Skyrmion Chirality Inversion in Ta/FeCoB/TaO$_x$ trilayers

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Abstract:

Skyrmions are nontrivial spiral spin textures considered as potential building blocks for ultrafast and power efficient spintronic memory and logic devices. Controlling their chirality would provide an additional degree of freedom and enable new functionalities in these devices. Achieving such control requires adjusting the interfacial Dzyaloshinskii-Moriya interaction (DMI). Thanks to Brillouin Light scattering measurements in Ta/FeCoB/TaO$_x$ trilayer, we have evidenced a DMI sign crossover when tuning TaO$_x$ oxidation and suspected another DMI sign crossover when tuning FeCoB thickness. Moreover, using polar magneto-optical Kerr effect microscopy, we demonstrate skyrmion chirality inversion through their opposite current induced motion direction either by changing FeCoB thickness or TaO$_x$ oxidation rate. This chirality inversion enables a more versatile manipulation of skyrmions, paving the way towards multidirectional devices.

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I- Introduction

Magnetic skyrmions [1] are swirling chiral magnetic textures. They have recently garnered much attention since they are considered as potential building blocks for next-generation ultra-fast and power efficient memory and logic devices [2,3]. Initially, skyrmions were observed at low temperature in bulk magnetic materials lacking inversion symmetry such as MnSi [4], FeGe [5] and FeCoSi [6,7]. The bulk-induced symmetry breaking leads to skyrmions with a chiral Bloch domain wall, so called Bloch skyrmions. More recently, Néel skyrmions have been observed at room temperature in heavy metal/ferromagnet/oxide (HM/FM/MO$_x$) [8,9,10] and HM/FM/HM [9,11,12] trilayers. The Dzyaloshinskii-Moriya interaction (DMI) [13,14], which favors noncollinear magnetic moments, together with dipolar energy allows to stabilize skyrmions with a given chirality, i.e. sense of rotation of spins when crossing the domain wall [15,16]. In structures where symmetry is broken due to artificial stacking of different materials, for instance in HM/FM/MO$_x$ and HM/FM/HM with different HM, DMI has an interfacial origin. It is mediated by the conduction electrons at the HM/FM interface (Fert-Levy type DMI) [17,18] and by the presence of a Rashba field at the FM/MO$_x$ interface (Rashba type DMI) [19,20].

Skyrmions can be moved efficiently by spin-orbit torque (SOT) produced in a heavy-metal layer via the spin Hall effect [9,10,21,22,23]. Their undistorted motion with low current densities makes them promising candidates for next-generation power-efficient spintronic devices.

Moreover, the SOT current-induced motion of skyrmions is governed by both the sign of the spin Hall angle of the HM and the skyrmion chirality [9,10,22,24,25]. This chirality is determined by the sign of DMI, which, in HM/FM/MO$_x$ trilayers, depends on the spin orbit coupling in the adjacent heavy metal, the structural symmetry of the magnetic layer [13,14] and the oxidation state of FM/MO$_x$ interface [20,26]. Total DMI in such a trilayer is usually considered to be the sum of the DMI stemming from the two interfaces and is often called effective DMI. For thick enough ferromagnet, they are independent, but may become coupled when ferromagnet thickness is below 2-3 monolayers [20,27]. The same interface with inversion of the stacking order is expected to give the same magnitude of DMI, but with a change of sign. However, this is expected only if the crystalline structure remains the same and film growth direction often breaks this symmetry. For nearly zero initial DMI values, non-equivalent intermixing of top and bottom interfaces due to ion irradiation may result in non-zero DMI [28].

Different material variations have been used to achieve a modulation of the strength and in some cases of the sign of effective DMI constant $D$: variation of the FM thickness [29], modification of the adjacent heavy-metal underlayer in HM/CoFeB/MgO heterostructure [30] and insertion of a Pt wedge layer in Ta/FeCoB/Pt(wedge)/MgO thin films [31]. Very recently, it has been experimentally shown that the total DMI of Cu/CoFe/CoFeO$_x$/Ta and Cu/CoFe/Ta(t)/TaO$_x$/Ta could be changed when the oxide type (either CoFeO$_x$ or TaO$_x$) or Ta thickness is modified [32]. A sign crossover of DMI has been also theoretically predicted in Ir/Fe/O depending on the O coverage [33]. Besides material modification, DMI can also be tuned by other means. For example, we demonstrated a huge 130% increase of DMI by an applied gate voltage, which induced a modification of Rashba field at FeCoB/TaO$_x$ interface in a Ta/FeCoB/TaO$_x$ trilayer [34]. Finally, current injection has also been shown to modify the measured DMI sign and magnitude [35].

However, despite several reports on DMI sign crossover, and theoretical prediction of chirality switch by local heating [36] or picosecond magnetic field pulses [37], a systematic study demonstrating a conclusive
chirality inversion of skyrmions, evidenced by their current induced motion is still missing. Such inversion would add a decisive degree of freedom to control the dynamics of skyrmions.

Here, we present a study of bottom-Ta(3nm)/FeCoB/top-TaOₓ samples and report on the observation of a static DMI sign crossover driven by the top oxide (FeCoB/TaOₓ) interface oxygen content and another crossover driven by the FeCoB thickness. DMI has been measured by Brillouin Light Spectroscopy (BLS). Furthermore, a detailed study of the motion of skyrmions under DC current in the positive, negative and zero DMI regions using polar magneto-optical Kerr Effect (p-MOKE) microscopy, confirms these two DMI sign crossovers that are accompanied with inversion of skyrmions motion and thus of their chirality.

For BLS measurements, we have studied samples of uniform thickness for various thicknesses of the ferromagnet and of the top-Ta, the latter giving rise to various oxidation state at the FeCoB/TaOₓ interface. For p-MOKE measurements, a double wedge sample with varying thicknesses of the top Ta and FeCoB layers (ref [38], and see section II-A) enables us to extensively and independently investigate the effect on DMI of FeCoB thickness and of FeCoB/TaOₓ interface state by tuning the oxidation state x of TaOₓ. This combinatorial approach allows to avoid sample-to-sample fluctuations.

II-Materials and methods

A-Sample preparation and skyrmion nucleation

Our samples consist in substrate/Ta(3)/FeCoB(0.6-1.6)/TaOₓ(0.6-1.0)/Al(0.5) (thicknesses in nm) grown by sputtering deposition on a thermally oxidized Si/SiO₂ 100 mm-diameter wafer. The composition of the ferromagnet is Fe₇₂Co₈B₂₀. Samples with uniform layer thickness were deposited with the substrate facing the target (on-axis deposition) whereas thickness gradients were obtained by shifting the sample with respect to the target (off-axis deposition). For double-wedge samples, the thickness gradient of FeCoB, along x-axis was perpendicular to the following wedge of Ta, along y-axis. After the Ta wedge deposition, the oxide was created by a natural oxidation step (150 mbar oxygen pressure for 10 s). The thickness given in the text for top TaOₓ is in fact the deposited Ta thickness. The 0.5 nm cap layer of Al oxidizes in air and protects the trilayer from further oxidation before depositing a thicker oxide. Afterwards, a layer of HfO₂, a high-k dielectric, was deposited atop the magnetic stack to protect the sample from further evolution. Transparent Indium Tin Oxide (ITO) pads (rectangles of 100 by 800 µm) have been deposited atop as reference locations on the sample.

To create skyrmions for imaging and observe their current-induced motion, we have applied a small perpendicular magnetic field (between 40-400 µT depending on location) after saturation with the same field polarity.

B- BLS measurements
BLS measurements have been performed on uniform thickness samples at room temperature. The magnetic field was applied in the film plane, perpendicular to the laser incidence plane, allowing the probing of spin waves propagating in the direction perpendicular to the applied magnetic field in the Damon–Eshbach (DE) geometry. The applied magnetic field is above the saturation field of the sample, as determined from magnetometry loops. The frequency shift $\Delta f$ between Stokes ($f_S$) and anti-Stokes ($f_{AS}$) frequency lines is analyzed using a $2 \times 3$ Fabry-Perot interferometer (3-300 GHz) and is calculated by $\Delta f = |f_S| - |f_{AS}|$. The offset due to the set-up was evaluated from surface acoustic line positions. The spin-wave vector is given by $k_{sw} = 4\pi \sin(\theta) / \lambda$, where $\theta$ (between 0 and 60°) is the angle of incidence and $\lambda = 532$ nm the wavelength of the incident laser. See also Supplemental Material at [URL will be inserted by publisher], section S.I.a Fig. S1 for [examples of spectra].

C- p-MOKE hysteresis and imaging

p-MOKE measurements have been performed on double wedge samples at room temperature. A p-MOKE magnetometer (NanoMOKE®3 by Durham Magneto Optics Ltd.) was used to measure the hysteresis loops on the double wedge samples, at every 2 mm (or 250 μm for the high-resolution maps) throughout 100 mm sample and a color-map of the remanence (remant magnetization/saturation magnetization*100) was generated. See also Supplemental Material at [URL will be inserted by publisher], section S.II Fig. S3 for [examples of evolution of hysteresis loops along the wedges]. The evolution of the static magnetic properties (saturation magnetization $M_s$, coercivity $H_c$ and magnetic anisotropy) was thus studied as a function of thickness of top Ta and FeCoB layers. A p-MOKE microscope was used for imaging and performing local hysteresis loops (see more details in Supplemental Material at [URL will be inserted by publisher], section S.II Fig. S4 for [examples of hysteresis loops and images]) and observe skyrmion motion under injected current (see section II.D).

D- Current induced motion of skyrmions

Wires have been connected on the ITO pads to inject current. The use of microbonding machine allows to locally break the insulating oxide and create a “point” contact, expected to be of typical diameter 5-25 μm. The pads visible on p-MOKE images are connected to the positive ($I^+$) current source, while the ground current electrode is far from the pad (see Supplemental Material at [URL will be inserted by publisher], section S.III.b Fig. S6 for [a photo of the sample with contact location]). In Fig. 4, the current flows outwards the electrodes as represented by thick white arrows in the corresponding figures. See also Supplemental Material at [URL will be inserted by publisher], section S.III.a for [expected motion depending on chirality, setup details and videos]. The magnetic configuration (stripe domains and skyrmionic bubbles, the latter will further be called skyrmions as they share the same topology) and its current-induced motion is observed using p-MOKE microscope.

III- DMI sign crossover by BLS measurements

The generated remanence map measured under magnetic field perpendicular to the layer plane of the double wedge sample shows perpendicular magnetic anisotropy (PMA), in-plane anisotropy (IPA) and paramagnetic (PM) state, depending on material thicknesses (see Fig. 1a). Such map was explained in our previous study [38]. The transition from PM to PMA to IPA for increasing FeCoB thickness is usual. For
low FeCoB thicknesses, the dead layer and finite size effect on Curie temperature explains the PM region. For increasing FeCoB thickness, the effective anisotropy changes sign from positive, dominated by surface contribution in the PMA region, to negative, dominated by the dipolar contribution in the IPA region. The transition from PM to PMA to PM for increasing top Ta was explained by the increase of dead layers for either too thin Ta, thus overoxidation of FeCoB, or too thick Ta, i.e. underoxidized interface.

At room temperature and under zero applied magnetic field, thermally activated demagnetization leads to the formation of high-density stripe domains in regions close to transition between PMA and IPA regions and between PMA and PM regions [34]. Moreover, on application of a small out-of-plane magnetic field (40 to 400 μT), the stripes transform into micron size skyrmions [38].

As explained previously, the DMI of a trilayer with a ferromagnet inserted between two non-magnetic layers is usually considered to be the sum of the DMI from the two interfaces. In the present study, the bottom Ta/FeCoB interface gives a small, Fert-Levy type DMI [17], which surface value $D_s = Dt$, with $t$ the ferromagnet thickness, is of the order of 20-30 $fJ/m$ [28,39]. By contrast, the top oxide interface, FeCoB/TaO$_x$, is expected to possess Rashba type DMI [19,20,26,34]. Moreover, Rashba-DMI, being linked to the interfacial dipole, is expected to be very dependent on the oxidation state and charge transfer at this FeCoB/TaO$_x$ interface. Thus the oxidation state of this interface will be of primary importance to tune DMI in this system [20,32,33].

In order to explore the effect of varying thickness of top Ta and FeCoB on DMI energy, we have performed BLS measurements. Our convention, similar to ref [34], is that positive (resp. negative) DMI corresponds to right-handed or clockwise (resp. left-handed or anticlockwise) chirality. In the BLS experiment (see section II.B), the frequency difference $\Delta f$ between Stokes and anti-Stokes frequency lines $\Delta f = |f_S| - |f_{AS}|$ (see also Supplemental Material at [URL will be inserted by publisher], section S.I.a Fig. S1 for [examples of BLS spectra]) is linked to DMI: $\Delta f = \frac{2\gamma}{\pi M_S} k_{SW} D$, where $\gamma$ is the gyromagnetic ratio, and $M_S$ is the saturation magnetization of the sample. A plot of measured $\Delta f$ and deduced $D$ as a function of top Ta thickness is shown in Fig. 1b. For the thinner Ta region ($t_{\text{top-Ta}} = 0.6$ nm), which thus corresponds to over-/optimally oxidized region, $\Delta f$ is positive, which results in a positive effective DMI ($D > 0$). For thicker Ta ($t_{\text{top-Ta}} \geq 0.7$ nm), in optimally-/underoxidized region, $\Delta f$ changes its sign and becomes negative which results in a negative effective DMI ($D < 0$).

We also observed that the DMI sign seems to be modified by the thickness of FeCoB layer (see Fig. 1c): for thin FeCoB layer ($t_{\text{FeCoB}} = 1.2$ nm) a positive slope of $\Delta f$ as a function of the wave vector, $k_{SW}$, is obtained, which indicates $D > 0$, as shown by green squares in Fig. 1c. Interestingly, for thicker FeCoB layer ($t_{\text{FeCoB}} = 1.5$ nm) a very small negative slope of $\Delta f$ as a function of $k_{SW}$ is obtained, as represented by the purple circles in Fig. 1c, which leads to $D < 0$. We note here that the error bars of ±100 MHz on the frequency difference $\Delta f$, represented in Fig. 1c, indicates that the very small negative DMI for 1.5nm FeCoB falls within the error bar of BLS measurement. Hence, a complementary method has been used to confirm this second DMI sign crossover, and will be discussed in section IV.

To understand this sign change of $\Delta f$ and effective DMI for both FeCoB and top Ta thickness, we have to keep in mind that the measured (effective) DMI results from the contribution of the bottom Ta/FeCoB interface (which is the same in all samples) and the top interface that may change when oxidation and/or the FeCoB thickness is different. From the literature [28,39], we have taken a bottom surface DMI contribution around $D_s = +30 fJ/m$ and deduced the contribution from top interface in our system and its variation along the two wedges (Fig. 2c and d). For measurements as a function of Ta thickness, this
contribution from the bottom interface is small with respect to the contribution from the top interface, despite having taken an upper limit for the bottom interface contribution. For the variation with FeCoB thickness, the two contributions are of the same order of magnitude. We further discuss in supplemental materials section S.I.b [URL will be inserted by publisher], the comparison of thicknesses between the different series of samples and stress that the combinatorial approach of the double wedge helps avoiding thickness reproducibility issues, inherent to ultrathin film deposition.

First, we discuss the effect of oxidation on the DMI magnitude and sign (Fig. 2a,c), which has been predicted to be dependent on oxidation state at the ferromagnet/oxide interface due to hybridization of 3d orbitals of the transition metal (Fe and Co) with the 2p orbitals of oxygen [33]. Our observations are that DMI is positive for FeCoB/TaOx interface, which is more oxidized and becomes negative for a lower oxidation state, i.e. thicker top Ta. A Rashba mechanism for optimally/overoxidized interface thus leads to positive DMI. The underoxidized FeCoB/Ta/TaOx interface leads to a negative DMI. This latter is consistent with a Fert-Levy origin known to lead to positive DMI in Ta/FeCoB, thus to negative DMI for the inverted stack. However, the effective negative DMI means that the absolute value is larger for the top FeCoB/Ta/TaOx interface. This might be due to an intermixed interface related to the deposition of the heavy metal on top of FeCoB [28] and/or to non-uniform interface likely composed of FeCoB/Ta and FeCoB/TaOx. Similar BLS measurement in Pt/Co/TaOx system gives the same trend (see also Supplemental Material at [URL will be inserted by publisher], section S.I.c Fig. S2 for [BLS spectra of the two systems]), but the curve is shifted by the negative DMI from the bottom Pt/Co interface, which is the main contribution in the effective DMI typically of the order of -1.5 to -2 mJ/m² (-1.3 to -2.2 pJ/m) [8,40,41]. The effective DMI is much smaller and closer to zero in the case of Ta/FeCoB/TaOx, which allows to change its sign more easily.

This sign crossover is similar to the one reported by Arora et al. [32], except that signs are opposite, which might be explained by the different samples. In their study, the ferromagnet is Co rich (Co₉₀Fe₁₀) with no B and with a (111) texture due to the underlayer Cu and the samples are not annealed. By contrast, in our present study the ferromagnet is Fe rich, with the presence of B (Fe₇₂Co₈B₂₀), with a bcc structure obtained after annealing. The presence of B, that should migrate to the bottom Ta layer, or possibly in the top Ta regions, when underoxidized, and the annealing leading to smoother interfaces might thus be critical parameters for DMI.

Second, let us quickly discuss the evolution of DMI with FeCoB thickness for a given oxidation state (Fig. 2b and d). After removing the contribution from the bottom interface, we see that for such slightly underoxidized FeCoB/TaOx interface, its contribution to DMI seems either positive for thin FeCoB or negative for thicker FeCoB.

The mechanism for the sign crossover when varying FeCoB thickness, confirmed in section IV, is not clear and might be due to the fact that the magnetic dead layer for this Ta thickness and oxidation level is relatively large (around 0.4 nm) due to underoxidization [38]. The effective thickness of the ferromagnet is thus lower than the nominal one, used in the text, and the variation of DMI may be due to the beginning of a coupling between the two interfaces [20,27]. Moreover, we may also think of a change in band structure or a stronger contribution of roughness to explain this trend.

IV- Skyrmion chirality inversion
In order to check on the double wedge sample the presence of these DMI sign changes and their effect on domain wall chirality, we have probed the dynamics of skyrmions i.e. velocity and direction of motion under the influence of an injected DC current. Left-handed (resp. right-handed) Néel skyrmion with negative spin-Hall angle (as known for Ta) is expected to give motion along (resp. against) the electron flow [24] (see also Supplemental Material at [URL will be inserted by publisher], section S.III Fig. S4 for [details of this expected motion]). For large skyrmion velocities, a transverse motion adds to the longitudinal one, leading to skyrmion Hall effect [42,43]. In the present case, motion of skyrmions occurs at low current densities and low velocities, in the creep regime, so that we do not observe a significant transverse motion.

Current-induced motion at several locations in zone A, are presented in Fig. 3 (see also Supplemental Material at [URL will be inserted by publisher], section S.III.c Fig. S6 and videos SV1-6 for [p-MOKE videos of current-induced motion of skyrmions]). The locations of these measurements in zone A are represented in Fig. 4a. It is visible that in the region of thick FeCoB and thick Ta (location #2, right part of zone A), the skyrmions move along the electron flow and opposite to the injected current direction (Fig. 3(a-c)). In this region, DMI is expected to be negative from BLS measurements. By contrast, in the region of thinner FeCoB (location #4, left part of zone A, which is close to the region studied in [38]), corresponding to positive DMI, they move along the current i.e. opposite to the electron flow (Fig. 3(d-f)). Similarly, in zone B, for thick FeCoB and thin Ta (location #7, lower part of zone B), skyrmions move along the injected current direction but, due to the proximity to the D ≈ 0 region their velocity is small (see Supplemental Material at [URL will be inserted by publisher], section S.III.e and videos SV7,8 for [more details on skyrmions motion in zone B]). This direction of motion is consistent with our previous studies, which focused on the same material stack with even thinner Ta and showed relatively high pinning [38].

Hence, the observed SOT induced motion of skyrmions in opposite directions in the positive and negative DMI regions, as determined by BLS measurements, clearly demonstrates the inversion of chirality of skyrmions from a right-handed in D > 0 region to a left-handed chirality in D < 0 region. This result is consistent with the BLS sign crossover observed when FeCoB/TaOx oxidation level increases and confirms the BLS sign crossover measured for FeCoB thickness increase.

Interestingly, we have found an intermediate region between these regions with D > 0 and D < 0, where a high density of bubbles is present: except usual Brownian motion, expected for 300K experiments, these bubbles do not move under current, as presented in Fig. 3(g-i) (see also Supplemental Material at [URL will be inserted by publisher], section S.III.f, videos SV3-4 for [p-MOKE videos of this experiment]). We interpret this observation as the effect of strong dipolar interactions between the bubbles which might have two possible chiralities for this D ≈ 0 region and thus are pushed in opposite directions (see also Supplemental Material at [URL will be inserted by publisher], section S.III.e for [discussion on current induced motion in D ≈ 0 region]).

Additionally, to better localize this DMI sign crossover on the double wedge, high-resolution remanence maps of zone A (varying FeCoB thickness) and zone B (varying Ta thickness and thus oxidation) are generated (Fig. 4a and b). On the double wedge, the inversion of skyrmion motion occurs at two positions on the transition, one along the FeCoB wedge (approx. t_{FeCoB}=1.05 nm, and t_{top-Ta}= 0.92 nm) and another one along the TaOx wedge (approx. t_{top-Ta}= 0.8 nm, and t_{FeCoB}=1.2 nm). Again, we stress that the wedge
deposition and uniform thickness deposition of BLS measured samples give different nominal thicknesses for the sign crossover, as explained in Supplemental Material at [URL will be inserted by publisher], section S.I.b.

Finally, the SOT induced dynamics of skyrmions has been explored on several locations to study more quantitatively the effect of DMI magnitude and sign. We measured the motion along FeCoB wedge, in zone A, at five locations: in the negative DMI region (#1-2), in the DMI sign crossover region (#3) and in the positive DMI region (#4-5), as shown in Fig. 4a. The velocity of skyrmions is measured as a function of location (Fig. 4c). A positive velocity corresponds to motion along the injected current direction. When crossing the region where DMI changes sign, the chirality of skyrmions is inverted, and a change in the sign of velocity of skyrmions is observed (see also Supplemental Material at [URL will be inserted by publisher], S.III.f, videos SV1-6 for [p-MOKE videos of these experiments]). For similar current density, the typical velocities at locations # 1 (D < 0) is around -53 $\mu$m/s, whereas it is only about +10 $\mu$m/s at location # 5 (D > 0). The current densities used and observed velocities are of the same order of magnitude as reported by other teams for this system [10] (see also Supplemental Material at [URL will be inserted by publisher], section S.III.c and S.III.d for [detailed discussion about these velocities]).

In our previous measurements in the PMA to IPA region with positive DMI, i.e. even below the lower part of zone B, we observed much more pinning than in the D > 0 PM to PMA transition region, i.e. around the left part of zone A [38]. It thus seems that the higher mobility of skyrmions we report in this paper is specific to this negative DMI region, and not to its proximity to the PMA to IPA transition. Interestingly, in this negative DMI region, skyrmions can be moved with a lower current (0.5 mA, roughly corresponding to 1.3 $10^8$ A/m$^2$ current density at 150 $\mu$m distance from point contact) as compared to the positive DMI region, which makes this region with large mobility and low pinning very promising for applications.

Finally, we have shown large skyrmion mobilities and the control of DMI sign by material parameters, such as ferromagnetic thickness and/or oxidation of top interface. Thus, using lithographic processes to locally pattern regions with different oxidation and/or ferromagnetic thickness would allow the definition of high mobility skyrmion regions with opposite motion direction. Moreover, as oxidation is a key parameter for DMI sign in this system, ion migration controlled by a gate voltage [44] could be further used to dynamically switch DMI and skyrmion chirality.

V- Conclusions

In Ta/FeCoB/TaO$_x$ samples, we have reported a DMI sign inversion depending on FeCoB thickness and TaO$_x$ oxidation rate using Brillouin light spectroscopy. We demonstrate that it also results in a chirality inversion of skyrmions, as observed by the inversion of their direction of spin-orbit torque induced motion using p-MOKE microscopy. The observed DMI sign reversal along the oxidation gradient is understood in terms of the change of mechanism, from Fer-Lee to Rashba when increasing oxidation level. The chirality inversion confirmed by the current induced motion of skyrmions also shows that the intermediate region where DMI is zero leads to no current induced motion, likely due to strong dipolar interaction between skyrmions. The region with negative DMI shows even higher mobility of skyrmions than the previously studied regions of these double wedges. The observed chirality inversion, which enables a desired control on the dynamics of skyrmions, may lead to novel functionalities in the skyrmion based logic, memory and neuromorphic devices and opens perspectives towards gate-controlled chirality.
Fig. 1. (a) Perpendicular remanence map of the Ta(3)/FeCoB(0.6-1.6)/Ta(0.5-1)Ox/Al(0.5) double wedge generated by performing the p-MOKE hysteresis loops every 2 mm. PMA, IPA and paramagnetic or dead layer phase are indicated. In zones A and B skyrmion motion has been studied by p-MOKE microscopy, see text. (b) BLS frequency difference $\Delta f$ at $k_{sw}=20.45$ $\mu$m$^{-1}$ and corresponding DMI energy ($D$) on the right scale vs. top Ta thickness for FeCoB thickness of 1.15 nm, measured on uniform thickness samples. Error bars are for $\Delta f$ data. For thicker Ta, i.e. underoxidized TaOx, $\Delta f$ and thus DMI are negative while for thinner Ta, i.e. optimally/overoxidized interface, $\Delta f$ and DMI are positive. (c) BLS shift frequency $\Delta f$ vs. wave vector ($k_{sw}$) for fixed Ta thickness of 0.8 nm, corresponding to slight underoxidized interface for two FeCoB thicknesses, measured on uniform thickness samples. For the thinner FeCoB ($t_{FeCoB}=1.2$ nm), the slope $\Delta f$ vs. $k_{sw}$ is positive indicating a positive DMI. By contrast for the thicker FeCoB ($t_{FeCoB}=1.5$ nm) the small negative slope indicates a near zero or small negative DMI.
Fig. 2. Schematic cross section of double wedge Ta/FeCoB/TaO$_x$ sample: (a) along the y-axis, the FeCoB nominal thickness is constant and the top Ta thickness is varying. The wedge of Ta leads to over-oxidized, optimally oxidized and underoxidized regions along the wedge. (b) Along the x-axis, the oxide thickness is constant and the FeCoB layer has a thickness gradient. (c-d) Variation of the total DMI, measured by BLS on uniform samples (same data as in Fig 1b and c), bottom DMI, taken from the literature and deduced top DMI as a function of Ta thickness (c) and FeCoB thickness for underoxidized TaO$_x$ (d). The red lines in a,b show the corresponding location on the double wedge cross section of the uniform samples used for BLS measurements.
Fig. 3. p-MOKE microscopy images taken under 400 µT perpendicular field while applying a DC current of 5 mA and captured at 0, 0.5 and 1 s time intervals, demonstrating SOT induced motion of skyrmions: (a-c) in the negative DMI region (D < 0) where skyrmions move opposite to the injected current direction, (d-f) in the positive DMI region (D > 0) where skyrmions move along the injected current direction and (g-i) in the DMI sign crossover region (D ≈ 0), where no detectable movement of bubbles is observed, except usual Brownian motion. The direction of injected current is shown by the white arrows. The red circles and arrows highlight the typical behavior of skyrmions in the different regions. The edges of the large ITO pads are visible (right part of images a-f, left part of images g-i). These pads are used to locate the positions and inject current in the trilayer with a point contact through the thick HfO$_x$ oxide (connecting wire giving blurry black/grey/white contrast on the ITO pad). Locations #2, #3 and #4 are the same as in Fig. 4.
Fig. 4. High resolution remanence map of A (a) and B (b) zones, (as defined in Fig.1), generated by performing the p-MOKE hysteresis loops measurements on the sample. The sign of DMI is deduced from the direction of motion of skyrmions under current. (c) Skyrmion velocity under 5 mA, measured at locations #1-2 in the area of negative DMI energy (D < 0), location #3 in the area where DMI is close to zero and locations #4-5 in the area of positive DMI energy (D > 0). A positive velocity corresponds to motion of skyrmions away from the top contact and along the injected current direction. The thick dashed lines in a-c represent an estimated location where the direction of skyrmion motion is inverted. The error bars mainly come from the statistical dispersion of velocities measured on several skyrmions.

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Notes
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Supplementary materials “Skyrmion Chirality Inversion in Ta/FeCoB/TaO_x trilayers”

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S.I BLS spectroscopy measurements:

I.a Examples of BLS spectra

We show in Fig. S1 the BLS spectra taken at incidence angle of 60°, corresponding to k_{sw}=20.45 μm^{-1} for Ta/FeCoB/TaO_x with varying top Ta thickness.

![BLS Spectra](image)

Fig S1. (a) BLS spectra (symbols) for Ta/FeCoB/TaO_x samples for varying top Ta thickness taken at k_{sw}=20.45 μm^{-1} corresponding to the points in Fig 1.b in the main text. The solid lines are Lorentzian fits to the spectra. (b) Zoom in the positive frequency region, close to the anti-Stokes resonance. The thin solid lines are the Lorentzian fits of anti-Stokes resonance and the thick solid lines are the fits of the Stokes resonance with opposite frequencies to better show the frequency difference between Stokes and anti-Stokes resonances.

I.b Discussion about thicknesses
The samples measured with BLS were deposited with a constant thickness (on-axis sputtering). By contrast, a sample with thickness gradients (double wedge, off-axis sputtering deposition) was used for p-MOKE microscopy in order to avoid reproducibility issues. The uniform thickness was monitored with deposition time after a calibration for 30 nm thick films. Thus the real thickness cannot be precisely compared to the ones of the double wedge: a lack of reproducibility of the thickness may occur for such small deposition times. Moreover, the use of on-axis deposition, where the sample is rotating during deposition and off-axis, where it is not, may also introduce differences in terms of film growth and thus of planar anisotropies, and possibly DMI.

We notice that the nominal thicknesses are not consistent between the two sets of measurements of Fig. 2c and d of main text: for 0.8nm of Ta, DMI is either positive for a FeCoB thickness of 1.2nm (Fig. 2d) or negative for a thickness of 1.15nm (Fig. 2c). Given the small DMI values and the very small thickness difference between the two measurements, close to the 5% accuracy given by sputtering deposition, we cannot ensure that the absolute nominal thicknesses are relevant. We will thus only discuss the variations within one given set of samples and then use the combinatorial approach of the double wedge sample to avoid these reproducibility issues.

I.c Comparison of Ta/FeCoB/TaO$_x$ and Pt/Co/TaO$_x$ systems:

We report in Fig. S2, the effective DMI for varying Ta thickness in the Ta/FeCoB/TaO$_x$ (same data as in main text) and in Pt/Co/TaO$_x$ system, that we have deposited under the same conditions. A similar trend is observed Pt/Co/TaO$_x$ for Ta thicknesses between 0.7 and 0.9nm, ie. the frequency difference goes more negative for increasing Ta. In the Pt/Co/TaO$_x$ case, this may be explained by the large Pt/Co contribution to DMI of typically -1.5 to -2 mJ/m$^2$. For this system, DMI reaches a maximum negative DMI of -1 mJ/m$^2$ for Ta 0.9nm. This is probably an indication
that the top Co interface is Co/Ta instead of Co/TaOx, ie. is underoxidized. The results obtained on Ta/FeCoB/TaOx show the same trend.

![Graph showing the evolution of BLS frequency difference effective DMI vs. Ta thickness for Pt(3)/Co(1.2)/TaOx and Ta(3)/FeCoB(1.15)/TaOx samples (thicknesses in nm).](image)

**Fig. S2.** Evolution of BLS frequency difference effective DMI vs. Ta thickness for Pt(3)/Co(1.2)/TaOx and Ta(3)/FeCoB(1.15)/TaOx samples (thicknesses in nm). The error bars are estimated from the uncertainty on Δf measurement and are only slightly larger than the symbols.

### S.II p-MOKE imaging and hysteresis measurements:

A commercial MOKE magnetometer, NanoMOKE®3 by Durham was used to characterize the magnetic properties of the magnetic stack used for this study. The hysteresis loop measurements were performed in the presence of a perpendicular ($H_z$) applied magnetic field. The MOKE laser spot was scanned through the specified positions using a motorized x-y stage at every 2 mm or 0.25 mm distance for low or high resolutions scans, respectively for Fig. 1 and 4 of main text. This type of automated hysteresis measurements enables precise characterization of the magnetic property throughout the 100 mm wafer and any change induced by the variation of FM and HM thickness in our double wedge sample can easily be detected.
The high remanence blue color region in the remanence map corresponds to the PMA region and the low remanence red color region in the thicker side of magnetic layer corresponds to the IPA region. Moreover, an additional zero and noisy remanence region shown in red color in the thinner side of FeCoB magnetic layer corresponds to the paramagnetic or magnetically dead layer region (PM). This double wedge has been slightly shifted in FeCoB thicknesses with respect to the one studied in ref 38 of main text, in order to have the full PMA region on the wafer. Moreover, we notice a difference here because we do not have a full in-plane remanence for IPA region, contrary to ref 38. This is due to the magnetic field orientation that was nearly completely perpendicular to the plane for the present case and with both in-plane and perpendicular to the plane components for ref 38.

Hysteresis loops obtained as a function of FM layer thickness and HM layer thickness, along the red and black line represented on the remanence map in Fig. S3a, are shown in Fig. S3b-c. To get a clear picture of the PMA, PMA to IPA transition and IPA regions, our setup allows to invert the sign between polar and longitudinal Kerr signals, thus inducing an inversion of hysteresis loops in the IPA region. This helps identifying the IPA region with respect to PMA region as shown in Fig. S3c, for thick FeCoB. In the thinner FeCoB region, the loops show only noise and thus correspond to PM/dead layer. For increasing FeCoB thickness, the loops have a square shape indicating a region of perpendicular magnetic anisotropy (PMA). The shape of the hysteresis loops obtained for thicker FM layer shows a (more noisy) inverted loop thus indicating IPA.
**Fig. S3a-c.** (a) Remanence map obtained from MOKE measurement for Ta/FeCoB/TaOx sample, same figure as in main text. High and low remanence regions are represented in blue and red respectively. The PMA anisotropy, IPA anisotropy and the paramagnetic PM/dead layer regions are marked. The black (resp. red) dashed line corresponds to the location of the hysteresis loops along the (b) Ta (resp. (c) FeCoB) thickness. (b) Hysteresis loops along the black dashed line of (a) showing the PM to PMA transition with varying top Ta thickness. (c) Hysteresis loops along the dashed red line in (a) showing the transition from PM to PMA to IPA regions as a function of the FM layer thickness. In all cases, the noisy signal with no loop represents the paramagnetic or dead layer region. The hysteresis loops have been normalized and shifted vertically to show their evolution along the Ta or FeCoB gradient.

Moreover, p-MOKE imaging and hysteresis measurements were performed at the same location using a p-MOKE microscope. An expected square loop with strong PMA was observed in the
PMA region as shown in Fig. S4a. An open butterfly loop was observed in the thermally demagnetized stripe domains region as shown in Fig. S4b and a closed butterfly loop was observed in the high density skyrmions region as represented in Fig. S4c. The p-MOKE images of the same locations are shown in Fig. S4d-f.

Fig. S4a-f. M vs H hysteresis loops measurements (a, b, c) and corresponding p-MOKE images (d, e, f) in the PMA regions showing square loop (a,d), in the stripes region showing an open “butterfly” loop (b,e), and in the skyrmions region showing a closed “butterfly loop” (c,f). In (e) skyrmions of approximate 1 µm diameter are clearly visible in the p-MOKE images (white dots).

S.III Current induced skyrmions motion.

III.a Expected direction of motion under current

Schematics of a Bloch skyrmion, a Néel skyrmion with a right-handed chirality and a Néel skyrmion with a left-handed chirality are shown in Fig S5a-c respectively. Under an injected DC current, the skyrmions experience a spin-orbit torque that leads to an undistorted motion of
The spin-orbit torque ($\tau_{SOT}$) induced by an injected current is equivalent to an effective spin-Hall field ($H_{SHE}$) oriented along the z-axis, and is given by the following equation:

$$\tau_{SOT} = -\gamma_0 \vec{m} \times \left( \frac{h_0 \gamma_{SHE} J_c}{2\mu_0 e M_s t} \vec{m} \times \vec{u}_y \right) = -\gamma_0 \vec{m} \times \vec{H}_{SHE},$$

where $M_s$ is the saturation magnetization, $\vec{m}$ is a unit vector along the magnetization, $\vec{u}_y$ is a unit vector parallel to the spins of accumulated conduction electrons at the HM/FM interface, $t$ is the thickness of FM film, $\gamma_0$ is the gyromagnetic ration, $J_c$ is the applied current, $e$ is the elementary charge and $h$ is the reduced Planck constant.

The direction of $H_{SHE}$ is determined by the sense of rotation of magnetization inside the Néel DWs (references 9, 10 of main text). Thus $H_{SHE}$ leads to a uniform and undistorted motion of skyrmions due to the identical chirality of Néel DWs in the skyrmion. Following the analogy presented by Emori et al., (reference 24 of main text) the skyrmions with a right-handed chirality driven by the spin-orbit torque produced by a HM with a negative spin-Hall angle ($\theta_{SHE} < 0$), like Ta, would move against the electron flow and along the injected current direction and vice versa as presented in Fig. S5e-f whereas a Bloch skyrmion is expected to show transverse motion as shown in Fig. S5a. Intermediate Dzyaloshinskii-walls[1] are expected to present an intermediate motion between these two cases, with a continuous change of the angle. Like for Néel or Bloch wall, their chirality will also be of importance for the motion direction with respect to the current. A small DMI might lead to these Dzyaloshinskii walls of only one chirality and thus give a unique motion direction of the skyrmions.

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Fig. S5a-f. (a) A schematic of Bloch skyrmions in the zero DMI (D=0) region. (b-c) schematics of Néel skyrmions in the positive DMI (D>0) region with right-handed chirality and the negative (D<0) DMI regions with left-handed chirality. The magnetization at the edge of skyrmions is out-of-plane, along z-axis, which gradually reverses its orientation at the center. (d) schematics showing the direction of SOT induced force (black arrow) on a Bloch skyrmion. (e-f) schematics showing the direction of SOT induced force on Néel skyrmions with right-handed (D>0) and left-handed (D<0) chiralities. The direction of an injected current (opposite to electron motion) is shown by the large orange arrow.

III.b Current induced measurement setup, location of contacts

For current induced motion, an ultrasonic wire-bonding machine was used to bond 25 µm diameter Al wires to the sample and the copper pads on a PCB. The location of indium tin oxide (ITO) transparent pads was used to locate the regions of interest on the sample. The contacts pads on the PCB were connected to a Keithley 2400 SMU. A schematic is shown in Fig. S6, superimposed on a picture of the sample. The ground terminal of the current source was connected to the reference
electrode on the sample and the source connector was switched from electrode # 1 to 7 to observe motion of skyrmions at these electrodes as shown in Fig. S6. We checked (see videos SV 1-6) that opposite currents induce opposite motion of skyrmions. In zone A, the electrodes #1-2 were located in D < 0 region and # 4-5 in D > 0 region, however, #3 was located in the D ≈ 0 crossover region. In zone B, the electrode #6 was located in D < 0 region and #7 in D > 0 region.

**Fig. S6.** A schematic of the setup used to study the current induced motion of the skyrmions. Real picture of the sample with visible ITO pads. The red empty circles correspond to the locations numbered in the main paper. The reference electrode for current injection is represented as a black full circle. The current was injected with a Keithley 2400 SMU. Skyrmions show different behavior under positive current between the left and right part of zone A (and between the top and bottom part of zone B) as delimited by the black dashed line. In zone A, on the left, i.e. corresponding to a region with positive DMI, for a positive current, the skyrmions go away from the electrode (black rectangle), as represented on the left of the figure. In zone A, on the right, i.e. a region with negative DMI, the skyrmions move towards the electrode, as represented on the right of the figure. The same trend is observed in zone B: above the black dashed line the skyrmions go towards the electrode and below, the skyrmions go away from the electrode. Along the black line no coherent motion is observed except Brownian fluctuations.
III.c Current induced measurements

Average skyrmion velocity was measured as follows: from the videos, we have extracted the displacement of several skyrmions, typically five, between two images separated by 20 frames, which corresponds to about 1s (videos at 24 frames/s). The location of selected skyrmions were all approximatively 150µm from the center of the contact in order to have the same current density. Error bars in Fig. S7 and Fig. 4 in the main text comes from the statistics on several skyrmions.

We showed in the main text Fig. 4 the direction of motion with +5 mA injected DC current in several locations of zone A. It is governed by DMI sign and spin-Hall angle ($\theta_{\text{SHE}} < 0$ for Ta) as discussed in the main text and in section S.III.a. In the positive DMI region ($D > 0$), the skyrmions have a motion along the current flow and vice versa. Here, we add a measurement of skyrmion velocity at location #2 with varying magnitude of the injected current in the negative DMI region ($D < 0$), see Fig. S7 and the corresponding videos SV 9-12.
Fig. S7. Skyrmion velocity as a function of injected current amplitude at location #2 in the negative DMI (D < 0) region taken at approximatively 150 µm from the contact: the speed linearly increases with the injected current. Error bars take into account statistical spread on several skyrmions.

A roughly linear variation of the velocity of skyrmions with the magnitude of injected current is observed. However, as the current is applied using a localized contact on a full sheet sample, the current lines are not straight. They are going out of the localized contact in a radial way in the proximity of the point contact. The measurement of the velocity of skyrmions with increasing current is thus not very accurate because when the skyrmions come closer to the contact, the current density increases.

The extracted velocities are thus estimations of velocity (see S.III.d) done at approximately the same location with respect to the contact (ie. 150 µm). More precise measurements on patterned
samples would give more accurate velocity versus current density, but this is beyond the present paper scope.

Interestingly, in the negative DMI region (D < 0), skyrmions can be moved with a low current as small as 0.5 mA, which confirms a significantly larger mobility and lower pinning of skyrmions in this region.

In Fig. S6, we may notice that the reference electrode is closer to D < 0 region. However, the larger velocity observed at location #2 cannot be ascribed to the position of the reference electrode since similar difference of velocity between location #2 (D < 0) and the D > 0 region skyrmion velocities was observed when using reference electrodes at other locations, for instance closer to D > 0 region.

Current induced motion has also been checked in region B (see video SV7 for D < 0 at location #6 and video SV8 for D > 0 at location #7). We see a coherent motion of skyrmions only very close to the electrode and they are relatively slow (applied current is -40mA, nearly 10 times larger than for videos SV 1-6). At location #6, (elongated) skyrmions move along electron flow, ie. outwards the electrode (negative current) and at location #7, skyrmions move along the current, ie. towards the electrode (negative current). The motion directions are again consistent with DMI sign measured by BLS. The velocities are small, this might be due to the proximity to the D ≈ 0 transition.

In our previous measurements in the PMA to IPA region with positive DMI, ref 38 in main text, we observed strong pinning: the skyrmions were not drifting but rather stretching under current flow. This region was situated even below the lower part of region B in the present manuscript. On the contrary, close to PM to PMA transition, which corresponds in the present paper to the left part of zone A, skyrmions moved with no distortion. Here, we study a new region with negative
DMI, where we observe higher mobility/lower pinning of skyrmions, if we go far enough from the $D \approx 0$ transition line (ie. above region B, or in the right part of region A). It thus seems that this higher mobility/lower pinning of skyrmions is specific to this new negative DMI region, and not to its proximity to the PMA to IPA transition.

III.d Discussion about the current density

As explained above, we measured skyrmion velocity very close to the point contact with respect to the distance to the reference electrode. The current lines are thus roughly radial close to this point contact, as can be observed by the motion of skyrmions in the videos SV1-2, SV5-6. Using current conservation, we may thus estimate the current density of around $5.3 \times 10^8$ A/m$^2$ for 2 mA applied current at a distance of 150 µm ($\frac{2mA}{4nm \times 2\pi \times 150 \mu m}$). At location #2 ($D < 0$), our obtained velocities are similar to measurements in reference 10 of the main text (they obtained around $+25$ µm/s for $4.5 \times 10^8$ A/m$^2$, we have $-11$ µm/s for $5.3 \times 10^8$ A/m$^2$) that were made in the same nominal system, but with positive DMI.

By using the tabulated resistivities of Ta, Fe and Co, ($R_{\square}(Ta) \approx 44 \Omega/\square$ and $R_{\square}(Fe_{0.9}Co_{0.1}) \approx 95 \Omega/\square$) we find that roughly one third of the current flows through the FeCoB and two thirds in the Ta. If spin-transfer torque would occur with the small current density in the FeCoB, skyrmions would always move in the direction of electrons. This is clearly not the dominant mechanism as in the $D > 0$ region, the skyrmions move against electron flow.

One may also argue that heating may occur, which may thus unpin skyrmions and increase their velocities. Close to the point contact, this may have an impact, but we observe the skyrmions in a
region far from the contact (typically 150 µm) where the current density is small and thus heating is negligible.

**III.e Discussion on current induced measurements for D ≈ 0 region**

As discussed in the main text, at the boundary between D > 0 and D < 0 regions, there is a high density of bubbles: except usual Brownian motion, expected for 300K experiments, nearly no motion of bubbles is observed under current, as presented in Fig. 4(g-i) of main text and in videos SV3-4. In this region, that is intermediate between the two DMI sign regions, we expect a nearly zero DMI. Thus, domain walls close to Bloch walls are expected. In the case of exactly zero DMI, we may expect either Bloch skyrmions with both chiralities or non-skyrmionic bubbles, ie. in which the domain wall is not the same type or chirality along the bubble. Non-skyrmionic bubbles are expected to be distorted by the injected current: a part of the domain wall might move in one direction and another part in another direction, which would lead to an expansion or contraction of the bubble. We do not observe such expansion or contraction, and this hypothesis is thus unlikely.

On the other hand, Bloch skyrmions should show a transverse motion with respect to the current, but in opposite direction for the two possible Bloch chiralities. The presence of these two chiralities, together with the fact that the skyrmions are closely packed, could explain the lack of global motion because of strong dipolar interaction that prevent them to move in opposite directions. In this region, skyrmions are thus probably not homochiral (i.e. a fraction of them is of one chirality and the other part is of the other chirality) and possibly of Bloch type.
III.f Videos of p-MOKE microscope images under DC current

**Video SV1.mp4** p-MOKE microscopy video under 400 µT magnetic field and +5 mA current for location #2 in the negative DMI region (D<0), zone A.

**Video SV2.mp4** p-MOKE microscopy video under 400 µT magnetic field and -5 mA current for location #2 in the negative DMI region (D<0), zone A.

**Video SV3.mp4** p-MOKE microscopy video under 400 µT magnetic field and +5 mA current for location #3 in the zero DMI region (D≈0), zone A.

**Video SV4.mp4** p-MOKE microscopy video under 400 µT magnetic field and -5 mA current for location #3 in the zero DMI region (D≈0), zone A.

**Video SV5.mp4** p-MOKE microscopy video under 400 µT magnetic field and +5 mA current for location #4 in the positive DMI region (D>0), zone A.

**Video SV6.mp4** p-MOKE microscopy video under 400 µT magnetic field and -5 mA current for location #4 in the positive DMI region (D>0), zone A.

**Video SV7.mp4** p-MOKE microscopy video under 400 µT magnetic field and -40 mA current for location #6 in the negative DMI region (D<0), zone B. The image size corresponds to 118 by 73 µm.

**Video SV8.mp4** p-MOKE microscopy video under 400 µT magnetic field and -40 mA current for location #7 in the positive DMI region (D>0), zone B. The image size corresponds to 126 by 99 µm.

**Videos SV9.mp4 to SV12.mp4** p-MOKE microscopy video under 400 µT magnetic field with a varying current (SV9: 0mA, SV10: 2mA, SV11: 5mA, SV12: 10mA) for location #2 in the negative DMI region (D<0), zone A.