Unconventional anomalous Hall effect in 3d/5d multilayers mediated by the nonlocal spin-conductivity.

T. Huong Dang, Q. Barbedienne, A. Jouy, N. Reyren, F. Godel, S. Collin, J. M. George, and H. Jaffrès

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We evidenced unconventional Anomalous Hall Effects (AHE) in 3d/5d (Co0.2nm/Ni0.6nm)N multilayers grown on a thin Pt layer or thin Au:W alloy. The inversion observed on AHE originates from the opposite sign of the spin-orbit coupling of Pt compared to Ni. Via advanced simulations methods for the description of the spin-current profiles based on the spin-dependent Boltzmann formalism, we extracted the spin Hall angle (SHA) of Pt and (Co/Ni) as well as the relevant transport parameters. The extracted SHA for Pt, +20%, is opposite to the one of (Co/Ni), giving rise to an effective AHE inversion for thin (Co/Ni) multilayers (N < 17). The spin Hall angle in Pt is found to be larger than the one previously measured in combined spin-pumping inverse spin-Hall effect experiments in a geometry of current perpendicular to plane. Whereas magnetic proximity effects cannot explain the effect, spin-current leakage and anisotropic electron scattering at Pt/(Co,Ni) interfaces fit the experiments.

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Recently, the field of spinorbitronics has emerged as a new route to generate spin-currents able to excite small magnetic elements [1–3] or to move domain walls [4–6]. This is made possible via the so-called intrinsic spin Hall effect (SHE) [7–9] provided by heavy metals, e. g. Pt [10–15], Ta [2, 3] and W [16] or via the extrinsic SHE of metallic diluted alloys [17–22]. SHE [24] borrows its concept from the well-established principles of the anomalous Hall effect (AHE) [26] whereby the relativistic spin-orbit interactions (SOI) may promote an asymmetric deflection of the electron flow depending on their spin. Early studies of AHE mostly deals with bulk ferromagnetic (FM) metals [27] and their alloys [28–30]. With the fast development of spinorbitronics, AHE now starts to be largely investigated in ultrathin multilayers such as Co/X (X=Au [35], Pd [36–41], Pt [42], or more recently Co/Ni [43]) investigated for its perpendicular magnetic anisotropy (PMA) properties often required for devices [44].

AHE and SHE involving 3d transition metals (Fe, Co, Ni) or 5d noble metals are presently the basis of numerous fundamental investigations dealing with an intrinsic mechanism originating from the Berry phase [27, 45, 46], skew-scattering [28, 31–34] or side-jump [29, 31] extrinsic phenomena. Less works dealt with multilayers wherein interfaces may bring new insights in the spin-orbit assisted scatterings. Examples of such important roles of interfaces are the magnetic proximity effects appearing at the scale of a few atomic planes [47–49], and the spin-current depolarization at 3d-5d interface by local SOI or spin-memory loss (SML) as revealed in several sets of recent papers [12, 13, 50–54]. Regarding spin-current depolarization, another recent matter of debate is the value of the spin Hall angle (SHA) of 5d heavy metals such as Pt including both disorder [45] and SML at interfaces once considering the non-local spin conductivity in multilayers [55]. Like AHE, the spin-dependent SHE properties are scaled by the off-diagonal spin-dependent conductivity tensor \( \sigma_{xy} \) involving either intrinsic \( \sigma_{xy}^{int,s} \) and extrinsic skew-scattering \( \theta^{sk,s}\sigma_{xy}^{s} \) and side-jump \( \sigma_{xy}^{s, sj} \) contributions [24, 25]):

\[
\sigma_{xy}^{s} = \theta^{sk,s}\sigma_{xy}^{s} + \sigma_{xy}^{int,s} + \sigma_{xy}^{s, sj}
\]

where \( s \) are the \( \uparrow, \downarrow \) spin index, the subscripts \( (sk),(sj),(int) \) denote respectively the extrinsic skew scattering, extrinsic side-jump and intrinsic terms [25, 56]. The expression for the off-diagonal conductivity responsible for AHE, \( \sigma_{xy} \), is the sum of the two spin channels with \( \sigma_{xy} = \sigma_{xy}^{\uparrow} + \sigma_{xy}^{\downarrow} \). AHE and SHE in 3d ferromagnetic materials, as in Co [57, 58] and Ni [59, 60], are mainly expected to possess an intrinsic origin with opposite sign of \( \sigma_{xy}^{int} \) as experimentally determined and calculated [62–65]. The correspondence between SHE and AHE in ferromagnet has been recently debated [32, 33].

In this letter, we present unconventional results of non-local AHE in a series of Pt/(Co0.2nm/Ni0.6nm)_N and Au:W/(Co0.2nm/Ni0.6nm)_N multilayers (MLs) involving different numbers of (Co/Ni) sequences and corresponding interfaces. (Co/Ni) is known to possess a specific interface anisotropy exhibiting PMA [44, 66, 67] also involving Dzyaloshinskii-Moriya (DMI) interactions [68, 69]. By taking advantage of the relatively small AHE of (Co0.2/Ni0.6)_N MLs and when \( N < 17 \), we demonstrate an AHE sign change in structures grown...
on Pt. We used an advanced Boltzmann analysis algorithm for the necessary determination of the spin-currents with adequate boundary conditions at interfaces and based on the extension of the Fuchs-Sondheimer approach [74]. It is based on the combination of electron scattering in multilayers [78] and SOI-assisted spin and charge deflection inside layers. The different contributions have been carefully addressed involving off-diagonal spin-flip terms in the diffusive potentials. We thus demonstrate that the AHE sign inversion originates from the non-local properties of the spin-conductivities [70, 71] more than induced magnetization in Pt (magnetic proximity effects or MPE) like recently invoked [47–49]. Physical mechanisms of non-local AHE effect in those systems relies thus on the combination of the SOI-dependent scattering of a polarized current generated in (Co/Ni) and subsequent ISHE process in bulk adjacent heavy metal (Pt, Au:W). Our experiments demonstrate an opposite sign of the SOI between Pt and (Co0.2/Ni0.6) or Ni in AHE phenomena. This also reveals a characteristic large positive SHA (+20%) for Pt, at Co/Pt interface much stronger [13, 53, 55] than determined previously in combined spin pumping-ISHE experiments [12] ; and that we assign to an anisotropy in the scattering time. By choosing consistent physical parameters, we find an excellent agreement with the experimental trends.

Samples preparation and measurements: Samples are deposited on thermally oxidized Si wafers at room temperature using magnetron sputtering. They are made of a 6-nm-thick heavy metal layer, Pt or Au:W alloys, covered by a magnetic multilayers composed of N repetitions of (Co0.2 nm/Ni0.6 nm) bilayers. Such (Co/Ni) stack is used to keep a large perpendicular magnetic anisotropy, about constant at least up to N ≈ 40 or more [44]. The samples are finally covered with a 5-nm-thick Al layer to prevent oxidation of the (Co/Ni) stack. Pt and Au:W buffer layers possess the same or opposite spin-Hall angle (SHA) depending on the W content in Au:W and depending thus of its resistivity. According to our recent published work [22] the SHA is counted positive, that of the same sign than that of Pt, for an Au:W alloy resistivity (ρ) typically less than 110µΩ·cm and, negative (same sign than Ni or (Co0.2/Ni0.6) for ρ > 130µΩ·cm. The bulk resistivity of Pt equals ρ_{Pt} = 17µΩ·cm whereas, in the present case, the one of Au:W is ρ varying from 80 to 130µΩ·cm. We denote Au:W_ρ the Au:W alloy of resistivity ρ in µΩ·cm. Devices are then patterned into Hall cross bars of different widths ranging from 3 to 6 µm and of 600 µm length by optical UV lithography and Ar ion etching process.

Fig. 1 displays our main results AHE that we have identified. Data are acquired at room temperature (RT) but the main trends remain for experiments led at low temperature. Panel 1b shows the transverse resistance measurements, i.e. the AHE of the Pt6/(Co0.2/Ni0.6)20 device (with N=20). AHE characteristics of amplitude ΔR_{AHE}=−6 mΩ has to be associated to a negative sign following the general convention of negative AHE of Ni (compared to Co and Pt). This is also true for (Co/Ni) multilayers when Ni, with a larger intrinsic AHE, is thicker than Co and when N is sufficiently large for (Co/Ni)N to dominate the conduction (negligible shunt in the buffer layer). In that spirit, the same conclusions holds for the samples grown on the 6 nm Au:W_{130} buffer layers for N=40 (panel 1d) corresponding to a large N and minimum current shunt in the buffer layer. More intriguing is the behaviour of the device made of Pt with N = 3 (panel 1a). This figure displays a clear positive AHE signal, ΔR_{AHE} ≃ +55 mΩ, emphasizing thus a clear opposite sign compared to the reference Pt6/(Co0.2/Ni0.6)20 sample with large N. This has to be related to an apparent opposite sign of the SOI in that particular sample. When the thin 6 nm Pt buffer is changed into a Au:W_{130} alloy, no sign change is observed keeping the overall AHE signal negative at small N=5 (panel 1c) whatever the resistivity ρ = 80−130µΩ·cm (refer to Fig. 2 for ρ = 80 µΩ·cm).
The whole experimental results for the two series of samples and for both AHE resistances (or resistivities) as well as longitudinal resistivities, vs. the number sequences \( N \) varying from 3 to 40 are presented in Fig. 2. In those experiments, the structure of the (Co/Ni) bilayers is fixed. Fig. 2a highlights the typical crossover from positive to negative experimental values acquired at RT, for the AHE resistance \( R_{xy} \) in the case of Pt. The cross-over is obtained for \( N \approx 17 \) corresponding to a total (Co/Ni) thickness of about 15 nm. This point indicates an exact compensation of the AHE current provided by Pt (positive) and (Co/Ni) (negative) layers. Fig. 2c gives the same plot in the scale of resistivity \( \rho_{xy} \) whereas Fig. 2d displays the increase of the resistivity from the one of Pt to the one of (Co/Ni) when \( N \) increases to give the bulk value of (Co/Ni). Fig. 2b compares the AHE values obtained with both Pt and Au:W series (Au:W_{80} - purple point- and Au:W_{130} - blue triangle-) compared to the Pt samples. The Au:W_{80} experimental point lies between the corresponding Pt and Au:W_{130} samples because of its intermediate SHA value between the ones of the two materials. One of our main conclusions is that, for thin ferromagnetic stacks (\( N \) small), AHE may be dependent on the heavy-metal buffer often used for PMA properties.

**Model for SHE-AHE and calculations.** How to explain such observations? The spin-current is generated in (Co/Ni) multilayers with a given spin-polarization from the ferromagnetic bulk properties. However, as the number of sequences \( N \) for (Co/Ni) (\( N = 3 - 5 \)) remains small, the current partly spin-polarized, occurs to be dominant in Pt compared to the (Co/Ni) region of reduced thickness [25]. This occurs up to a given threshold limit of \( N \) above which the conduction becomes dominant in (Co/Ni) like in the case of Pt/(Co/Ni)_{20} and Au:W_{x}/(Co/Ni)_{40} (Fig. 1). The semi-phenomenological theory of current-in-plane (CIP) spin-currents [78] indeed shows that (Co/Ni) is able to provide the necessary polarized current within the whole stacks Pt. The existence of such spin-polarized proximity current is converted into a transverse current via the local SOI and ISHE. We thus demonstrate that Pt possesses a positive spin Hall angle [12, 22] while (Co/Ni) with thicker Ni possesses a negative SHE sign. This is corroborated by following modeling and simulations presented in that second part.

In order to retain the main physical principles driving the SHE and AHE in MLs with Pt related interfaces, our idea is to distinguish the four possible different mechanisms of AHE in (Co/Ni): (i) an intrinsic AHE-SHE phenomenon in (Co/Ni) viewed as an effective material and characterized by an average spin-Hall conductivity (SHC=\( \sigma_{xy}^{int} \)), a pure extrinsic SHE mechanism acting either on (ii) the majority or (iii) the minority spin channels with an overall extrinsic SHA given by \( \theta_{eff} = \frac{\theta \sigma_{s} \sigma_{s}}{\sigma_{s} + \sigma_{p}} \) [25]. A larger majority spin-current is expected (\( \sigma_{p} > \sigma_{s} \)) whereas a larger SHA is expected in the spin minority band (\( \langle \theta \rangle < \langle \theta \rangle \)) by enhanced sp-d band mixing and necessary phase shift for skew-scattering phenomena [72, 73] giving uncertainties between scenario (ii) and (iii) for extrinsic mechanism like suggested in Ref. [30, 61]. In that sense, our approach is slightly different from considering an identical SHA for both spin channel [34]. The last scenario (iv) is the one of magnetic induced moment in Pt (MPE) generating spin-currents and AHE in Pt close to the Co interface at the scale of a few (typically 2) atomic planes [47–49].

Apart from spin-dependent electronic diffusions in bulk, one may emphasize the relevant boundary conditions to match for the out-of-equilibrium Fermi distribution in the framework of Fuchs-Sondheimer model [74]. This is generally performed by including possible specular [75, 76] or diffusive electron reflection (R)/transmission(T) at interfaces [25] in the CIP spin-dependent Boltzmann equations involving layer- and spin-dependent electronic mean free path \( \lambda_{s} \). One also has to consider the corresponding SOI spin-mixing terms in a 2 \( \times \) 2 Pauli matrix form and related spin-flip probability [77]. This is particularly true at the Co/Pt interface where the spin-loss is known to be large. It is parameterized, here, by a spin-flip coefficient \( p_{sf} \) related to the spin-memory loss (SML) \( \delta \) parameter [12] according to \( p_{sf} = 1 - \exp(-\delta) \). SML at 3d – 5d interfaces plays unavoidable role in spin-pumping in FMR experiments [12, 53]. Moreover, one introduces the overall longitudinal resistivity \( \rho_{xx}^{s} \) (or conductivity \( \sigma_{xx}^{s} \)) and transverse resistivities \( \rho_{xy}^{s} \) (or transverse conductivity \( \sigma_{xy}^{s} \)) of the MLs as:

\[
R_{xx} = \rho_{xx}^{s} \frac{L}{Wt} \approx \frac{L}{W t \sigma_{xx}^{s}} = \frac{L}{W \sum_{i,s} \sigma_{xx,i}^{s} t_{i}}, \quad (2)
\]

\[
R_{xy} \approx \frac{\rho_{xy}^{s}}{t} = \frac{\sigma_{xy}^{s}}{(\sigma_{xx}^{s})^2} = \frac{\sum_{i,s} (\sigma_{xy,i}^{s} t_{i})}{(\sum_{i,s} \sigma_{xx,i}^{s} t_{i})^2}, \quad (3)
\]

where \( L, W \) represents the length and width of the Hall cross bars, \( t \) is the overall thickness of MLs and \( \sigma_{xx,i}^{s} \) the local longitudinal spin-conductivity of the \( i^{th} \) layer of thickness \( t_{i} \) and \( \sigma_{xy,i}^{s} \) the local off-diagonal spin-conductivity in the layer \( i \).

Two different cases may be distinguished according to the (i) extrinsic or (ii) and (iii) the intrin-
sic nature of the AHE into play. For the case (ii) and (iii) $\sigma_{xy,i}$ may be expressed as $\sum_s \theta^i_s \sigma^s_{xx,i}$ for both ferromagnetic and normal metals with $\theta^i_s$ the local spin Hall angle of layer (i) for the s-spin channel [25, 30]. One has $\theta^i_s = -\theta^i_{s'}$ for non-magnetic materials whereas no equivalent relationship exists for a ferromagnet because of the spin-degeneracy lift making $\theta^i$ and $\theta^i_s$ different in absolute value [30].

However, one may generally assume that $\theta^i_s$ and $\theta^i$ are of opposite sign. For those calculations [25], the current density for the $s$-spin channel in the MLs is calculated via the relationship:

$$J^s(z) = -\frac{e}{2} \int_S v_x g^s(z, v_z) d^3v$$

where $g^s(z, v_z)$ are the out-of-equilibrium Fermi distributions for spin $s$, solution of the Boltzmann equation within the MLs, $v_{x,z}$ the Fermi velocity along the current direction ($x$) or along the perpendicular ($z$) to the layers and $S$ the section. $g^s(z, v_z)$ possess two components, one for to the bulk and the other decreasing in $z$ related to the spin-dependent scattering at interfaces that should be found self-consistently. After integration of Eq. 4, one has access to $\sigma^s_{xx,i}$ and $\sigma^s_{xy,i}$ respectively. The transverse current is calculated by considering the local transverse conductivity and by summing all contributions.

**Data fitting by numerical methods:** On the basis of the aforementioned arguments of extrinsic vs. intrinsic SHE mechanism in (Co/Ni) we have proceeded to the four different fitting procedures for AHE in the Pt and AuW series and retained the best fit given on Fig. 2a-b with the same set of parameters. In the present case, SML is taken into account in the interfacial scattering matrix at each $Pt/(Co)$ (with $\delta = 0.9$ [12, 52, 53] or equivalently $\rho_f = 0.6$) and (Co/Ni):$(\delta = 0.25$ or equivalently $\rho_f = 0.3$) interfaces as given in Ref. [79]. Details of the extended CIP and the $S$-scattering formalism treating the case of MLs are given in [25]. The fits have been obtained with a SHA for Pt equal to $\theta^i_{Pt} = \theta^i_{Co} = +20 \pm 2\%$, $\theta^i_{AuW80} = +10 \pm 1\%$ and $\theta^i_{AuW130} = -0.3 \pm 0.1\%$ whereas the different models yield (see table 1 of SI):

- (i) intrinsic SHE mechanism in (Co/Ni) giving $\sigma_{xy}^{intr} = -85 S.cm^{-1}$;
- (ii) extrinsic SHE mechanism in (Co/Ni) giving $\theta^i_{(Co/Ni)} = -0.9\% (\theta^i_{(Co/Ni)} = 0)$ (blue curve);
- (iii) extrinsic SHE mechanism in (Co/Ni) on the minority spin-channel effect giving $\theta^i_{(Co/Ni)} = -2.2\% (\theta^i_{(Co/Ni)} = 0)$ (magenta curve).

The conductivity for (Co/Ni), $\sigma_{xy}^{int}$, should be compared to the extrinsic one with balanced spin-Hall effect on both spin up and spin down channel $\theta^i_{(Co/Ni)} = -\theta^i_{(Co/Ni)} = -1.5\%$ with the corresponding relationship $\sigma_{xy}^{int} \simeq \theta^i_{(Co/Ni)} \sigma_{xx}$ with $\theta^i_{(Co/Ni)} = 1\%$ whereas the different fits have been obtained with a SHA for Pt equal to $\theta^i_{Pt} = \theta^i_{Co} = +20 \pm 2\%$, $\theta^i_{AuW80} = +10 \pm 1\%$ and $\theta^i_{AuW130} = -0.3 \pm 0.1\%$ whereas the different models yield (see table 1 of SI):

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extrinsic vs. intrinsic models/simulations for AHE concerning (Co/Ni) for that range of \( N = 3 \sim 40 \) and bilayer thicknesses, two major conclusions may be raised. The first one is that, in any case, the consistent spin Hall angle of Pt, +20\%\), is observed to be clearly enhanced compared to its value extracted from spin pumping-ISHE experiments [12]. Such enhancement of the spin-Hall angle in Pt has been already observed in STT-FMR experiments [13, 50, 51, 53], lateral spin-valve (LSV) geometry [81] as well as Spin-Hall magnetoresistance experiments [52, 53, 55, 56]. Such enhancement of the spin-Hall angle value extracted from spin pumping-ISHE experiments observed to be clearly enhanced compared to its consistent spin Hall angle of Pt, +20\%, is opposite to the one of (Co/Ni), giving rise to AHE inversion for thin (Co/Ni) multilayers. The large SHA cannot be explained by magnetic proximity effects, and is found to be larger than previously measured in spin pumping-ISHE experiments, effect that we can understand by considering the anisotropy in the electron scattering time in the multilayers. Moreover, we can conclude that the AHE effect can probe main properties of the interfacial spin-orbit interactions appended by heavy metals.

In conclusions, we evidenced inverted anomalous Hall effect in (Co/Ni) based multilayers grown on thin Pt buffer via spin-polarized transport proximity effects. Using advanced simulation methods for the description of the current and spin-current profiles within multilayers, we have extracted the opposite spin Hall angles for Pt and (Co/Ni) and the relevant transport parameters. The extracted SHA for Pt, +20\%, is opposite to the one of (Co/Ni), giving rise to AHE inversion for thin (Co/Ni) multilayers. The large SHA cannot be explained by magnetic proximity effects, and is found to be larger than previously measured in spin pumping-ISHE experiments, effect that we can understand by considering the anisotropy in the electron scattering time in the multilayers. Moreover, we can conclude that the AHE effect can probe main properties of the interfacial spin-orbit interactions appended by heavy metals.

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