Giant interfacial Rashba spin-orbit torque in atomically thin metallic Pt|$\text{Co}|\text{Al}|\text{Pt}$ heterostructures

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Spin-orbit interactions in metals and their ability to generate spin-current has been the hallmark of spintronics in the last decade. Beyond its fundamental interest as a source of spin-current, a quest in today’s spintronics, the manipulation of magnetization via the transfer of spin angular momentum has proven to be energy efficient for spin-based memory, spin Hall nano-oscillator as well as for neuromorphic devices. Therein, charge to spin conversion is believed to be essentially governed by two main mechanisms: the spin Hall effect (SHE) in heavy metals and the Rashba effect (RE) at interfaces although the latter being often considered negligible in all-metallic interfaces. Here, we have examined the impact of light metal element interface on SOT and its efficiency in terms of damping-like ($H_{DL}$) and field-like ($H_{FL}$) effective fields in ultrathin ferromagnets. Unexpectedly, we have observed large $H_{FL}/H_{DL}$ ratio ($\sim$2.5) upon inserting a 1.4 nm thin Al layer in Pt|$\text{Co}|\text{Al}|\text{Pt}$. From our modelling, experimental results strongly evidence the presence of a giant interfacial Rashba effect at Co|$\text{Al}$ interface producing a very large $H_{FL}$. The occurrence of these efficient Rashba states is further validated by reducing the SHE contribution from Pt as well as by demonstrating current-induced magnetization reversal showing a significant decrease of the critical current for the switching. Hence we believe that our results are of a prime importance as they provide a clear route for reaching ultimate spin torque efficiency.

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A turning point has been made in the field of spintronics with the emergence of the spinor-bitronics that exploits spin-orbit coupling (SOC) to provide new phenomena such as strong perpendicular anisotropy and interfacial antisymmetric exchange interactions. Moreover, SOC may generate efficient spin-current via the spin Hall effect (SHE) useful to switch the magnetization or drive chiral spin textures in heavy-metal/ferromagnet stacks. In atomically thin layers or interfaces with broken inversion symmetry, SOC also leads to Rashba spin splitting that modifies the electronic ground states affecting the spin transport through the Rashba-Edelstein effect (REE). Both SHE and REE possess the property to convert a charge current into either an out-of-equilibrium spin-current or a spin accumulation carrying angular momentum. Upon current injection, the spin-accumulation and the spin-current may transfer their angular momentum to the adjacent ferromagnetic layer by precessing around the exchange field. To conserve the total angular momentum, the local magnetization experiences the so-called spin-orbit torques (SOTs). This questions about the characteristic length over which the transverse component of the spin (often termed as spin decoherence length and is predicited to be in the range of a few atomic lattice parameters) relaxes and how it impacts the nature and the amplitude of SOTs.

The torques, predominantly related to SHE, occur from 4d or 5d host metallic materials and are often described by a flow of spins transverse to the charge current satisfying the quantum law \( \mathbf{J}_\sigma \propto \theta_{\text{SHE}} \mathbf{J}_c \times \hat{\sigma} \) with \( \theta_{\text{SHE}} \), the spin-Hall angle (SHA) being a scaling parameter. It results in a linear relationship between the SHE-SOT and the applied electric field \( \mathcal{E} \) as \( \tau_{\text{SOT}} \propto \sigma_{\text{SHE}}^{\text{eff}} \mathcal{E} \) with \( \sigma_{\text{SHE}}^{\text{eff}} \) is the spin Hall conductivity. Concerning the SOT related to REE, it arises from the charge current injected in the plane of an inversion asymmetric stack that produces a non-equilibrium spin-accumulation owing to a Rashba spin-structure generally accompanying the spin-momentum locking (SML). The corresponding linear relationship is given by \( \hat{\mu} \propto \kappa_{\text{IEE}} \mathcal{E} \), where \( \kappa_{\text{IEE}} = \mathcal{N}^{\text{DOS}} \lambda_{\text{IEE}} \) is the Rashba-Edelstein tensor, \( \lambda_{\text{IEE}} \) the Rashba-Edelstein length and \( \mathcal{N}^{\text{DOS}} \) the 2-dimensional density of states (DOS). Importantly, this makes the REE to be distinct in nature from SHE. However, both spin-current and spin-accumulation generated by either way, may act in concert to exert net SOTs onto a magnetization vector with two different components: the damping-like torque (DLT), \( \tau_{\text{DL}} \propto \hat{\mathbf{m}} \times (\hat{\mu} \times \hat{\mathbf{m}}) \) and the field-like torque (FLT), \( \tau_{\text{FLT}} \propto \hat{\mu} \times \hat{\mathbf{m}} \) giving rise correspondingly to damping-like (\( H_{\text{DL}} \)) and field-like (\( H_{\text{FLT}} \)) effective fields.

Manipulating the magnetization direction via torques generated by SOC has gained great attention in the last decade, but disentangling the exact origin of SOTs still remains today a complex
problem, particularly in case of very thin magnetic films. Beyond the fundamental interest, this research is also largely motivated by the contact need for improvement of the SOT efficiency for writing the information in future SOT based devices. In the pioneer experiments of by Miron et al in Pt|Co|AlO$_x$\textsuperscript{15}, the origin of the spin generation responsible for the magnetization switching by DLT was pointed out to be predominantly from REE at the Co|AlO$_x$ interface. Note that the REE also gives rise to a significant FLT in this system \textsuperscript{14,16}, suggesting the role of a large electric field at Co|Oxide interface. Nevertheless, in all metallic heterostructures\textsuperscript{13,17–20}, SHE is largely considered as the primary source of DLT, even though recently, a large interfacial REE has been measured in Ir|ferromagnetic|Ta stacks with a significant contribution from DLT\textsuperscript{21}. Other effects such as spin-filtering or quantum confinement of charges and subsequent change of the electronic band structure in the limit of atomically thin ferromagnetic layer may also contribute to the net SOTs\textsuperscript{22–27}, notably at surface states of topological insulators (TIs)\textsuperscript{28} and in 2D electron gases \textsuperscript{29}.

In order to address this fundamental question, we have investigated the properties of SOTs in atomically thin metallic structures, namely Co layers ranging from 1.4 nm down to 0.4 nm, sandwiched between Pt and light element based (Al, Cu)|Pt overlayers. Using the second harmonic Hall measurement technique, we precisely determined the two SOT components and identified their thickness dependence with respect to the ferromagnetic, heavy-metal and overlayers. We unambiguously measure a substantial increase in the DLT by more than \textasciitilde 30\% and the FLT by more than \textasciitilde 190\% compared to the best-reported results in the literature\textsuperscript{14}. By combining the experimental results with calculations, we explicitly demonstrate the existence of an enhanced SOT mainly due to a strong REE in these asymmetric Pt|Co|Al stacks as witnessed by a FLT over DLT ratio as large as 2.5 for 0.55 nm thick Co, the highest value ever reported in metallic systems. We believe that our results shed some light on the electronic transport and its spin polarization at the interfaces between the classical regime and quantum (2D-like) one.

I. DETERMINING TORQUE ANATOMY FROM EXPERIMENTS

A. Harmonic Hall measurements and fitting methods

To quantify the SOTs acting on a Co ferromagnet (FM) of thickness $t_{Co}$, we have used the second harmonic Hall measurement technique. The measurement geometries are shown in the top panel of Fig. 1. In Fig. 1a-b, we display the dependence of second harmonic Hall voltage ($V_{2f}$, in green) acquired on a Pt(8)|Co(0.9)|Al(1.4)|Pt(3) sample vs. the in-plane magnetic fields (normalized with
Figure 1. Harmonic Hall measurements of damping-like (DL) and field-like (FL) spin-orbit effective fields in Pt(8)|Co(0.55-1.4)|Al(1.4)|Pt(3). The top panel illustrates the DL geometry when $H \parallel I$ ($H_x$) and the FL geometry when $H \perp I$ ($H_y$) for a thin Co layer of magnetization $M$, at position $(\theta, \phi)$. a-b) In-plane field dependence of $V_{2f}$ (green) for $t_{Co} = 0.9$ nm in (a) DL geometry and in (b) FL geometry. The solid black lines are fits to extract DLT and FLT. The horizontal axis is normalized with respect to the anisotropy field, $H_K$. The inset of Fig. 1a shows normalized $V_f$ (green) as a function of in-plane magnetic field and Stoner-Wohlfarth fit (solid black lines). The data are shown after correcting the contributions from inhomogeneous device geometry as well as the thermal contributions. c) DL and d) FL SOT fields vs. $I$ for $t_{Co} = 0.55$ nm (disc), 0.9 nm (square) and 1.4 nm (star). The dotted lines are the linear fit forced to cross through (0,0) with uncertainty shown in shadow.
respect to the anisotropy field, $H_K$) in DL geometry and FL geometry (thicknesses in parentheses are given in nm). The inset of Fig. 1a shows the variation of first harmonic Hall voltage ($V_f$) vs. $H_x$ fitted with Stoner-Wohlfarth model. The fit allows to extract the anisotropy fields ($H_K$) and the magnetization angle ($\theta$) away from the easy-axis ($z$). Note that the accurate measurements of $H_K$ and $\theta$ are critical prerequisites to extract precise values of $H_{DL}$ and $H_{FL}$ from $V_{2f}$. It must be pointed out that extracting the effective fields by restricting to the linear regime of the $V_{2f}$ may give rise to a strong discrepancy, especially in the estimation of $H_{FL}$ values.

The $H_{DL}$ and $H_{FL}$ SOT components are determined by fitting $V_{2f}$ vs. $H_x$ and $H_y$ respectively, and taking into account the contribution from the anomalous Hall (AHE) and planar Hall effects (PHE)$^{30}$ (see SI-I), even though, the PHE contribution is one order of magnitude smaller than the AHE. The fits are displayed by black continuous lines in Fig. 1a-b. In Fig. 1c-d, we show the $H_{DL}$ and $H_{FL}$ vs. $I = I_{RMS}$ for three Pt(8)$|$Co($t_{Co}$)$|$Al(1.4)$|$Pt(3) samples with Co thicknesses, $t_{Co} = 0.55, 0.9$ and $1.4$ nm, which are in the range, or below the typical transverse spin decoherence length ($\lambda_\perp \approx 1.5 − 2$ nm) of Co$^{10,11}$. We note that $H_{DL}$ changes sign with $\mathbf{m}$, whereas $H_{FL}$ does not, in agreement with the expected SOT symmetries. The $H_{DL}$ and $H_{FL}$ are measured at several currents and the slope of $H_{DL,FL}$ vs. $I$ (dashed lines in Fig. 1(c-d)) is considered to extract the SOT fields with a high accuracy. Furthermore, the SOT fields show linear scaling with current as expected.

B. Anatomy of SOT from the Co thickness dependence

In order to unravel the SOT mechanisms acting on an atomically thin ferromagnet, we have investigated two series of samples having Al and Cu at the top Co interface: Pt(8)$|$Co($t_{Co}$)$|$Al(1.4)$|$Pt(3) and Pt(8)$|$Co($t_{Co}$)$|$Cu(1.4)$|$Pt(3). Starting from negligible torque in fully symmetric Pt(8)$|$Co(0.9)$|$Pt(8) stack (see SI-IV), the asymmetric ‘control’ sample Pt(8)$|$Co(0.9)$|$Pt(3) gives $H_{DL}=0.80 \pm 0.05$ mT and $H_{FL} = 0.58\pm0.15$ mT for a current density $\mathcal{J}_{Pt}$ in Pt = $10^{11}$ A/m$^2$. To obtain an integrated torque in the whole ferromagnet of thickness $t_{Co}$, we define a normalized quantity by multiplying $H_{DL,FL}$ with $t_{Co}$, obtained for a $10^{11}$ A/m$^2$ current density in Pt. As demonstrated in forthcoming data analyses, the Cu samples series will be used as an almost pure ISHE reference sample, i.e. free of significant REE contribution. Several features can be identified highlighting similarities and differences between the two series. First, as shown in Fig. 2a, the integrated DL component $(H_{DL} \times t_{Co})/\mathcal{J}_{Pt}$ is about 65% larger in Pt|$Co|Al(1.4)|Pt$ samples compared to Pt|$Co|Cu(1.4)|Pt$. At $t_{Co} = 0.9$ nm, the value of DLT is about 2.5 mT$\times$nm$/\left(10^{11}$Am$^{-2}\right)$ for Al
Figure 2. Co layer thickness dependence of SOT effective fields in Co|Al(1.4) and Co|Cu(1.4) systems. a) DL field and (b) FL field multiplied by Co thickness as a function of $t_{Co}$ for $J_{Pt} = 10^{11}$ A/m$^2$ in Pt(8)|Co(0.55-1.4)|Al(1.4)|Pt(3) and Pt(8)|Co(0.4-1.2)|Cu(1.4)|Pt(3). c) $t_{Co}$ dependence of the ratio $\zeta$ of $H_{FL}$ and $H_{DL}$. The red and blue dashed lines are fits to experimental data points using our theoretical model and the black dashed line are calculated $\zeta$ values without any Rashba interface (see SI-VII).

and 1.5 mT×nm/(10$^{11}$ Am$^{-2}$) for Cu. Second, $(H_{DL} \times t_{Co})/J_{Pt}$ exhibits a similar qualitative maximum in 1.0 – 1.2 nm window. Third, we find that, in both series, FLT exhibits an increase on reducing $t_{Co}$ (Fig. 2b) resulting in a dominating FLT at small $t_{Co}$. If the trend of the thickness dependence of the SOT resembles between the Al(1.4) and Cu(1.4) series, their magnitude strongly differs. In particular, we observe a substantial increase in the FLT for Al(1.4) series i.e. from 1.7 mT×nm/(10$^{11}$ Am$^{-2}$) for $t_{Co} = 1.4$ nm to 5.2 mT×nm/(10$^{11}$ Am$^{-2}$) for $t_{Co} = 0.55$ nm, whereas the maximum value of FLT reaches only 2 mT×nm/(10$^{11}$ Am$^{-2}$) in the Cu series. As per our knowledge, such a large increase in atomically thin Co layer has never been reported before.

Another striking result is the amplitude of the FLT to DLT ratio ($\zeta = H_{FL}/H_{DL}$) for Pt|Co|Al|Pt. A
rise in FLT is expected (i) in the case of SHE, when the spin decoherence length is larger than the magnetic thickness as shown by black dotted curve in Fig. 2 and (ii) from the interfacial REE. To characterize the respective weight of the two components, we define the ratio \( \zeta = \frac{H_{FL}}{H_{DL}} \) as a figure of merit. The evolution of \( \zeta \) as a function of \( t_{Co} \) is plotted in Fig. 2c.

The ratio \( \zeta = \frac{H_{FL}}{H_{DL}} \) then appears as a main figure of merit whose evolution vs. \( t_{Co} \) is plotted in Fig. 2c. The SOT ratio shows again a similar behaviour for the two series: it saturates for \( t_{Co} \gtrsim 1.2 \) nm with comparable magnitude, \( \zeta \approx 0.7 \). \( \zeta < 1 \) is indeed expected in the limit of a ‘thick’ ferromagnet\(^{17} \) when only SHE is considered without Rashba (as for Cu but we consider a small Rashba for Cu series, the black and blue curves are different). Note that similar value of \( \zeta \) were also obtained in our previous studies on 4 nm thick Co|Ni ferromagnetic multilayer\(^{31} \). However, reducing \( t_{Co} \) below 1.2 nm, we find here that \( \zeta \) increases sharply for Pt|Co|Al|Pt series and eventually reaches at 2.5 for \( t_{Co} \approx 0.55 \) nm, whereas it remains smaller than one down to \( t_{Co} \approx 0.7 \) nm for Pt|Co|Cu|Pt. We assert that such enhancement of the SOT ratio by a factor larger than three, strongly challenges the SHE origin of SOT in stacks involving Al.

C. Role of the Co|Al and Co|Cu interfaces

In Fig. 3, we display the evolution of \( H_{DL}/J_{Pt} \) and \( \zeta = \frac{H_{FL}}{H_{DL}} \) as a function of the thickness of the light element i.e. Al (red points) and Cu (blue points), showing several striking behavior. First we find that DLT exhibits an unexpected strong increase with \( t_{Al} \) in the 1-3 nm window from \( \sim 1.5 \) mT/(10\(^{11}\) A/m\(^2\)) for \( t_{Al} = 1 \) nm to a saturation value \( \sim 4.5 \) mT/(10\(^{11}\) A/m\(^2\)) for \( t_{Al} \geq 3 \) nm. In parallel, the SOT ratio \( \zeta \) increases much more with \( t_{Al} \) (Fig. 3b) from \( \zeta = 0.75 \) for \( t_{Al} = 0 \) nm to \( \zeta = 1.75 \) for \( t_{Al} \geq 3 \) nm manifesting again a peculiar behaviour. These behavior are very different from what we obtain in the Cu series on varying \( t_{Co} \) in which \( H_{DL}/J_{Pt} \approx 1.5 \) mT/(10\(^{11}\) A/m\(^2\)) and \( \zeta \approx 0.7 \) remain roughly constant vs. \( t_{Cu} \), as shown by blue open triangles in Fig. 3a and Fig. 3b. This makes that FLT rises by at least a factor of 6 by inserting 2 nm Al on top of Co compared to Cu.

Note that in these two samples series, the light element film is covered by a 3nm Pt layer, that could be also a source of spin-charge conversion, and thus of SOT. Indeed two major conclusions can be draught emphasizing i) a large electronic transmission across the top Co|Cu|Pt interfaces\(^{32,33} \) partially compensating the SHE torque from the bottom Pt, irrespective of \( t_{Cu} \) and ii) a progressive extinction of SHE action from the top Pt with increasing the thickness of ‘Al barrier’. Interestingly, the effect of the electronic transmission across Al or Cu also appeared in our AHE measurements.
Figure 3. Al and Cu layer thickness dependence of SOTs in Pt(8)|Co(0.9)|Al(tAl),Cu(tCu)|Pt(3). a) $H_{DL\text{ field}}$ vs. Al (red) and Cu (blue) layer thickness. The change in $H_{DL}$ as a function of Al thickness is fitted as indicated by the dashed lines (in red) using our model detailed in the text and SI (see SI-VII). We used $l_{Pt}^{sf}=1.5$ nm, $\tilde{r}_s=2$, $\theta_{SHE}^{Pt(0)}=0.09$ and $T$ varying from $T=0.8$ for $t_{Al}=0$ nm to $T=0$ for $t_{Al}>3$ nm. b) $\zeta=H_{FL}/H_{DL}$ ratio vs. Al (red) and Cu (blue) layer thickness. The thick and constant blue lines are guide to the eye to indicate that SOTs are independent of Cu thickness.

As a matter of fact, we observe the signature of a smaller scattering probability in the top Pt layer, and thus the smaller AHE observed in Al samples compared to Cu may be due to a stronger reflection of the polarised electrons in Co layer by the Al layer (Fig.S5 and see SI-III).

More quantitatively, the effective spin Hall angle (SHA) arising from SHE ($\theta_{SHE}^{eff}$) may be put in the following form:

$$\theta_{SHE}^{eff} \approx \theta_{SHE}^{bulk\ Pt} \left[ 1 - L \right] \left[ F(8, T_{Co\|Pt}) - F(3, T_{Co\|Al\|Pt}) \right],$$

where, $\theta_{SHE}^{bulk\ Pt}$, is the SHA of bulk Pt, $T$ is the transmission coefficient, $L$ is the spin memory loss$^{32,34,35}$ at the interface as calculated theoretically$^{36-38}$ and the function $F$ defines the spin-back flow in Pt and is given by

$$F(t_{Pt}, T) \approx \frac{T_{T_{s}^{Pt}} \cosh \left( \frac{t_{Pt}}{\lambda_{sf}^{Pt}} \right) \left[ 1 - \text{sech} \left( \frac{t_{Pt}}{\lambda_{sf}^{Pt}} \right) \right]}{1 + T_{T_{s}^{Pt}} \cosh \left( \frac{t_{Pt}}{\lambda_{sf}^{Pt}} \right)}$$

$\lambda_{sf}^{Pt}$ is the spin-diffusion length (SDL) in Pt and $r_{s}^{Pt}$ is the spin-resistance or resistance to spin-flip (see SI-VII). From our fit shown as dotted line in Fig. 3a, we extract $\lambda_{sf}^{Pt} \approx 1.5$ nm in agreement with Ref.$^{33}$, $r_{s}^{Pt} \approx 2$. $\theta_{SHE}^{Pt}=0.09$ and $\theta_{SHE}^{bulk\ Pt} \approx 0.22$ in agreement with the literature for Pt of resistivity $\rho = 25 \mu \Omega \text{cm}^{34,39}$. We have considered $T = 0.8$ and $\mathcal{L} \approx 0.5$. Similar
Figure 4. Dependence of the effective $\theta_{SHE}^{\text{eff}}$ and $\zeta = H_{FL}/H_{DL}$ ratio on $t_{Pt}$ in the $\text{Pt}(t_{Pt})|\text{Co}(0.9)|\text{Al}(5)$ series. a) Effective $\theta_{SHE}^{\text{eff}}$ vs. $t_{Pt}$ (green points). The solid line is the result of the fit performed enable to extract $t_{sf}^{\text{Pt}} = 1.5$ nm, bulk $\theta_{SHE}^{\text{eff}} = 0.09$ of Pt and $t_{s}^{\text{Pt}} = 2$ using Eq. 2. b) The $\zeta$ ratio as a function of $t_{Pt}$.

values of $\theta_{SHE}^{\text{Bulk Pt}}$ are determined from our AHE measurements by considering $T = 0.8$ and $L = 0.5 \approx 0.5$ nm. SHE compensation from the top injection then approaches $\frac{2}{3}$ in our control $\text{Pt}(8)|\text{Co}(0.9)|\text{Pt}(3)$ structure. The main consequence is that the large increase of $H_{FL}$ and the SOT ratio $\zeta$ in the Al series cannot be simply explained because of such bottom/top SHE compensation, even partially. This provides an additional signature of the existence of a large, and unexpected REE from Co|Al.

D. Rashba-Edelstein vs. spin Hall effect

In order to better discard the different contributions to the effective SOT, our approach has been then to reduce the SHE action by decreasing the bottom Pt thickness in $\text{Pt}(t_{Pt})|\text{Co}(0.9)|\text{Al}(5)$ series. We first quantify the SHE contribution in the charge to spin conversion ratio from Pt by extracting $\theta_{SHE}$ from the DL field according to $\theta_{SHE}^{\text{eff}} = \frac{2e}{\hbar} \frac{\mu_{0} H_{DL}}{J_{Pt} M_{s} M_{Co}}$, where $e$ is the electron charge, $M_{s}$ the saturation magnetization. In Fig. 4a, we show that $\theta_{SHE}^{\text{eff}}$ increases with $t_{Pt}$ according to a standard law $\propto \theta_{SHE}^{\text{Bulk Pt}} \times F(t_{Pt})$ (see. Eq.[2]) before reaching a saturation for $t_{Pt} \geq 5$ nm. From the fit, the spin-diffusion length (SDL) and $\theta_{SHE}^{\text{eff}}$ are determined to be $\approx 1.5$ nm and $\approx 0.09$ in agreement with the data of Fig. 3. In Fig. 4b, we show the impact of REE from the Co|Al interfaces on the SOTs by restricting ourselves to $t_{Co} = 0.9$ nm. For the chosen Co thickness, the FLT dominates the DLT over all the Pt thickness range. In addition, a remarkable increase...
of $\zeta$ is observed in the $t_{Pt} = 2-5$ nm range, i.e. in the range where SHE has not yet reached its maximum efficiency. These results hence undoubtedly support the role of a significant Co|Al Rashba interface superimposed to SHE from Pt for Al based samples. Our data also yield a clear quantitative understanding of how the Rashba effect at metallic interfaces may strongly affect the $H_{FL}$ in chemically asymmetric Pt|Co|Al|Pt stacks as also described in Fig. 2 and Fig. 3.

II. MODELLING AND NUMERICAL ANALYSIS OF SOT.

From fundamentals, SOT is generally determined via the real and imaginary parts of the spin-mixing conductance$^{19,20,40,41}$ However, for $t_{Co}$ smaller than the decoherence length, the description becomes much more complex$^{42}$. In order to tackle this issue corresponding to our experimental conditions, we have developed a model and numerical analyses (described in detail in (see SI-VII)) gathering main of the ingredients discussed above. To this aim, we have adapted the generalized drift-precession-diffusion equations$^{18,42–44}$ in agreement with the spin-dependent Boltzmann theory$^{19,20}$. We provide here a subsequent numerical insights for $H_{DL}$ and $H_{FL}$, considering the integrated value of the torque $\tau_{SOT}$ generated from the $s$-$d$ exchange ($J_{xc}$). By reciprocity, the SOT, acting on the out-of equilibrium spin-density $\hat{\mu}_F$ in Co gives$^{45}$:

$$\tau_{SOT} = \int_V \frac{d\hat{M}}{dt} dV \approx \int_V \hat{\mu}_F \times \hat{m} \tau_J dV. \quad (3)$$

$V$ is the volume of the magnet and $\mu_F$ are expressed in the same unit $\mu_B/V$ and $\tau_J = \frac{\hbar}{J_{xc}}$. Based on our present knowledge for Co|Pt$^{32,46,47}$, the resulting calculated SOT fields are in excellent agreement with the experimental results for both DL and FL components (see dashed lines in Fig. 2 and Fig. 3). From these fits, we are able to evaluate the precession length $l_J = v_F \tau_J = 0.75$ nm and a transverse decoherence length $l_{\perp} = v_F \tau_\Delta = 1.7$ nm (in the range of the values given in Ref.44) for Co with $T_{Co|Pt} = \frac{G_{Co|Pt}}{G_{sh}} = 0.8$. Here, $G_s$ is the spin surface conductance and $G_{sh}$ is the Sharvin conductance. From our previous experimental data, the spin resistances were fixed to $\tilde{r}_{s}^{Pt} = 2$ and $\tilde{r}_{s}^{Al|Pt} = 3$ in unit of $\frac{1}{G_{sh}}$ (see SI-VII). We point out that a clear difference between Co|Al and Co|Cu is the large transmission coefficient for the top interface in Co|Cu|Pt if compared to that of Co|Al|Pt. It can be determined: $T_{Co|Cu|Pt} = \frac{G_{Co|Cu|Pt}}{G_{sh}} = 0.5$ and $T_{Co|Al|Pt} = \frac{G_{Co|Al|Pt}}{G_{sh}} = 0.2$. In fact, as aforementioned, such a large $T_{Co|Cu|Pt}$ can be also deduced from our measured AHE amplitude in this system compared to the Co|Al|Pt. This feature indicates an enhancement of AHE via non-local spin-current effect in Co|Cu|Pt when electrons are allowed to scatter in Pt$^{39}$. 


In order to fit the experimental data, we must account for by the spin-accumulation \( \mu_F = \mu_{REE} \) at Co|Al(Cu) due to REE. For Pt|Co|Cu|Pt, we considered an homogeneous \( \mu_{REE} \) in Co and constant vs. \( t_{Co} \). From the fit, we estimate that REE contributes \( \approx 45\% \) to the FLT (\( \approx 55\% \) due to SHE) for \( t_{Co} = 0.4 \) nm in Cu samples. In contrast, for the Pt|Co|Al|Pt series, we found a much larger spin accumulation \( \mu_{REE} \) with a strong dependence on \( t_{Co} \) to fit the data. The Larmor precession length and decoherence length were fixed to \( \lambda_J = 4.7 \) nm and \( \lambda_\perp = 1.7 \) nm respectively. \( \lambda_J \) is in very close agreement with the one extracted in other 3d NiFe transition metal ferromagnet from orbital Hall effects measurements\(^48\). In the Al series, \( \mu_{REE} \) varies within a lengthscale of the order of 0.75 nm in Co as extracted from the fit. As a consequence the FLT contribution from REE reaches \( \approx 75\% \) for \( t_{Co} = 0.55 - 1.2 \) nm (\( \approx 25\% \) from SHE), whereas it contributes only by 60 \% for \( t_{Co} = 1.4 \) nm (\( \approx 40\% \) from SHE). We finally notice that a possible explanation for the observation of large FLT in Pt|Co|Al|Pt stacks may be the workfunction difference between Co and Al generating a strong interfacial electric field possibly mediated by charge transfer from top to bottom Co interfaces. We recently referred to this effect in order to elude the large effective interfacial Dzyaloshinskii-Moriya interaction (DMI) found in Pt|Co|Al stacks\(^49\). In conclusion, the FLT may largely exceed the DLT in the limit of a small \( t_{Co} \) below \( \lambda_\perp^{Co} \) that we have found to be 1.7 nm, in agreement with the literature\(^10,11,48\) (see SI-VII).

**A. Enhanced magnetization switching efficiency through optimized SO torques**

In order to further demonstrate the improvement provided by the REE contribution at the Co/Al metallic interface, we show the SOT-induced current magnetization switching experiments in the various samples explored. Such experiments consist of performing magnetization reversal of a thin 0.9 nm Co layer in Pt(8)/Co(0.9)/Al(\( t_{AL} \))/Pt(3) made of the same sized Hall cross-bars (5 \( \mu \)m wide and 40 \( \mu \)m long) through injection of a large current density in the multilayer and in particular in the bottom Pt. During the experimental protocol, an external magnetic field \( H_x \), either positive of negative, is applied along the current injection in order to break the mirror symmetry and to observe the magnetization reversal with one or the other magnetic cycle polarity. The results of SOT-induced magnetization reversal are shown in Fig. 5 for respective positive \( H_x = +250, 373, 495, 608 \) mT (Fig. 5a) or equivalent negative \( H_x = -240, -368, -495, -600 \) mT (Fig. 5b) magnetic fields for the representative \( t_{AL} = 3 \) nm sample. These exhibit, in each configuration, a clear magnetization reversal in the range \( J_{Pt} \approx \pm 2 - 2.5 \times 10^{11} \) A/m\(^2\). Three main features can be formulated from these measurement series: First a magnetization reversal cycle (whose
Figure 5. **SOT-induced magnetization reversal experiments**: Al thickness dependence of current-induced magnetization reversal: (a) Current-induced magnetization reversal in Pt(8)/Co(0.9)/Al(3)/Pt(3) sample for different (a) positive and (b) negative in-plane magnetic fields. The inversion of R-I loops with field polarity confirms that SOT are at play in the switching mechanism. (c) $R_{AHE}$ as a function of pulsed current in Pt(8)/Co(0.9)/Al($t_{Al}$)/Pt(3) series of samples for a positive magnetic field of $H_x = +250$ mT in each case. All the measurements were performed at room temperature (300 K). The width of pulse was 100 $\mu$s and the $R_{AHE}$ was measured after a delay of 40 $\mu$s.

polarity or chirality that is the occurrence of the switching from positive to negative resistance states at positive or negative current) depends on the sign of $H_x$ as expected from the SOT symmetry. Second the critical current density decreases as $|H_x|$ increases as also observed in previous experiments$^{50,51}$ with typical current density reaching $J_{Pt} = 3-3.5 \times 10^{11} A/m^2$. Third, the magnetization reversal is fully achieved between the two saturated states (parallel vs. antiparallel) owing to the respective level of resistances matching with the AHE data measured in the saturated states. In Fig. 5c-d), we compare the different magnetization reversal cycles acquired at $H_x = +250$ mT for different Al thickness $t_{Al} = 1, 2, 3, 4$ and 5 nm. For $t_{Al} = 3$ and 4 nm, we find very similar critical current densities for the complete magnetization reversal (see Fig. 3a and b)). On the contrary, we find a much stronger critical current for $t_{Al} = 2$ and 1 nm, and more importantly, we could not reach the complete magnetization reversal, as shown by the intermediate AHE levels. These results hence confirm that increasing the Al thickness in the Pt(8)/Co(0.9)/Al($t_{Al}$)/Pt(3)
series makes \(i\) the SOT components larger mainly \textit{via} an enhanced Rashba SOT and \(ii\) allows a reduction of the critical current density for switching by more than 30\% with the possibility of a full magnetization switching. Moreover, our samples have a large anisotropy (in the range of 1.5 to 2T), that is essential for thermal stability of SOT devices.

III. CONCLUSIONS

In summary, we demonstrate that SOTs in atomically thin ferromagnetic layers ranging from 0.4 nm to 1.4 nm, are only partially controlled by spin-current generation in the bulk of the bottom Pt layer via SHE mechanism. Surprisingly, the amplitude of SOTs varies significantly with a light element insertion that is either Al or Cu, on account of change in the electronic properties (band structure) at the top interface. As a result, we observe much stronger SOTs, both DLT and even more FLT in Pt|Co|Al|Pt system than the Pt|Co|Cu|Pt stacks. Although the SHE contribution to the SOTs from the bulk of heavy-metals is widely recognized, our discovery emphasizes the impact of Co|Al interface on the SOTs, as demonstrated by enhanced magnetization switching efficiency. In particular, we observe a giant increase in the field-like component and the corresponding ratio of \(H_{FL}/H_{DL}\) upon reducing the Co thickness in single atomic thickness range. Combined with the analytical calculations, this observation demonstrates that an additional charge to spin conversion mechanism, the REE, exists at all-metallic Co|Al interface that dominates over the bulk SHE contributions. Our experimental findings suggest the significant impact of charge transfer effects and the presence of large interfacial spin-orbit related conversion effect at Co|Al interface\(^{49}\). Such mechanisms of spin-current generation are indeed existing at oxide interfaces, in 2DEG systems and at surface states of topological insulators, our demonstration that they do play a crucial role in all-metallic interfaces allows for a giant charge to spin conversion in the limit of atomically thin ferromagnets and opens to our opinion a new research direction for CMOS-compatible and cost-effective spinorbitronic based technology.

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AUTHOR CONTRIBUTIONS

S.K., N.R., H.J. and V.C. conceived the project. S.K., Y.S. and F. A. deposited the magnetic multilayers and fabricated the devices with the help of S.C. S.K performed the transport measurements with the help of N.R. and J.M.G. S.K., H.J., V.C. and A.F. discussed and analysed the results. H.J. developed the analytical and theoretical modelling. S.K., H.J. and V.C. prepared the manuscript and all authors discussed and contributed to the final manuscript.
METHODS

Sample preparation: The samples were deposited on a thermally oxidized Si/SiO$_2$ wafer using d.c. magnetron sputtering at an Ar$^+$ pressure $2.5 \times 10^{-3}$ mbar and the base pressure was $8 \times 10^{-8}$ mbar. The thickness of SiO$_2$ is 280 nm. They all exhibit perpendicular magnetic anisotropy, characterized by an anisotropy field, $H_K$ (see SI-IV). The thin films were grown after depositing a 5 nm Ta seed layer to improve the perpendicular magnetic anisotropy and adhesion. The thicknesses of materials were calibrated using x-ray reflectometry. To perform electrical measurements, the thin films were fabricated in 5 µm wide and 40 µm long Hall bar structures using optical lithography and Ar$^+$ ion-milling etching technique.

Electrical and harmonic Hall voltage measurements: The devices were wire-bonded on a commercial chip to perform magneto-electric transport and harmonic Hall measurements. All the measurements were performed at room temperature. The van der Pauw method is used to measure the resistivity of thin films (see SI-VI). The resistivity of Pt, Cu, Ta and Co was found to be 24.2 µΩ×cm, 11.5 µΩ×cm, 169 µΩ×cm, and 20.2 µΩ×cm, respectively. A parallel resistivity model is used to estimate current shunt through each layer. Harmonic Hall voltage measurements were performed at an a.c. frequency of 733 Hz. The anomalous Hall voltage ($V_H$) can be expressed as:

$$V_H = V_0 + V_1 \sin(2\pi ft) + V_{2f} \cos(4\pi ft). \quad (4)$$

The 1$^{st}$ ($V_1$) and 2$^{nd}$ ($V_{2f}$) harmonics of the Hall voltages were measured simultaneously with multiple harmonic lock-in amplifier. A large external in-plane magnetic field, exceeding the anisotropy field ($H_K$) of the sample, was swept along (transverse to) the current to measure $H_{DL}$ ($H_{FL}$). The in-plane field was applied with a tilt $\delta \theta_H \approx 7 \pm 1^\circ$ to ensure a single magnetic domain state within the Hall cross. The effective fields are obtained by analysing the Hall voltage signal, considering the entire field dependence i.e., from $H=0$ to $H>H_K$. As shown in Fig.1, the $H_{DL}$ ($H_{FL}$) are extracted by fitting $V_{2f}$ vs $H$ using:

$$V_{2f}^{DL} = -\frac{1}{2} \sin(\theta) \left( \frac{H_{DL} R_{AHE}}{H_K \cos(2\theta) - H \sin(\delta \theta_H - \theta)} + \frac{2H_{FL} R_{PHE} \sec(\delta \theta_H) \sin(\theta)}{H} \right) I_0 \quad (5)$$

$$V_{2f}^{FL} = \frac{1}{2} \cos(\theta) \sin(\theta) \left( \frac{H_{FL} R_{AHE}}{H_K \cos(2\theta) - H \sin(\delta \theta_H - \theta)} \right) I_0 + \frac{1}{2} \cos(\theta) \sin(\theta) \left( \frac{2R_{PHE} H_{DL} \sec(\delta \theta_H) \sin(\theta)}{H} \right) I_0. \quad (6)$$
Where $\theta(H)$ is the magnetization angle from z-axis, $R_{AHE}$ is the anomalous Hall resistance and $R_{PHE}$ is the planar Hall resistance. $\theta(H)$, $\delta\theta_H$ and $H_K$ are extracted from $V_I$ using the Stoner-Wohlfarth model. The fittings were performed after subtracting thermal contributions as well as constant offset due to asymmetric voltage leads from the raw data. More details are available in SI-I and SI-II.

DATA AVAILABILITY

All relevant data within this paper are available from the corresponding author upon reasonable request.

CODE AVAILABILITY

The code is available from the corresponding author upon reasonable request.

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