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## Large Interfacial Rashba Interaction Generating Strong Spin–Orbit Torques in Atomically Thin Metallic Heterostructures

Sachin Krishnia, Yanis Sassi, Fernando Ajejas, Nicolas Sebe, Nicolas Reyren, Sophie Collin, Thibaud Denneulin, András Kovács, Rafal E. Dunin-Borkowski, Albert Fert, Jean-Marie George, Vincent Cros,\* and Henri Jaffrès\*



further validated by demonstrating current-induced magnetization reversal showing a significant decrease of the critical current for switching.

KEYWORDS: Rashba-Edelstein effect, Spin-orbit torques, Spintronics, Magnetization switching

In modern nanomagnetism, spin-orbit coupling (SOC) at interfaces is exploited to generate strong interfacial perpendicular anisotropy (PMA) or antisymmetric exchange interactions.<sup>1</sup> In spintronics, SOC is the key interaction to generate spin-current via the spin Hall effect (SHE) in heavy metals, enabling magnetization switching $^{2-6}$  and driving skyrmions<sup>7</sup> and domain walls. In atomically thin layers with broken inversion symmetry, the interplay of low-dimensionality and SOC may also lead to a Rashba spin-splitting<sup>8-10</sup> modifying the electronic ground state, thus affecting the spin transport via the Rashba-Edelstein (REE)<sup>11</sup> or orbital Rashba effects (ORE).<sup>12,13</sup> These phenomena, namely, SHE<sup>5,14,15</sup> and REE (or ORE),<sup>5,10,11,15,16</sup> may convert a charge current  $\mathcal{J}_c$ into either an out-of-equilibrium spin-current  $\mathcal{J}_\sigma$  or spin accumulation  $\hat{\mu}$  (out-of equilibrium spin-density) carrying an angular momentum. Upon current injection, those can, in turn, transfer their angular momentum to the adjacent ferromagnetic layer by precessing around the exchange field.

Both  $\mathcal{J}_{\sigma}$  and  $\hat{\boldsymbol{\mu}}$  may exert spin-orbit torques (SOTs) onto a magnetization vector  $\mathcal{M} = M\hat{\mathbf{m}}$  with two components: damping-like torque (DLT),  $\hat{\tau}_{\text{DL}} \propto \hat{\mathbf{m}} \times (\hat{\boldsymbol{\mu}} \times \hat{\mathbf{m}})$ , and field-like torque (FLT),  $\hat{\tau}_{\text{FL}} \propto \hat{\boldsymbol{\mu}} \times \hat{\mathbf{m}}$ . The nature and strength of the corresponding DL ( $H_{\text{DL}}$ ) and FL ( $H_{\text{FL}}$ ) effective fields strongly depend on the relaxation of the transverse spin i.e. on the spin decoherence length, which is predicted to be a few

atomic lattice parameters.<sup>17,18</sup> Manipulating the magnetization via SOTs and disentangling the exact origin of SOTs to use them adequately are still challenging today for writing processes in magnetoelectronic devices.

In the pioneer experiments by Miron et al. in PtlColAlO<sub>x</sub><sup>2</sup> SOT was discussed in terms of dominating FLT arising from REE at the ColOxide interface, larger than SHE-DLT from Pt, suggesting the crucial role of a large interfacial electric field at the ferromagnet/oxide interface. Although, in metals, SHE is largely considered as the primary source of DLT,<sup>3,19–21</sup> a significant interfacial REE has been recently observed in Irl ferromagnetic|Ta contributing to DLT.<sup>22</sup> In the limit of atomically thin magnetic layers, additional mechanisms might also influence the strength of the SOTs: spin-filtering or charge quantum confinement,<sup>23–27</sup> surface states of topological insulators (TIs)<sup>28</sup> and 2D electron gases.<sup>29</sup>

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Moreover, recent theory has evidenced additional orbital currents in light elements from orbital Hall effects (OHE) and/or orbital Rashba effects (ORE) superimposing to the spin currents.<sup>30</sup> These orbital currents may result in the generation of orbital torques<sup>12,31</sup> and orbital Rashba magnetoresistance.<sup>13,32</sup> This questions the actual origin of the magnetic torques in multilayers with light elements.

In this study, we investigate 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 overlayers, i.e., Al or Cu. Using the second harmonic Hall measurement technique,<sup>33,34</sup> we precisely determine the amplitude of the SOT vs the ferromagnetic film thickness, the light-element overlayer, and the heavy-element. Compared to the best-reported results,<sup>34</sup> we find an increase of the DLT by ~30% together with a drastic rise in the FLT by more than ~190%. With the help of our spin-dependent Boltzmann calculations, we explicitly demonstrate the occurrence of enhanced SOT due to a strong REE in asymmetric PtlColAll Pt stacks, from a FLT over DLT ratio as large as 2.5 for 0.55 nm thick Co, one of the highest values ever reported in metals.

Samples were grown using magnetron sputtering at the base pressure  $6 \times 10^{-8}$  mbar. In Figure 1a, we show X-ray



Figure 1. (a) XRR measurements (blue) and fittings (red) on Pt(8)ICo(0.7)IAl(1.4)IPt(3) sample. (b) Measurements of XPS on similar samples are shown by red color together with a reduced Pt capping (2 nm) with oxidized Al (in black). (c) HAADF image in STEM of the sample with corresponding EDX elemental map shown in (d).

reflectivity (XRR) measurements (blue) and fit (red) on a Ta(5)|Pt(8)|Co(0.7)|Al(1.4)|Pt(3) sample where the layer thicknesses are found to be in good agreement with nominal values (<3% error). The XRR measurements reveal 0.29 and 0.12 nm roughness at PtlCo and ColAl interfaces, respectively, comparable with the SiO<sub>2</sub>|Ta interface (0.45 nm). We have also performed X-ray photoelectron spectroscopy (XPS) on Pt(8)|Co(0.7)|Al(1.4)|Pt(t<sub>Pt</sub>) with  $t_{Pt} = 2$  and 3 nm to probe the chemistry of Al underneath Pt (Figure 1b with black and red colors respectively). As the Al 2*p* peak (71 eV) is totally screened by the 4*f* Pt peaks, the 2*s* Al bonds are probed. The peak for the 2 nm Pt capping can be fitted with two contributions attributed to AlOx (dashed) and metallic Al (dotted) respectively whereas the 3 nm Pt capping sample only reveals a single metallic state of Al thus discarding any

oxidization state. In Figure 1c and d, we show the energy dispersive X-ray spectroscopy (EDX) measurements in scanning transmission electron microscopy (STEM). The Ta, Pt Co, and Al layers can be clearly distinguished by bright and black Z-contrasts in high-angle annular dark-field (HAADF) image. In Figure 1d, we show elemental distribution of Ta (white), Pt (green), Co (blue), and Al (red). Together with XRR, the HAADF and EDX characterizations clearly show homogeneous and flat interfaces without any sign of oxidation of the Al film.

To evaluate the two components of the SOTs, we employed the harmonic Hall measurement technique on 5  $\mu$ m wide Hall bars with an AC excitation of 727 Hz (SI-I). First, we compare the results from the two series of multilayers with Al and Cu light element interfaced with Co:  $Pt(8)|Co(t_{Co})|Al(1.4)|Pt(3)$ and  $Pt(8)|Co(t_{Co})|Cu(1.4)|Pt(3)$ . Starting from negligible torques in a fully symmetric Pt(8)|Co(0.9)|Pt(8) stack as expected (SI-V), the asymmetric "control" sample Pt(8) Co(0.9)|Pt(3) gives  $H_{\rm DL}$  = 0.80 ± 0.05 mT and  $H_{\rm FL}$  0.58 ± 0.15 mT for a current density  $\mathcal{J}_{Pt}$  in Pt = 10<sup>11</sup> A/m<sup>2</sup> (later used as a reference value). To obtain integrated torques over the whole ferromagnetic layer of thickness  $t_{Co}$  and hence to be able to make an accurate comparison, we define a normalized quantity by multiplying  $H_{DL,FL}$  with  $t_{Co}$  for a 10<sup>11</sup> A/m<sup>2</sup> current density in Pt. This corresponds to the equivalent torque values for an applied electric field  $\mathcal{E} = \rho_{\rm Pt} \times \mathcal{J}_{\rm Pt} = 2.5 \times 10^4 \, {\rm V/m}$ owing to the Pt resistivity  $\rho_{\rm Pt}$  = 25  $\mu\Omega$ ·cm (SI-III). Our analyses mainly depend on the anisotropy field  $H_k$  but not on the saturation magnetization at the limit of thin Co  $M_{\rm s}$  = 1.1  $\pm$ 0.1 MA/m, remaining however quite independent of the Co thickness  $t_{Co}$  for both the Al and Cu series (SI-IV).

Several features may be highlighted with similarities and differences between the Al and Cu series. First, as shown in Figure 2a, the integrated DL component  $(H_{\rm DL} \times t_{\rm Co})/\mathcal{J}_{\rm Pt}$  is



**Figure 2.** (a) Damping-like (DL) field and (b) field-like (FL) field multiplied by  $t_{Co}$  for  $\mathcal{J}_{Pt} = 10^{11} \text{ A/m}^2$  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  $\zeta = H_{FL}/H_{DL}$ . Red and blue dashed lines are fits to experimental data points using our theoretical model, and black dashed line in (c) is calculated  $\zeta$  values without any Rashba interface (see SI-VII).

about 65% larger in PtlColAl(1.4)lPt samples compared to Ptl ColCu(1.4)lPt for almost the whole Co thickness. At  $t_{\rm Co} = 0.9$  nm, the value of DLT is about 2.5 mT × nm/(10<sup>11</sup> A·m<sup>-2</sup>) for Al and 1.5 mT × nm/(10<sup>11</sup> A·m<sup>-2</sup>) for Cu. We note however that the  $(H_{\rm DL} \times t_{\rm Co})/\mathcal{J}_{\rm Pt}$  product exhibits in each case a similar maximum close to  $t_{\rm Co} = 1.0-1.2$  nm that we ascribe to the spin decoherence length at small thickness. Second, we find



**Figure 3.** (a)  $H_{\text{DL}}$  vs Al (red) and Cu (blue) layer thickness. The variation in  $H_{\text{DL}}$  is fitted by the dashed red lines (SI-VII). We used  $l_{\text{sf}}^{\text{pt}} = 1.5 \text{ nm}$ ,  $\tilde{r}_s = 2$ ,  $\mathcal{L} = 0.4$ ,  $\theta_{\text{SHE}}^{\text{Pt}(0)} = 0.09$  and the transmission coefficient  $\mathcal{T}$  varying linearly from  $\mathcal{T} = 0.8$  for  $t_{\text{Al}} = 0 \text{ nm}$  to  $\mathcal{T} = 0$  when  $t_{\text{Al}} > 3 \text{ nm}$ . (b)  $H_{\text{FL}}$  vs Al (red) and Cu (blue) layer thickness. (c)  $\zeta = H_{\text{FL}}/H_{\text{DL}}$  vs Al (red) and Cu (blue) layer thickness. Thick and constant blue lines are a guide to the eye.

that the FLT exhibits an increase in reducing  $t_{\rm Co}$  (Figure 2b) in both series, resulting in a dominating FLT component at small  $t_{\rm Co}$ . Nevertheless, their SOT magnitude strongly differs. We observe a substantial increase of the FLT for Al(1.4), i.e., from 1.7 mT × nm/(10<sup>11</sup> A·m<sup>-2</sup>) for  $t_{\rm Co} = 1.4$  nm to 5.2 mT × nm/ (10<sup>11</sup> A·m<sup>-2</sup>) for  $t_{\rm Co} = 0.55$  nm, whereas the maximum value of FLT reaches only 2 mT × nm/(10<sup>11</sup> A·m<sup>-2</sup>) for Cu. Such a large FLT increase in the atomically thin Co layer has never been reported to our knowledge. This striking result manifests also by a rise of the FLT to DLT ratio  $\zeta = \frac{H_{\rm FL}}{H_{\rm DL}}$  for PtlColAllPt that we will consider as a new figure of merit (see Figure 2c).

Such a behavior is expected either for SHE when  $t_{\rm Co}$  becomes smaller than the spin-decoherence length (black curve in Figure 2c from our modeling (SI-VII) or for an increased weight of REE (red dotted line for Al and blue for Cu in Figure 2c). We find that  $\zeta = \frac{H_{\rm FL}}{H_{\rm DL}}$  saturates for  $t_{\rm Co} \gtrsim 1.2$  nm with a magnitude  $\zeta \approx 0.7$  in the "thick" ferromagnet limit for SHE.<sup>19</sup> This corresponds also to the case of the Cu series with moderate Rashba effects. Similar value of  $\zeta \simeq 0.7$  were also obtained on 4 nm thick ColNi multilayers.<sup>35</sup> Here, on reducing  $t_{\rm Co}$  below 1.2 nm,  $\zeta$  increases sharply for the PtlColAll Pt series (Figure 2c) and eventually reaches 2.5 for  $t_{\rm Co} = 0.55$  nm. This ratio remains smaller than unity down to  $t_{\rm Co} = 0.7$  nm for PtlColCulPt. Such FLT enhancement by a factor larger than three from  $t_{\rm Co} = 1.4$  nm to  $t_{\rm Co} = 0.55$  nm strongly challenges the SHE (and/or OHE) origin of SOT with Al.

In Figure 3, we present the evolution of  $H_{\rm DL}/\mathcal{J}_{\rm Pt}$  (Figure 3a),  $H_{\rm FL}/\mathcal{J}_{\rm Pt}$  (Figure 3b), and their ratio  $\zeta = \frac{H_{\rm FL}}{H_{\rm DL}}$  vs Al (red points) and Cu (blue points) thickness (Figure 3c). Both DLT and FLT exhibit a strong enhancement with  $t_{\rm Al}$  in the 1–3 nm window, particularly pronounced for the FLT. We find that FLT (respectively DLT) increases from ~0.65 mT/10<sup>11</sup> A·m<sup>-2</sup> (~0.8 mT/10<sup>11</sup> A·m<sup>-2</sup>) for  $t_{\rm Al} = 0$  nm to a saturation value of ~7 mT/10<sup>11</sup> A·m<sup>-2</sup> (~4.4 mT/10<sup>11</sup> A·m<sup>-2</sup>) for  $t_{\rm Al} = 3$  nm. This corresponds to a boost of the FLT by more than a factor of 10 upon inserting 3 nm of Al on top of 0.9 nm Co. Conversely, the ratio  $\zeta$  is also enhanced with  $t_{\rm Al}$  (Figure 3c) from  $\zeta = 0.8$  for  $t_{\rm Al} = 0$  to  $\zeta = 1.6$  for  $t_{\rm Al} \ge 3$  nm. First, we comment the behavior of DLT. Based on the results from our fits (SI-VII), we assign the rise of DLT vs  $t_{\rm Al}$  to the decrease of the transmission coefficient  $\mathcal{T}_{\uparrow\downarrow}^{\rm Co/Al/Pt}$  at the top of the

structure varying linearly from  $\mathcal{T}_{\uparrow\downarrow}^{\text{Co/Pt}} \cong 0.8$  for  $t_{\text{Al}} = 0$  nm to  $\mathcal{T}_{\uparrow\downarrow}^{\text{Co/Al3/Pt}} \cong 0$  when  $t_{\text{Al}} > 3$  nm. This results in the quenching of the opposite SHE from the top Pt. However, part of the DLT enhancement also originates from the conversion of the FLT into the DLT in the Co thickness via the spin precession mechanism. This trend is very different in the "reference" Cu series vs  $t_{\text{Cu}}$  as we find that  $H_{\text{DL}}/\mathcal{J}_{\text{Pt}} \approx 1.5 \text{ mT}/10^{11} \text{ A} \cdot \text{m}^{-2}$  and  $\zeta \approx 0.7$  remain roughly constant vs  $t_{\text{Cu}}$  (blue open triangles in Figure 3a and b).

Two important conclusions may then be drawn for the DLT: (i) a progressive extinction of the SHE action from the top Pt on increasing the thickness of Al through the quench of the electron transmission at the ColAllPt interfaces and (ii) a larger electronic transmission across ColCulPt<sup>36,37</sup> partially compensating the SHE torque from the bottom Pt (irrespective of  $t_{Cu}$ ). Considering both bottom and top Pt layers, the effective spin Hall angle (SHA) ( $\theta_{SHE}^{eff}$ ) is

$$\theta_{\text{SHE}}^{\text{eff}} \approx \theta_{\text{SHE}}^{\text{bulkPt}} [1 - \mathcal{L}] [\mathcal{F}(8, \mathcal{T}_{\text{ColPt}}) - \mathcal{F}(3, \mathcal{T}_{\text{ColAllPt}})]$$
(1)

where  $\theta_{\text{SHE}}^{\text{bulkPt}}$  is the bare SHA of bulk Pt,  $\mathcal{T}$  is the effective transmission coefficient at the top, and  $\mathcal{L}$  is the spinloss.<sup>36,38,39</sup> The function  $\mathcal{F}$ :

$$\mathcal{F}(t_{\mathrm{Pt}}, \mathcal{T}) \approx \frac{\mathcal{T}\tilde{r}_{s}^{\mathrm{Pt}} \mathrm{coth}\left(\frac{t_{\mathrm{Pt}}}{\lambda_{sf}^{\mathrm{Pt}}}\right) \left[1 - \mathrm{sech}\left(\frac{t_{\mathrm{Pt}}}{\lambda_{sf}^{\mathrm{Pt}}}\right)\right]}{1 + \mathcal{T}\tilde{r}_{s}^{\mathrm{Pt}} \mathrm{coth}\left(\frac{t_{\mathrm{Pt}}}{\lambda_{sf}^{\mathrm{Pt}}}\right)}$$
(2)

describes the spin back-flow in Pt with  $\lambda_{sf}^{pt} \approx 1.5$  nm being its spin-diffusion length (SDL) in Pt as in ref 37 and  $\tilde{r}_{s}^{pt} \approx 2$  is its spin-resistance in the unit of the inverse of the Sharvin conductance (SI-VII). From our fitting procedure (Figure 3a), we could extract  $\theta_{SHE}^{eff} = (1 - \mathcal{L})\mathcal{T}\theta_{SHE}^{bulkPt} = 0.075 \pm 0.01$ using  $\theta_{SHE}^{bulkPt} \approx 0.225 \pm 0.025$  in agreement with the literature for a Pt resistivity of  $\rho = 25 \,\mu\Omega \cdot \text{cm.}^{37,40}$  Similar values of  $\theta_{SHE}^{bulkPt}$ were determined from AHE.<sup>40</sup> We considered here  $\mathcal{T}_{\uparrow\downarrow}^{Co/Pt} = 0.8$  and  $\mathcal{L} \approx 0.4$ . Our fitting procedure applied to AHE (SI-IV) uses the same set of parameter values as for SOTs. The SHE compensation from the top injection then approaches 75% in our 'control' Pt(8)|Co(0.9)|Pt(3) structure and 60% in the Pt(8)|Co(0.9)|CulPt(3) series. This implies that the expected increase of the DLT is 25% (0.75/0.6) from Pt(8)|Co(0.9)|Pt(3) to Pt(8)|Co(0.9)|Cu|Pt(3) and by a factor x2.5 to Pt(8)|Co(0.9)|Al(3)|Pt(3) (Figure 3a).

In order to highlight any additional REE (or ORE) arising from ColAl, we reduced the bottom Pt thickness in the Pt( $t_{Pt}$ )l Co(0.9)lAl(5) series to attenuate the SOTs originating from SHE. We first quantified this SHE contribution by extracting  $\theta_{SHE}$  from  $H_{DL}$  according to  $\theta_{SHE}^{\text{eff}} = \frac{2e}{\hbar} \frac{\mu_0 H_{DL} M_s t_{Co}}{\mathcal{J}_{Pt}}$  (e is the electron charge). In Figure 4a, we observe that  $\theta_{SHE}^{\text{eff}}$  increases



**Figure 4.** a) Effective  $\theta_{\text{SHE}}^{\text{eff}}$  vs  $t_{\text{Pt}}$  (green points). The solid line is the fit enabling to extract  $l_{\text{sf}}^{\text{Pt}} = 1.5$  nm, Pt bulk  $\theta_{\text{SHE}}^{\text{bulk Pt}} = 0.225 \pm 0.025$  (effective  $\theta_{\text{SHE}}^{\text{eff}} = 0.075 \pm 0.01$ ), and  $\tilde{r}_{\text{s}}^{\text{Pt}} = 2$  using eq 2. (b) Plot of the  $\zeta$  ratio vs  $t_{\text{Pt}}$ .

with  $t_{\rm Pt}$  according to  $\propto \theta_{\rm SHE}^{\rm bulkPt} \times \mathcal{F}(t_{\rm Pt})$  (eq 2) before reaching its saturation value for  $t_{\rm Pt} \geq 5$  nm. From the fitting procedure, the spin-diffusion length (SDL) and  $\theta_{\rm SHE}^{\rm eff}$  are, respectively,  $\simeq 1.5$  nm and  $\simeq 0.08$ , in agreement with data of Figure 3. In Figure 4b, we show the impact of the additional REE (or ORE) occurring from ColAl on the SOTs for  $t_{\rm Co} = 0.9$  nm. First, the FLT clearly dominates the DLT for all the Pt thicknesses. In addition, a remarkable increase of  $\zeta$  is observed at the lowest  $t_{\rm Pt} = 2-5$  nm thickness range, i.e., where SHE has not yet reached its maximum efficiency. This undoubtedly supports the role of a significant Rashba interaction (i.e., REE and/or ORE) superimposed to SHE. Our data also yield a clear quantitative understanding of how the Rashba effects at metallic interfaces may strongly affect  $H_{\rm FL}$  in chemically asymmetric PtlColAllPt stacks (see Figures 2 and 3).

We then performed SOT-driven magnetization switching experiments on Pt(8)|Co(0.9)|Al( $t_{Al}$ )|Pt(3) Hall cross-bars. A magnetic field  $H_x$ , either positive or negative, is applied along the current direction to break the mirror symmetry, and  $R_{\rm AHE}$ is measured vs the current. Typical results of SOT-induced magnetization reversal are shown in Figure 5 for respective positive  $\mu_0 H_r = +250, 373, 495, \text{ and } 608 \text{ mT}$  (Figure 5a) and equal negative  $\mu_0 H_x = -240$ , -368, -495, and -600 mT (Figure 5b) fields for  $t_{Al} = 3$  nm. A complete magnetization reversal is observed for  $J_c \approx \pm 2 - 2.5 \times 10^{11} \text{ A} \cdot \text{m}^{-2}$  with the polarity of the magnetization reversal cycle depending on the sign of  $\mu_0 H_x$  as expected. We also find that the critical current density decreases as  $|\mu_0 H_x|$  increases as observed in refs 35 and 41 with the typical current density reaching  $J_c = 3-3.5 \times 10^{11}$  $A \cdot m^{-2}$ . In Figure 5c, we compare magnetization reversal cycles for different  $t_{A1} = 1, 2, 3$ , and 4 nm, acquired under a field  $\mu_0 H_x$ = +250 mT much smaller than the anisotropy field  $H_K$  (SI-IV). Despite a larger  $H_K$  and larger coercive field, we find that a complete switching of magnetization (between P and AP states) may be achieved in devices with  $t_{Al} = 3$  and 4 nm, whereas the ones with  $t_{Al} = 1$  and 2 nm exhibit only partial reversal, thus in agreement with the enhanced SOT shown in



**Figure 5.** Current-induced magnetization reversal in Pt(8)|Co(0.9)| Al(3)|Pt(3) for different (a) positive and (b) negative  $\mu_0 H_x$ . (c) R<sub>AHE</sub> as a function of pulsed current in the Pt(8)|Co(0.9)|Al( $t_{Al}$ )|Pt(3) series of samples for  $\mu_0 H_x = +250$  mT. The pulse width was 100  $\mu$ s.

Figure 3. We hence conclude that increasing the Al thickness in the  $Pt(8)|Co(0.9)|Al(t_{Al})|Pt(3)$  series increases the SOT allowing a reduction of the critical current density for complete magnetization switching by more than 30%.

The ensemble of the results highlighting the existence of a large interfacial Rashba effect from the ColAl is supported by our modeling (SI-VII). In the case of thick ferromagnet, it is known that SOT from SHE is determined via the complex spin-mixing conductance.<sup>21,42,43</sup> For  $t_{\rm Co}$  smaller than the dephasing length, the description is far more complex owing to the remaining outgoing spin-currents and additional electronic reflections. We developed numerical analyses gathering most of the ingredients discussed above based on generalized drift-precession-diffusion equations<sup>20,44-46</sup> able to express the profile of the vectorial spin-accumulation  $\hat{\mu}_{F}$ . We thus provide subsequent insights for  $H_{\rm DL}$  and  $H_{\rm FL}$ , the integrated value of the torque  $au_{
m SOT}$ , by considering the precessing and dephasing processes via the s-d exchange coupling  $(\mathcal{J}_{xc})$ . In particular, this implies a complex value of the propagating length in Co. The SOT acting on the Co layer by the out-of equilibrium spin  $\hat{\mu}_{F}$  then writes:<sup>4</sup>

$$\boldsymbol{\tau}_{\text{SOT}} = \int_{\mathcal{V}} \frac{\mathrm{d}\hat{\mathcal{M}}}{\mathrm{d}t} \mathrm{d}\mathcal{V} \approx \int_{\mathcal{V}} \frac{\hat{\boldsymbol{\mu}}_{F} \times \hat{\mathbf{m}}}{\tau_{J}} \mathrm{d}\mathcal{V}$$
(3)

where  $\mathcal{V}$  is the volume of Co and  $\mu_F$  is expressed in the same unit  $(\mu_B/\mathcal{V})$  as  $\hat{\mathcal{M}}$  and  $\tau_J = \frac{\hbar}{\mathcal{J}_{xc}}$  is the precession time. In our treatment, the interfaces are described by their proper interfacial  $G_s$  or spin-mixing  $G_{\uparrow\downarrow}$  (between Co and the nonmagnetic material) conductances normalized by their Sharvin values  $G_{sh}$ , or using corresponding transmission coefficients (spin-mixing transmission). It generally leads to possible spin-backflow processes<sup>48</sup> and additional spinaccumulation within the different layers. We considered:

(i) a typical precession (or Larmor) length  $\lambda_J = 2\pi v_F \tau_J = 4.7$  nm for the transverse part of the spin accumulation and spincurrent and a transverse spin-decoherence length  $\lambda_{\perp}$  of 1.7 nm in the range of the values given in ref 45 for Co.

(ii) a large spin-mixing transmission coefficient  $\mathcal{T}_{\uparrow\downarrow}^{\text{ColPt}} = \frac{G_{\uparrow\downarrow}^{\text{ColPt}}}{G_{\text{sh}}} \approx 0.8$  at the ColPt interface<sup>40,49</sup> with  $G_{\uparrow\downarrow}$  and

 $G_{\rm sh}$  being, respectively, the spin-mixing conductance and the Sharvin conductance.

(iii) a mean spin-mixing transmission coefficient  $\mathcal{T}_{\uparrow\downarrow}^{\text{ColCulPt}} = \frac{G_{\uparrow\downarrow}^{\text{ColCulPt}}}{G_{\text{sh}}} \approx 0.5 \pm 0.05 \text{ through Cu.}^{36,37}$ (iv) a mean spin-mixing transmission coefficient

 $\mathcal{T}_{\uparrow\downarrow}^{\text{ColAllPt}} = \frac{G_{\uparrow\downarrow}^{\text{ColAllPt}}}{G_{\text{sh}}} \text{ varying from 0.8 to 0 when the Al thickness}$  is increased from 0 to 3 nm and greater.

Based on our present knowledge for the ColPt inter-

face, <sup>36,49,50</sup> the predicted SOT fields are in excellent agreement with the experimental ones for both DL and FL (see dashed lines in Figures 2 and 3). We accounted for a spinaccumulation  $\mu_F = \mu_{REE}$  at the ColAl(Cu) interface due to REE. For PtlColCulPt, we consider that  $\mu_{REE}$  is *homogeneous* in Co and constant for all  $t_{Co}$ . We quantify that REE (SHE) contributes  $\approx 45\%$  to the FLT ( $\approx 55\%$ ) for  $t_{Co} = 0.4$  nm in the reference Cu samples. The important result is that, for the Ptl ColAllPt series, a much larger spin accumulation  $\mu_{REE}$  is required to fit the data with a certain dependence on  $t_{Co}$ varying within a length scale of the order of 0.75 nm. Consequently, the FLT contribution from REE (and/or ORE) reaches  $\approx 75\%$  for  $t_{Co} = 0.55-1.2$  nm ( $\approx 25\%$  from SHE), whereas it contributes only 60% for  $t_{Co} = 1.4$  nm ( $\approx 40\%$  from SHE).

Finally, we point out that, at this stage, and owing to the same symmetry of torques possibly generated by an ORE<sup>13</sup> compared to the one by spin Rashba effect, we cannot distinguish between the two mechanisms and thus cannot discard that a large part of the FLT increase in Al-based samples may indeed originate principally from ORE. Note, however, we can discard the role of the OHE in the Al light element (nonetheless calculated to be small<sup>30,51</sup>) from our results displayed in Figure 4 on the basis of the same arguments discussed for the spin Hall effect.

In conclusion, we have demonstrated that, in atomically thin Co, the SOT amplitudes, both for damping- and field-like symmetries, vary significantly when a light element such as Al is deposited on top of Co, exceeding the values existing in the literature. In particular, we find a large increase of the field-like component and the corresponding ratio of  $H_{\rm FL}/H_{\rm DL}$  at ColAll Pt interfaces within the single atomic Co thickness range. This demonstrates the occurrence of interfacial Rashba interactions, leading to significant conversion effects at the ColAl interface. Beyond the fundamental interest, these results in all-metallic systems in the limit of atomically thin ferromagnets also open a new research direction for CMOS-compatible and cost-effective spinorbitronic based technology.

#### ASSOCIATED CONTENT

#### **Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.2c05091.

(Detailed analysis of harmonic Hall measurements, measurements of resistivity and estimation of current shunt effects in the each layer of multilayers, theoretical modeling of anomalous Hall effect and spin-orbit torques PDF)

#### AUTHOR INFORMATION

#### **Corresponding Authors**

- Vincent Cros Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France;
  orcid.org/0000-0003-0272-3651; Email: vincent.cros@cnrs-thales.fr
- Henri Jaffrès Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France; Email: henri.jaffres@cnrs-thales.fr

#### Authors

- Sachin Krishnia Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France
- Yanis Sassi Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France
- Fernando Ajejas Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France; Present Address: Department of Physics and Center for Advanced Nanoscience, University of California, San Diego La Jolla, CA, 92093, USA
- Nicolas Sebe Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France
- Nicolas Reyren Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France; orcid.org/0000-0002-7745-7282
- Sophie Collin Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France
- Thibaud Denneulin Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons (ER-C 1) and Peter Grünberg Institut (PGI-5), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany
- András Kovács Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons (ER-C 1) and Peter Grünberg Institut (PGI-5), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany; © orcid.org/0000-0001-8485-991X
- Rafal E. Dunin-Borkowski Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons (ER-C 1) and Peter Grünberg Institut (PGI-5), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany; Octid.org/0000-0001-8082-0647
- Albert Fert Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France
- Jean-Marie George Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France

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#### Notes

The authors declare no competing financial interest.

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