{11}$ is considered to depend only on the band dispersion$^{46}$ at Pt/Co interface and its real part is assumed to be $10^{15} \Omega^1 m^{-2}$,$^{47,34}$ independent of $T$. Thus we obtain $\lambda_{IEE}^{eff}$ and $d_{R}^{eff}$ by fitting the experimental data in Fig. 2 (g), Fig. 3 (a) and (b) using Eq. (2) with different $t_p$. The resulting $\lambda_{IEE}^{eff}$ and $d_{R}^{eff}$ are shown as a function of $t_p$ in Figs. 3 (e) and (f). The $\lambda_{IEE}$ is obtained by fitting the $\lambda_{IEE}^{eff}$-$t_p$ plot with an empirical formula $\lambda_{IEE} = \lambda_{IEE}^{eff}[1 - \text{sech}(t_p/l_{dp})]$, where $l_{dp}$ is the spin dephasing length in Co, yielding a large $\lambda_{IEE}$ of a few nanometers (2.5 nm at 300 K and 5.3 nm at 4.2 K), which is about one order of magnitude greater than that in heavy metals (e.g. 0.2 nm for Pt and 0.43 nm for W),$^{16}$ and is comparable to that of an earlier study for the LaAlO$_3$/SrTiO$_3$ 2DES,$^{16}$ demonstrating exceptionally efficient interfacial charge-to-spin conversion. Moreover, the Rashba parameter ($\alpha_R$) is extracted from the relation $\alpha_R = h\lambda_{IEE}/\tau_p$, where $\tau_p$ is the momentum scattering time. Assuming a carrier density of $10^{28}$ m$^{-3}$ in Pt, we obtain a $T$-independent value of $\alpha_R$ in the order of $10^{-10}$ eVm (Fig. 4), providing a low bound of the Rashba SOC strength.

**Spin torque ferromagnetic resonance measurement**

From the REMR analysis, a large $\lambda_{IEE}$ of a few nanometers is obtained, suggesting efficient charge-to-spin current conversion originated from the Rashba SOC. In this section we study its influence on the SOTs by performing the spin torque ferromagnetic resonance (ST-FMR) measurement,$^{10}$ i.e. to quantitatively investigate the $\xi_{DL}$ and $\xi_{FL}$ in our samples. Figure 5 shows the experimental set-up with an external magnetic field ($H_{ext}$) applied at 45º to the micro-strip and
a RF microwave current applied to the ST-FMR devices with a conventional bias tee configuration. The output $V_{\text{mix}}$ is the d.c. voltage from mixing the RF current and the resultant oscillating device resistance via the oscillating anisotropic MR, and is expressed as follows:

$$V_{\text{mix}} = k \left[ \frac{S}{\Delta H^2 + (H_{\text{ext}} - H_R)^2} A \left( \frac{\Delta H (H_{\text{ext}} - H_R)}{\Delta H^2 + (H_{\text{ext}} - H_R)^2} \right) \right]$$  \hspace{1cm} (3)

where $k$ is a constant, $\Delta H$ is the half linewidth of the resonance spectrum, $H_R$ is the resonance field, $S$ and $A$ are the symmetric and anti-symmetric component of the ST-FMR resonance.

Typical ST-FMR spectra and the their fitting curves using Eq. (3) are shown in Fig. 5 (b) and (c), with the respective stacking structure of (b) poly-Pt(10)/Co(4.0) and (c) epi-Pt(10)/Co(3.0). The effective spin torque efficiency $\xi_{\text{ST}}$ is given by

$$\xi_{\text{ST}} = \frac{S e \mu_0 M_{\text{eff}} t_F t_N}{\hbar} (1 + \mu_0 M_{\text{eff}}/\mu_0 H_R)^{1/2}$$  \hspace{1cm} (4)

where $e$ is the electron charge, $M_{\text{eff}}$ is the effective demagnetization field, obtained by fitting the frequency as a function of the $H_R$ using the Kittel formula $f_R = (\gamma/2\pi) [\mu_0 H_R (\mu_0 H_R + \mu_0 M_{\text{eff}})]^{1/2}$. Figure 5 (d) and (e) show the calculated $\xi_{\text{ST}}$ based on the fitting results of Fig. 5 (b) and (c). Parasitic effects such as the spin pumping are estimated to be less than 5% in magnitude compared to the genuine ST-FMR signal (see supplementary materials). For the two samples above, $\xi_{\text{ST}}$ is estimated to be 0.15 and 0.1 for poly- and epi-films, respectively.

In the case that the FL-SOTs are negligible in the SO/FM bilayers, the $\xi_{\text{ST}}$ can be regarded as the $\xi_{\text{DL}}$, providing an estimation of the effective SOTs absorbed by the FM layer. However, when there exists considerable FL-SOTs in the system, they contribute to the anti-symmetric component ($A$) of the ST-FMR signal, thus resulting in incorrect estimation of both DL- and FL-SOTs. To separate the contributions of DL- and FL-SOTs to the overall SOTs and obtain the intrinsic $\xi_{\text{DL}}$ and $\xi_{\text{FL}}$, we performed the ST-FMR measurement for epi- and poly-samples with varying $t_F$ from 2.5 to 10 nm ($t_N$ is fixed to be 10 nm). Typical ST-FMR spectra with different $t_F$ for poly- and epi-samples are shown in Fig. 6 (a) and (b), respectively. Both $\xi_{\text{DL}}$ and $\xi_{\text{FL}}$ can be quantitatively determined using the following equation:
\[
\frac{1}{\xi_{ST}} = \frac{1}{\xi_{DL}} \left( 1 + \frac{\hbar}{e \mu_0 M_s t_F t_N} \right) \tag{5}
\]

where \(\xi_{DL}\) and \(\xi_{FL}\) are determined from the y-axis intercept and slope of the \(1/\xi_{ST} - 1/t_F\) linear fit, as shown in Fig. 6 (d). Note that as the \(t_F\) increases to more than 5 nm, the \(\xi_{ST}\) starts to deviate from the linear fitting, this is likely due to a phase change in Co, which was reported previously.\(^{34}\)

As a result, the average \(\xi_{DL}\) and \(\xi_{FL}\) for poly-samples turn out to be 0.16 and 0.015, consistent with previous reports for the Pt/Co bilayers.\(^{32-34}\) For epi-samples, both SOTs are enhanced: \(\xi_{DL}\) is determined to be 0.34 and more strikingly, \(\xi_{FL}\) is found to be 0.2, showing a tenfold increase compared to the poly-samples. Such large \(\xi_{FL}\) enhancement is another evidence for the enhanced Rashba SOC at the Pt/Co interface.\(^{24,47}\) Moreover, the \(\lambda_{IEE}\) in poly-samples is estimated to be one order of magnitude smaller (see supplementary materials), showing quantitative consistency regarding the interfacial charge-to-spin efficiency obtained independently from the REMR and ST-FMR measurement.

Based on the analysis of the REMR and ST-FMR results, a good agreement is attained that both the anomalously large \(\lambda_{IEE}\) and a tenfold enhancement in \(\xi_{FL}\) for epi-samples are attributed to the large Rashba SOC at Pt/Co interface. The remaining question would be: how to explain the nearly twofold increase in \(\xi_{DL}\) for epi-samples in which Pt and Co have smaller resistivity compared to poly-ones. Generally, the DL-SOTs are considered to mainly arise from the bulk SHE through the relation of \(\xi_{DL} = \theta_{SH} T_{int}\), where \(T_{int}\) is the so-called spin interface transparency.\(^{27,33,34}\) If DL-SOTs in poly-samples stem only from the SHE, the \(\theta_{SH}\) for poly-samples at RT is estimated to be 0.25 with \(T_{int}\) of 0.65,\(^{33}\) consistent with recent reports of poly-Pt/Co bilayers.\(^{33,34}\) By assuming an intrinsic contribution from the SHE (i.e. \(\theta_{SH} \propto \rho_N\)) for Pt,\(^{22,49}\) we estimate that in epi-samples, \(\xi_{DL}\) induced from the SHE ranges from 0.13 to 0.19 by taking a \(T_{int}\) from 0.65 to 1, with the averaging value to be 0.16. Thus, even by assuming a \(T_{int}\) close to unity at epi-Pt/Co interface, the \(\xi_{DL}\) originated from SHE alone is still much smaller (about half) than the value obtained from the ST-FMR measurement. Consequently, we conclude that the SHE
cannot be the mechanism that accounts for the 2 times enhancement in $\xi_{DL}$ for epi-samples compared to that of poly-ones, meaning that the other critical mechanism in SO system, \textit{i.e.} the Rashba effect, must play a critical role in changing the overall DL-SOTs in epi-samples. Actually, such a DL-SOT generated by the Rashba SOC has been proposed in earlier theoretical works,\textsuperscript{23,24} and been investigated lately.\textsuperscript{40,41} The coupling of Rashba field and the $s$-$d$ exchange field acts as the key to generating such a Rashba-induced DL-SOT.\textsuperscript{23,24,40}

\textbf{Conclusions}

Our results provide two highlights associated with the Rashba SOC: firstly, with a well-defined Pt/Co interface in epi-samples with favorable interlayer flatness, an exceptionally large $\lambda_{IEE}$ of a few nanometers is extracted from a detailed MR analysis with the origin of REE, demonstrating strikingly efficient interfacial charge-to-spin conversion that originates from the large Rashba SOC; the obtained $\lambda_{IEE}$ in epi-samples is about one order of magnitude larger than poly-samples, quantitatively consistent with an anomalous tenfold increase in the $\xi_{FL}$ compared to poly-samples; secondly, the twofold increase of $\xi_{DL}$ in epi-samples is attributed not to the bulk SHE mechanism, but probably to the Rashba SOC at Pt/Co interface. For the first point, the extracted $\lambda_{IEE}$ is self-explanatory in the sense that a $T$-independent $\alpha_R$ in the order of $10^{10} \, eV \, m$ is derived from it which is consistent with previous spin-resolved photoemission experiments and first-principles calculations,\textsuperscript{50,51} indicating that $\alpha_R$ is only associated with the electronic band property and not affected by external $T$. As for the striking tenfold increase in the $\xi_{FL}$ for epi-samples that suggests much enhanced Rashba SOC, one possible explanation is that a flat interface free from defects such as grain boundaries can change the transport properties\textsuperscript{52} and induce larger band splitting.\textsuperscript{14} For the second point, our work demonstrates a significant control of SOTs by the Rashba SOC in HM/FM bilayers, highlighting the unique role of REE in manipulating the SOTs that conventional SHE fails to capture. Our results suggest that both DL- and FL-SOTs can be significantly modulated by interface modification even without a drastic
increase in the material resistivity such as by oxygen incorporation. From this standpoint, such a method towards the SOT engineering might find its compatibility to electronics integration in view of lower impedance and power consumption.
Figures (captions included)

Figure 1 (a) Out-of-plane and (b) in-plane x-ray diffraction patterns of thin films of Pt(10)/Co(10)/cap deposited onto Al₂O₃(0001) (epi-) and Si/SiOₓ (poly-) substrates. Inset of (a) shows the reflection of high energy electron diffraction image for epitaxial Pt(1)/Co(1)/cap tri-layers. (c) X-ray reflection spectra for Pt(10)/Co(₁₅) bilayers. Unit in the brackets is in nanometer.
Figure 2 (a) Schematic demonstration of the system and coordinate axis for magnetoresistance measurement used in this study. (b) Illustration of the REE with a shift in the Fermi contour with an external electric field. (c), (d) $R-H$ curves of poly-Pt(1)/Co(3) and epi-Pt(1.8)/Co(3) with magnetic field applied in x, y and z directions. (e), (f) Inverse sheet resistance ($1/R_{xx}^0$) and Rashba-Edelstein magnetoresistance (REMR) for Pt($t_N$/Co(3)) at RT. (g) Temperature ($T$) dependence of REMR for Pt($t_N$/Co(6)). (h) $T$ dependence of resistivity for poly-and epi-Pt, Co films from $1/R_{xx}^0-t_N$ analysis.
Figure 3 (a), (b) REMR as a function of Pt layer thickness $t_N$ for different Co layer thickness $t_F$ at 300 K and 4.2 K. Solid lines are the fitting curves by assuming $\lambda_{sf} \cdot \theta_{SH} = 0.2$ nm and $\lambda_{sf} \cdot \sigma_N = 0.69 \times 10^{-15} \, \Omega \, m^2$ (c), (d) $t_N$ dependence of $1/R_{xx}^0$ for varying $t_F$ at 300 K and 4.2 K. (e), (f) $t_F$ dependence of $\lambda_{IEE}^{eff}$ and $d_{2eff}^{eff}$ at 300 K and 4.2 K extracted from (a) and (b) by assuming $\lambda_{sf} \cdot \theta_{SH} = 0.2$ nm and $\lambda_{sf} \cdot \sigma_N = (0.36 - 0.69) \times 10^{-15} \, \Omega \, m^2$. 
Figure 4 $\lambda_{\text{IEE}}$ and Rashba parameter $\alpha_R$ as a function of $T$. 
Figure 5  (a) Schematic demonstration of the measurement set-up for ST-FMR. (b) and (c) show the ST-FMR spectra and their fitting by Eq. (3) at different frequencies for poly-Pt(10)/Co(3) and epi-Pt(10)/Co(4); (e) and (f) show the corresponding $\xi_{ST}$ calculated using Eq. (4).
Figure 6 (a) and (b) show the ST-FMR spectra for poly- and epi-samples with bilayer thin film structure of Pt(10)/Co($t_F$). The $\xi_{ST}$ and $1/\xi_{ST}$ are shown as a function of $t_F$ and $1/\xi_{ST}$ in (c) and (d).
Methods
Sample fabrication.

Epitaxial Pt/Co bilayers were deposited onto Al₂O₃(0001) substrates at room temperature (RT) using ultrahigh vacuum rf sputtering facility with a base pressure better than 2×10⁻⁷ Pa. The operation pressure and power for sputtering were 0.2-0.4 Pa and 50-80 W for all the materials. As control samples, polycrystalline Pt/Co bilayers were deposited onto thermally oxidized Si substrates at RT. Prior to deposition, all the substrates were annealed at 200°C for 1 hour to remove residual gas. Amorphous 2 nm Al-O capping layer was deposited upon Co layer to prevent samples from oxidation. To measure the magnetoresistance (MR) from the Pt/Co bilayers, wedged thin films were fabricated into Hall bar devices that are 10 µm wide and 20-160 µm long using photo-lithography and Ar ion milling. Flat films with fixed layer thickness were used to calibrate the wedged layer thickness. To make ST-FMR samples, thin films were patterned into rectangular shapes that are 6 µm wide and 12-36 µm long using electron beam lithography with co-planar waveguide.

Electrical measurement

MR measurements were performed using physical property measurement system with a 360° rotator, the current applied to the Hall bars varies from 0.1-1.0 mA; ST-FMR measurements were performed by injecting a radio-frequency (rf) current into the sample micro-strip by signal generator E8257D (Agilent technologies) and by applying an external field at 45 deg to the strip for detecting the signal. The voltages were detected by means of lock-in amplifier using SR830 (Stanford) with an amplitude modulation of the rf current for a reference signal.
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**Author contributions**

Y.D. deposited the films and fabricated the devices. Y.D. performed the magnetoresistance measurement and corresponding data analysis with inputs from S.T., S.K., H.G., J.R, M.K. and J.N. Y.D. and S.K. contributed to the spin-torque ferromagnetic resonance measurement and data analysis with inputs from M.K. and J.N. S.T. developed the theory. Y.D. and J.N. drafted the manuscript. All authors discussed the results and commented on the manuscript. J.N. directed the research.

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Supplementary Figure S1: CALIBRATION OF THE SPIN-HALL RASHBA-EDELSTEIN MAGNETORESISTANCE (a) Schematic view of parallel configuration for Co and Pt conduction channel. (b) Parasitic MR in single Co layer at 300 K and 4.2 K. (c) Temperature dependence of parasitic MR for 6 nm Co layer.
Supplementary Figure S2

Supplementary Figure S2: **FIELD-ANGULAR DEPENDENCE OF ST-FMR SIGNAL.** The symmetric ($V_s$) and anti-symmetric ($V_{as}$) component of the ST-FMR signal are plotted as a function of the in-plane field angle, shown in (a) and (b). For symmetric component shown in (a), the total fit (red) is composed of four terms (Ref. 2): $\cos \varphi \sin 2\varphi$ (blue), $\sin \varphi \sin 2\varphi$ (pink), $\sin 2\varphi$ (green) and $\sin \varphi$ (orange) that represent respectively the main DL torque in y direction, a FL torque in x direction, a FL torque in z direction and the torque arising from spin pumping effects; for anti-symmetric component shown in (b), the total fit (red) is composed of three terms: $\cos \varphi \sin 2\varphi$ (blue), $\sin \varphi \sin 2\varphi$ (pink), $\sin 2\varphi$ (green) and that represent respectively the main FL torque in y direction, a DL torque in x direction, a DL torque in z direction.
Supplementary Figure S3: QUANTITATIVE ESTIMATION OF REE CONTRIBUTION IN POLY- SAMPLES. The stacking structure of the poly-sample is Pt(tN)/Co(3). (a) Pre-defined $\lambda_{sf}$ as a function of Pt conductivity $\sigma_{Pt}$. (b) MR data and the fitting using Eq. 2 in the main text for poly-Pt(tN)/Co(3) using $\lambda_{sf}$ shown in (a). A constant $d_R$ of 0.6 nm is used. (c) $\lambda_{IEE}$ and $\theta_{SH}$ extracted from the fitting. Inset shows the $\theta_{SH}$ as a function of Pt conductivity. (d) Ratio of $\lambda_{IEE}$(poly-) over $\lambda_{IEE}$(epi-).
Supplementary Figure S4: DERIVATION OF THE SPIN-HALL RASHBA-EDELSTEIN MAGNETORESISTANCE. Schematic demonstration of the system used for deriving the spin-Hall Rashba-Edelstein MR. The Rashba-Edelstein (RE) region with a thickness $d_2$ is sandwiched by the ferromagnetic layer (F) with a thickness of $d_F$ and non-magnetic layer (N) with a thickness of $d_1$. $\lambda_{EE}$ and $\lambda_{IEE}$ are the inverse and direct Edelstein length in the RE region. $\rho_N$, $\lambda_N$ and $\theta_{SH}$ are the resistivity, spin diffusion length and spin Hall angle in N.
Supplementary Note 1. CALIBRATION OF THE SPIN-HALL RASHBA-EDELSTEIN MAGNETORESISTANCE

Physical parameters such as the inverse Edelstein length ($\lambda_{IEE}$), effective thickness of the Rashba region ($d_R$) et al. in this work are extracted from magnetoresistance (MR) that is originated from the charge-spin current interplay, i.e. the spin Hall effect (SHE) and the Rashba-Edelstein effect (REE). Therefore, parasitic MR originated from e.g. the geometrical size effect\(^1\) has to be subtracted from the raw data to obtain genuine MR signal. To perform the MR calibration, we use a parallel circuit to describe the MR in the Pt/Co bilayer (Fig. S1). The longitudinal resistance with field applied in $z$ and $y$ direction are respectively

$$R_{xx}(H_z) = R_0 = \frac{R_1 R_2}{R_1 + R_2}$$

$$R_{xx}(H_y) = R'_0 = \frac{R'_1 R'_2}{R'_1 + R'_2}$$

where $R'_1 = R_1 + \Delta R_1$, $R'_2 = R_2 + \Delta R_2$. $\Delta R_1$ is the parasitic resistance change in Co layer and $\Delta R_2$ is the overall resistance change induced by SHE and REE. Thus we have

$$R_{xx}(H_y) = \frac{(R_1 + \Delta R_1)(R_2 + \Delta R_2)}{R_1 + R_2 + \Delta R_1 + \Delta R_2} \approx R_0 + R_0 \left( \frac{\Delta R_1}{R_1} + \frac{\Delta R_2}{R_2} \right)$$

The MR originated from the SHE and REE ($MR_{int}$) and the raw MR ratio without calibration reads

$$MR_{int} \approx \frac{R_1 (R_2 + \Delta R_2)}{R_1 + R_2 + \Delta R_2} - \frac{R_1 R_2}{R_1 + R_2} \approx \frac{\Delta R_2}{R_2}$$

$$MR_{raw} = \frac{R_{xx}(H_y) - R_{xx}(H_z)}{R_{xx}(H_z)} \approx \frac{\Delta R_1}{R_1} + \frac{\Delta R_2}{R_2} = MR_{int} + \frac{\Delta R_1}{R_1}$$

in which $\Delta R_1/R_1$ is the parasitic MR originated from single Co layer. Therefore, one obtains the $MR_{int}$ in Pt/Co bilayers by subtracting the parasitic MR from single Co layer with the same measurement geometry.
Supplementary Note 2. FIELD-ANGULAR DEPENDENCE OF ST-FMR SIGNAL

In order to evaluate the effect of spin pumping et al. on the ST-FMR signal, we measure the ST-FMR spectra by varying the direction of external magnetic field for sample with film structure of epi-Pt(10)/Co(10.2). The symmetric and anti-symmetric components of the signal were extracted for each field angle as plotted in Fig. S2 (a) and (b). The symmetric component is fitted with a sum of four phenomenological terms: \( \cos \varphi \sin 2\varphi \) (blue), \( \sin \varphi \sin 2\varphi \) (pink), \( \sin 2\varphi \) (green) and \( \sin \varphi \) (orange) that represent respectively the main DL torque in y direction, a FL torque in x direction, a FL torque in z direction and the torque arising from spin pumping effects; the anti-symmetric component is fitted with 3 terms: \( \cos \varphi \sin 2\varphi \) (blue), \( \sin \varphi \sin 2\varphi \) (pink), \( \sin 2\varphi \) (green) and that represent respectively the main FL torque in y direction, a DL torque in x direction, a DL torque in z direction. From the analysis it is found that the signals arising from spin pumping and other extrinsic effect (e.g. waveguide contact issue) are less than 5% of the genuine signal that we obtained. Therefore, we conclude that those parasitic effects are negligible for our ST-FMR analysis.
Supplementary Note 3. QUANTITATIVE ESTIMATION OF REE CONTRIBUTION IN POLY- SAMPLES

The REE effect in poly-samples is quantitatively estimated using Eq. 2 shown in the main context. The stacking structure of the sample is Pt(tN)/Co(3). Here the EY spin relaxation mechanism is assumed (Fig. S3 (a)), same as that for epi-samples. To perform the fitting, a constant $d_R$ of 0.6 nm is assumed independent of $T$, as suggested from epi-results. The $T$ dependent MR data can be well-fitted as shown in Fig. S3 (b). The extracted $\lambda_{IEE}$ and $\theta_{SH}$ are shown in Fig. S3 (c): the $\lambda_{IEE}$ increases as decreasing $T$, showing same tendency as that in epi-samples, while its amplitude is roughly one order of magnitude lower (Fig. S3 (d)), suggesting significantly reduced interfacial charge-to-spin conversion efficiency in poly-samples; the extracted $\theta_{SH}$ decreases with decreasing $T$, and follows the intrinsic/side-jump spin Hall mechanism (the inset of Fig. S3 (c)), consistent with previous reports.\textsuperscript{3}
Supplementary Note 4. DERIVATION OF THE SPIN-HALL RASHBA-EDELSTEIN MAGNETORESISTANCE

The system for drift-diffusion modeling in this work is shown in Fig. S4. The Rashba-Edelstein (RE) region with a thickness \(d_2\) is sandwiched by the ferromagnetic layer (F) with a thickness of \(d_F\) and non-magnetic layer (N) with a thickness of \(d_N\). By adding an Edelstein contribution \(e(\lambda_{\text{IEE}} \hat{z} \times \mathbf{E})\) of spin accumulation\(^{4,5}\) into the SMR theory\(^{6-8}\), the spin accumulation in N reads:

\[
\begin{align*}
\delta \mu_{N1}(z) &= a e^{-\frac{z-d_2}{\lambda_N}} + b e^{\frac{z-d_2}{\lambda_N}}, \quad (d_2 < z < d_1 + d_2) \\
\delta \mu_{N2}(z) &= c e^{-\frac{z}{\lambda_N}} + d e^{\frac{z}{\lambda_N}} + e \lambda_{\text{IEE}} \hat{z} \times \mathbf{E}, \quad (0 < z < d_2)
\end{align*}
\]

(1)

where \(\lambda_N\) is the spin diffusion length in N, \(e\) is the electron charge, \(\lambda_{\text{IEE}}\) is the inverse Edelstein length and \(\mathbf{E}\) is the external electric field. The spin current in region N1 and N2 reads:

\[
\begin{align*}
\mathbf{j}_s^{N1}(z) &= -\frac{1}{2eR_N A} \left[ -ae^{-\frac{z-d_2}{\lambda_N}} + be^{\frac{z-d_2}{\lambda_N}} \right] - \theta_1 j_c e_y \\
\mathbf{j}_s^{N2}(z) &= -\frac{1}{2eR_N A} \left[ -ce^{-\frac{z}{\lambda_N}} + de^{\frac{z}{\lambda_N}} \right] - \theta_2 j_c e_y
\end{align*}
\]

(2)

where \(R_N = \rho_N \lambda_N / A\), and \(\lambda_{\text{IEE}} = \alpha_R \tau / \hbar\). \(\lambda_{\text{IE}}, \alpha_R\) and \(\tau\) are Rashba parameter and effective momentum scattering time. The spin current going through F/N2 interface reads:

\[
\mathbf{j}' = \frac{g_{11}'}{2eA_j} \left( \mathbf{M} \times (\mathbf{M} \times \delta \mu(0)) \right) + \frac{g_{11}''}{2eA_j} (\mathbf{M} \times \delta \mu(0))
\]

(3)

where the spin mixing conductance \(g_{11} = g_{11}'+ i g_{11}''\). Solving Eq. (1), (2) and (3), the overall MR due to both SHE and REE reads:

\[
\frac{\Delta R_{xx}}{R_0} \approx - \frac{\lambda_N / \rho_N}{d_N / \rho_N + d_F / \rho_F} \left( \theta_{\text{SH}} \Lambda + \frac{\lambda_{\text{IEE}}}{2 \lambda_N} \Phi \right)^2 \text{Re} \left[ \frac{g_5}{1 + \Gamma g_5} \right]
\]

where
\[
\begin{aligned}
\Lambda &= \tanh \left( \frac{d_N}{2\lambda_N} \right) \\
\Phi &= \begin{cases} 
\sinh \left( \frac{d_N - d_2}{\lambda_N} \right) / \sinh \left( \frac{d_N}{\lambda_N} \right) & \text{for } d_N > d_2 \\
1 & \text{for } 0 < d_N < d_2
\end{cases} \\
\Gamma &= \coth \left( \frac{d_N}{\lambda_N} \right) \\
g_s &= 2\rho_N^2 \lambda_N G_{11}
\end{aligned}
\]

Considering an exponential decay in the REE spin accumulation, the overall MR reads:
\[
\frac{\Delta R_{xx}}{R_0} \approx -\frac{\lambda_N / \rho_N}{d_N / \rho_N + d_F / \rho_F} \text{Re} \left[ \frac{2\rho_N \lambda_N G_{11}}{1 + 2\rho_N \lambda_N G_{11} \coth(d_N / \lambda_N)} \right] \\
\times \left\{ \theta_{SH} \tanh \left( \frac{d_N}{2\lambda_N} \right)^2 + 2 \left( \frac{\lambda_{IEE}}{2\lambda_N} \right)^2 \sinh \left( \frac{d_N - d_2}{\lambda_N} \right) \right\}
\]

where
\[
\langle \sinh((d_N - d_2)/\lambda_N) \rangle = \frac{\sinh(d_N/\lambda_N) - r \cosh(d_N/\lambda_N) + re^{-(d_N/\lambda_N)/r}}{(1 - r^2)[1 - e^{-(d_N/\lambda_N)/r}]}
\]
\[
\langle \sinh^2((d_N - d_2)/\lambda_N) \rangle
\]

\[
= \frac{1}{2[1 - e^{-(d_N/\lambda_N)/r}]^2} \left[ \cosh(2d_N/\lambda_N) - 2r \sinh(2d_N/\lambda_N) - e^{-(d_N/\lambda_N)/r} \right]
\]
\[
+ e^{-(d_N/\lambda_N)/r} - 1
\]

where \langle \cdots \rangle denotes the average with respect to \(d_2\) weighted by \(e^{-d_2/d_R}\), \(r \equiv d_R / \lambda_N\), where \(d_R\) is the effective thickness of the RE region.
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