

## PHYSICS

# Hybrid chiral domain walls and skyrmions in magnetic multilayers

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Noncollinear spin textures in ferromagnetic ultrathin films are currently the subject of renewed interest since the discovery of the interfacial Dzyaloshinskii-Moriya interaction (DMI). This antisymmetric exchange interaction selects a given chirality for the spin textures and allows stabilizing configurations with nontrivial topology including chiral domain walls (DWs) and magnetic skyrmions. Moreover, it has many crucial consequences on the dynamical properties of these topological structures. In recent years, the study of noncollinear spin textures has been extended from single ultrathin layers to magnetic multilayers with broken inversion symmetry. This extension of the structures in the vertical dimension allows room temperature stability and very efficient current-induced motion for both Néel DWs and skyrmions. We show how, in these multilayered systems, the interlayer interactions can actually lead to hybrid chiral magnetization arrangements. The described thickness-dependent reorientation of DWs is experimentally confirmed by studying demagnetized multilayers through circular dichroism in x-ray resonant magnetic scattering. We also demonstrate a simple yet reliable method for determining the magnitude of the DMI from static domain measurements even in the presence of these hybrid chiral structures by taking into account the actual profile of the DWs. The existence of these novel hybrid chiral textures has far-reaching implications on how to stabilize and manipulate DWs, as well as skyrmionic structures in magnetic multilayers.

## INTRODUCTION

The Dzyaloshinskii-Moriya interaction (DMI) is a form of antisymmetric exchange (1–3) that promotes canting between adjacent magnetic moments. As the DMI in ultrathin films originates from the strong spin-orbit coupling of interfacial atoms neighboring the magnetic layer (4, 5), it is notably found in a ferromagnet (FM) interfaced with two different heavy metals, such as Pt/Co/Ir (6, 7), or in magnetic layers inserted between a heavy metal and an oxide, such as Pt/CoFe/MgO (8, 9). Because of the energy lowering of the Néel configuration [internal magnetization perpendicular to the domain wall (DW)] associated to the interfacial DMI, the demagnetizing effects inside the DW that usually favor the Bloch configuration (internal magnetization along the DW) can be overcome and the Néel orientation becomes favored (10). Depending on the types of interfacial atoms combined with the FM and on the stacking order, the DMI can also change sign, which determines whether clockwise (CW) or counterclockwise (CCW) chirality is preferred (6). This stabilization of chiral magnetic textures also helps to stabilize quasi-punctual solitonic structures called skyrmions, in the present case, Néel (hedgehog) skyrmions (11). To obtain stable and compact individual skyrmions at room temperature, a successful approach has been to stack up several repeats of an asymmetric combination of ultrathin layers (7, 12). In this way, it is possible to stabilize columnar-shaped skyrmions, in which the increased magnetic volume reinforces their stability against thermal fluctuations up to room temperature, while still preserving both interfacial perpendicular magnetic anisotropy (PMA) in each ultrathin FM and required interfacial DMI due to the absence of inversion symmetry of the structure.

Here, our purpose is to investigate the actual thickness-dependent internal profile of chiral magnetic DWs and chiral skyrmions in Pt/Co/Ir- and Pt/Co/AlO<sub>x</sub>-based multilayers with PMA. The studied

multilayers are stackings made of 5 up to 20 repetitions of these trilayers with various magnetic layer thicknesses. Our study combines theoretical predictions, together with a direct experimental observation, of a vertical position-dependent reorientation of the chirality inside the magnetization texture in multilayers. By focusing first on the simplest case of DWs, we predict through micromagnetic simulations not only a significant variation of the DW width through the thickness of the multilayers but also, most importantly, a twisting of the internal DW texture along the thickness. In case of a large number of repetitions in the stacking, we find that topmost and bottommost layers host Néel orientations of opposite chiralities (CW and CCW), one in accordance with the chirality favored by the interfacial DMI and the other one being opposed to it. This reorientation thus results in an uncommon type of chiral composite Néel-Bloch DW. Then, we provide an experimental demonstration of the described reorientation of the DW texture in Pt/Co/AlO<sub>x</sub> multilayers by directly observing the DW chirality close to the top surface through circular dichroism (CD) in x-ray resonant magnetic scattering (XRMS). These results involve reconsidering the common implicit assumption of a uniform magnetization through the thickness of the multilayers. With a better understanding of the actual depth profile of the DW structure through the thickness, we then describe a novel approach to measure the DMI in these multilayers using the experimentally measured size of parallel stripe domains. We next propose a simple model to predict the occurrence of DW twisting. We finally discuss how our findings open new opportunities to manipulate more efficiently this new type of composite chiral skyrmion in multilayers through a proper engineering of the interfacial spin-orbit torques (SOTs).

## RESULTS

### Dipolar field-induced reorientation of magnetic DWs

In ultrathin magnetic films, the interfacial DMI can be strong enough to stabilize chiral Néel DWs. Let us consider a single DW separating two perpendicularly magnetized domains of magnetization pointing

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down ( $\mathbf{m} \cdot \mathbf{z} = -1$ ) for  $x < 0$  and up ( $\mathbf{m} \cdot \mathbf{z} = +1$ ) for  $x > 0$ . Denoting  $\theta(x) = \arccos(\mathbf{m} \cdot \mathbf{z})$  the polar angle and  $\psi$  the (fixed) azimuthal angle of the magnetization inside the DW, the DW energy is given by (13)

$$\sigma_{\text{dw}} = A \int_{-\infty}^{+\infty} \left( \frac{d\theta}{dx} \right)^2 dx + D \cos \psi \int_{-\infty}^{+\infty} \frac{d\theta}{dx} dx + K \int_{-\infty}^{+\infty} \sin^2 \theta dx + \sigma_{\text{dem}}(\psi) \quad (1)$$

where  $A$  is the Heisenberg exchange interaction amplitude,  $D$  is the interfacial DMI,  $K$  is the uniaxial anisotropy, and  $\sigma_{\text{dem}}$  is the demagnetizing energy associated to dipolar fields. Without DMI, the  $\psi$  dependence of  $\sigma_{\text{dem}}$  favors Bloch-type DWs ( $\cos \psi = 0$ ). However, it is now well known that, in the presence of DMI, the energy of the DW can be lower for a Néel orientation ( $\cos \psi = \pm 1$ ), which causes the reorientation of the internal magnetization of the DW and explains the stabilization of Néel structures (5, 8, 14). These Néel DWs in ultrathin films exhibit a fixed chirality controlled by the sign of the DMI and, notably, have proven to present extremely interesting current-driven dynamics (10).

In the recent years, the study of these chiral spin textures has become a very important research field both theoretically and experimentally. Most analyses of the spin configurations in single layers (for DWs and skyrmions) (10) assume a pure Néel profile and, for DWs, an energy independent of the DW spacing (uncoupled DWs). However, most often in multilayered stacks, the DWs are packed close to each other, meaning that the uncoupled DW approximation does not apply. Another complexity is that if the demagnetizing fields are strong (that is, for a large number of repetitions of magnetic layers), a DW structure between Bloch and Néel may arise ( $0 < \psi < \pi/2$ ). These two effects have been recently considered by Lemesh *et al.* (15), who derived an analytical model of the demagnetizing energies for standard one-dimensional (1D) magnetization profiles. In this model, a classical DW profile with one free internal angle  $\psi$  is assumed, and the magnetic configuration is parameterized by  $\Delta$  (DW width),  $\lambda$  (domain periodicity), and  $\psi$  only. Knowing the material parameters, a set of three nonlinear equations needs to be solved to get the equilibrium value of  $(\Delta, \lambda, \psi)$ . As we will demonstrate later, this model is very accurate for pure Néel DWs and allows us to estimate the value of  $D$ . It also allows us to a threshold minimum value of  $D$  that prevents internal tilting of the magnetization inside the DWs toward Bloch configuration.

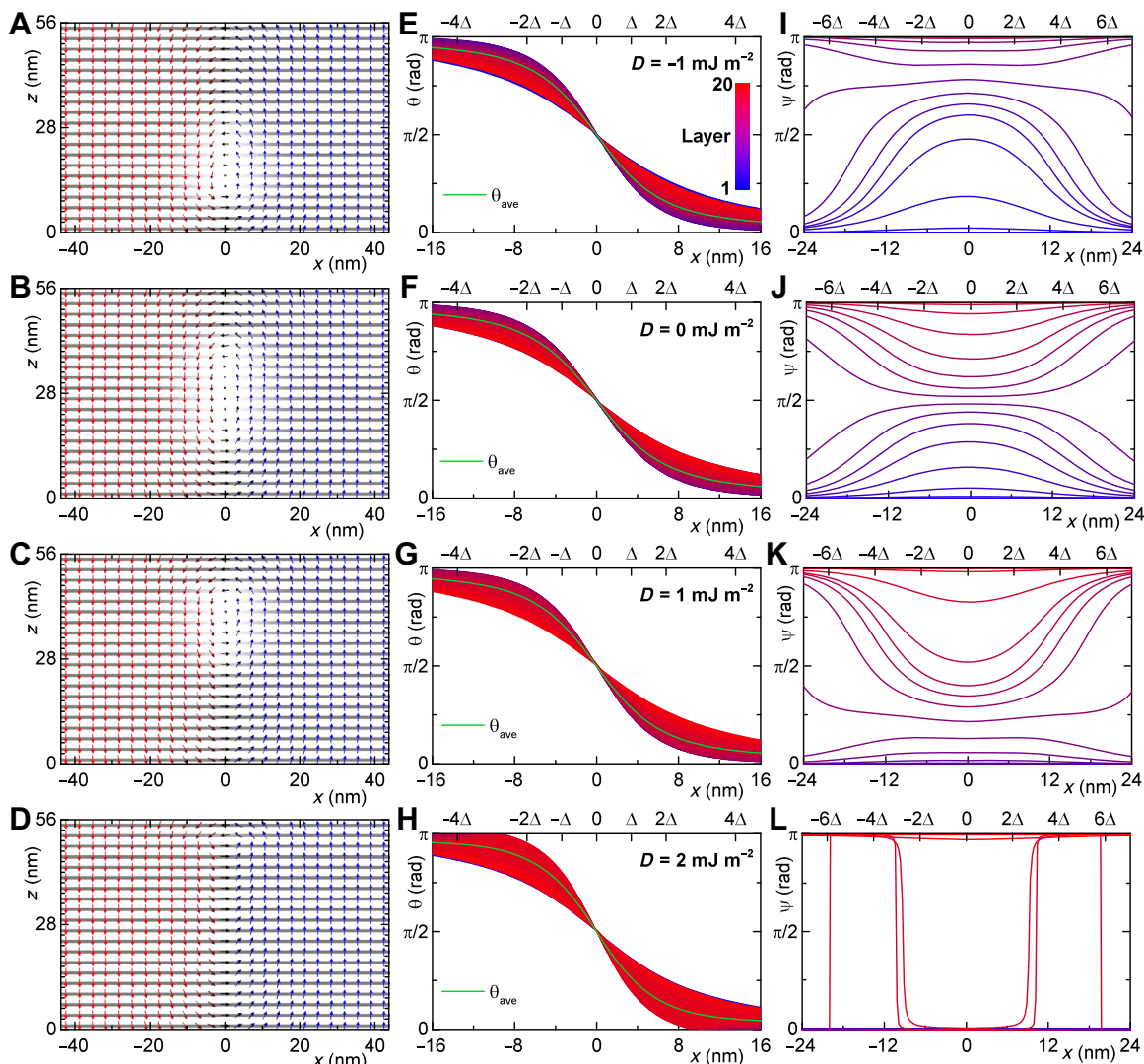
This  $(\Delta, \lambda, \psi)$  model is based on two strong assumptions: (i) The DW has a classical arctan profile without any  $x$  dependence for  $\psi$ , and (ii) all layers share the same DW profile, that is, without any  $z$  dependence. However, it is to be suspected that spin structures not uniform in the  $z$  direction can be stabilized in magnetic multilayers, as was suggested by micromagnetic simulations (16, 17) and by imaging of the emergent field of skyrmions (18). We aim here to demonstrate that, in most of the multilayered material systems considered in recent experiments on skyrmions at room-temperature, the actual magnetization structure through the thickness of the multilayer is more complex. As a result of the competition between the different interactions, a novel type of chiral hybrid magnetic texture is stabilized with some serious consequences on their spin torque-driven dynamics.

To illustrate the consequences of the magnetic texture varying through the multilayers, we perform a series of micromagnetic simulations of the DW structure in a multilayer comprising 20 repetitions of  $X(1 \text{ nm})/\text{Co}(0.8 \text{ nm})/\text{Z}(1 \text{ nm})$ , where  $X$  and  $Z$  are heavy metal or oxide

nonmagnetic spacers, which is typical among multilayers and allows for stabilization of the skyrmions (7, 12, 19–23). We hence perform the micromagnetic simulation of the stripe domain configuration in the full geometry including all magnetic layers and nonmagnetic spacers (see Materials and Methods). To study the influence of the DMI on the DW configuration, we vary  $D$  to be  $-1.0, 0.0, 1.0$ , and  $2.0 \text{ mJ m}^{-2}$ , for each of which the DW configuration has been computed. The results of these simulations are compiled in Fig. 1, in which we present the evolution of the magnetization profile as a function of the layer position inside the stacking in three different ways. The first column displays cross-sectional views of the magnetization profile (Fig. 1, A to D), the second column displays the spread of polar ( $\theta$ ) angle of the DW magnetization across the DW in each layer (Fig. 1, E to H), and the third column displays the azimuthal ( $\psi$ ) angle (Fig. 1, I to L). Each line of the figure then corresponds to the different DMI values. From these simulations, we find that, for a large range of DMI values, the DW internal magnetization is twisted between Néel and Bloch types along the direction (that is, the position of the magnetic layer in the stacking), leading to the formation of what we call a hybrid chiral DW structure.

One of the most striking results of these simulations is that the stabilized DW structures are not corresponding, whatever the value of  $D$ , neither to pure Bloch DWs ( $\cos \psi = 0$ ), as usually expected for  $D = 0$  in single layers (Fig. 1B), nor to pure Néel DWs ( $\cos \psi = \pm 1$ ) even for  $D$  as large as  $2.0 \text{ mJ m}^{-2}$  (Fig. 1D). We further notice that, even for intermediate  $D = \pm 1.0 \text{ mJ m}^{-2}$  (Fig. 1, A and C), the DWs do not exhibit an intermediate structure between Néel and Bloch types with a constant  $\psi$  angle of the magnetization inside the DW through the  $z$  direction but instead show a continuous variation of this  $\psi$  angle between all the magnetic layers of the stacking. These rather complex internal magnetization profiles, different from the single layer case, arise in these many-repeats multilayers from the competition between the interfacial DMI and the dipolar interactions between all layers. The resulting configuration can be described by the presence of a mostly Bloch ( $\cos \psi$  close to zero) DW part in some of the intermediate magnetic layers, whereas the combined action of the demagnetizing fields and DMI fields leads to the stabilization of mostly Néel DW parts in the bottommost and topmost layers. The DW thus has opposite internal magnetization directions in the topmost and bottommost layers, that is, considered separately, topmost and bottommost layers have opposite chiralities. This kind of magnetic configuration was known in the absence of DMI for DWs (24, 25) and bubbles (26) in magnetic thick layers. Here, we observe it for a large range of values of the DMI. We notice that the position of the Bloch DW part with respect to the central layer depends on the strength and sign of the DMI. For increasing  $|D|$  values, the preference for a single chirality shifts more and more the position of the most Bloch DW (minimal  $|\cos \psi|$ ) up or down, depending on the sign of  $D$ . For example, for  $D = 2.0 \text{ mJ m}^{-2}$ , only the two topmost layers contain Néel-like DWs with a sense of rotation opposite to all others (for a large negative  $D$ , the two bottommost DWs will have an opposite rotation). Only for a large enough value of  $D$  ( $|D| \geq 2.5 \text{ mJ m}^{-2}$  in the present case), the Bloch-like DWs are excluded, and the DW chirality is the same in all layers. The detailed description of the evolution of  $\theta(x)$  and  $\psi(x)$  profiles throughout the multilayer thickness is presented in section S1.

The discrepancy between the more elaborate micromagnetic simulations and the described analytical models stems from the absence, for the latter, of dependence of  $\Delta$  and  $\psi$  along  $z$ . As described, we instead predict a twisted DW magnetization through the thickness, constituting a new kind of hybrid chiral DW in DMI systems. The complexity of the



**Fig. 1. Micromagnetic simulations of hybrid chiral DWs for different DMI values.** (A to D) Cross-sectional view of a half simulation volume for  $[X(1)/Co(0.8)/Z(1)]_{20}$  multilayer with  $D = -1.0, 0.0, 1.0,$  and  $2.0 \text{ mJ m}^{-2}$  (top to bottom); the gray lines correspond to the Co layers. Arrows point in the direction of the magnetization,  $m_z$  is given by the color of the arrows from red (-1) to blue (+1), while  $m_y$  is displayed by the color of the grid from black (-1) to white (+1). (E to H) Polar angle  $\theta$  inside the DW in each layer for  $D = -1.0$  to  $2.0 \text{ mJ m}^{-2}$ . The blue-to-red gradient filling corresponds to the envelop of  $\theta$  of all profiles in the different layers from bottom to top [see color scale in (E)], while the green line is  $\theta_{ave}$ , the averaged  $\theta$  of all layers across the thickness. (I to L) Azimuthal angle  $\psi$  inside the DW in each layer for  $D = -1.0$  to  $2.0 \text{ mJ m}^{-2}$ . The blue-to-red lines again correspond to the layers from bottom to top.

DW (or skyrmion, as will be seen later on) profiles in multilayers is well depicted in the curves shown in Fig. 1 (I to L).

The important difference of hybrid chiral DWs with previous models originates from the competition between the interlayer magnetostatic interactions and DMI, which results in these flux-closure DW configurations that never correspond to DWs with a fixed internal magnetization angle in between pure Bloch and Néel types. We will see later that, in our multilayers (but more generally in all the multilayered systems experimentally studied so far), the actual value (27) of  $D$  is inferior to  $2.5 \text{ mJ m}^{-2}$ , which is the threshold value that allows pure Néel DWs through all layers in the present case. In consequence, it becomes essential to consider that the actual DW (or skyrmion boundary) textures are similar to the ones that we just described. As detailed in the end of this study, this finding has profound implications for the spin torque-induced manipulation of these composite DWs (28) or skyrmions (29).

### Direct observation of the internal twisting of the DWs

To experimentally verify the formation of the above-described composite DWs notably in the presence of a significant DMI, we grew several series of magnetic multilayered stackings by sputtering deposition (see Materials and Methods). The structures of the studied multilayers are reported in Table 1, starting from the substrate side //, where all thicknesses are given in nanometers and the numbers indexing the brackets are the number of repetitions of the base multilayer. The use of different buffer layers allows us to control the growth conditions and hence the amplitude of the magnetic anisotropy  $H_k$  and  $M_s$  values for similar stacks. The choice of opposite orders of the stackings allows us to get opposite signs of the DMI, while increasing the number of repetitions of the same trilayer base allows us to increase the influence of interlayer dipolar interactions.

To evidence the existence of the predicted DW twisting, we have measured the CD-XRMS, allowing us to directly probe the magnetization

**Table 1. List of the studied magnetic multilayers investigated.** The saturation magnetization  $M_s$ , the uniaxial anisotropy  $H_k$ , the measured domain periodicity, the estimated DW width, and the comparison of estimations of  $D$  with a fixed DW energy [ $K_{\text{eff}}$  model (39)], ( $\Delta, \lambda, \psi$ ) model, and the present full micromagnetic model are given for each multilayer. Two multilayers are not labeled as they have not been studied by CD-XRMS but only characterized by MFM to determine  $D$  with the different models.

| No.  | Multilayer stack  | $M_s$<br>(kA m <sup>-1</sup> ) | $H_k$<br>(mT) | $\lambda$<br>(nm) | $\Delta_{\Delta, \lambda, \psi}$<br>(nm) | $D_{\sigma} K_{\text{eff}}$<br>(mJ m <sup>-2</sup> ) | $D_{\Delta, \lambda, \psi}$<br>(mJ m <sup>-2</sup> ) | $D_{\text{full}}$<br>(mJ m <sup>-2</sup> ) |
|------|---|--------------------------------|---------------|-------------------|--|--|--|--|
| I    | //Pt 10/[Ir 1/Co 0.6/Pt 1] <sub>5</sub> /Pt 3                                   | 840                            | 330           | 167               | 7.27                                     | -1.46  | -1.43  | -2.30                                      |
| II   | //Pt 10/[Ir 1/Co 0.8/Pt 1] <sub>5</sub> /Pt 3                                   | 1229                           | 640           | 150               | 4.13                                     | -2.13  | -2.00  | -2.00                                      |
| III  | //Pt 11/[Co 0.8/Ir 1/Pt 1] <sub>5</sub> /Pt 3                                   | 637                            | 516           | 278               | 7.08                                     | 1.48   | 1.45   | 1.37                                       |
| IV   | //Ta 5/Pt 10/[Co 0.8/Ir 1/Pt 1] <sub>5</sub> /Pt 3                              | 683                            | 748           | 488               | 5.77                                     | 1.69   | 1.64   | 1.63                                       |
| V    | //Pt 11/[Co 0.8/Ir 1/Pt 1] <sub>10</sub> /Pt 3                                  | 637                            | 516           | 244               | 6.03                                     | 1.53   | 1.50   | 1.52                                       |
| VI   | //Ta 5/Pt 10/[Co 0.8/Ir 1/Pt 1] <sub>10</sub> /Pt 3                             | 847                            | 1088          | 250               | 4.11                                     | 2.11   | 2.08   | 2.06                                       |
| VII  | //Ta 15/Co 0.8/[Pt 1/Ir 1/Co 0.8] <sub>10</sub> /Pt 3                           | 957                            | 500           | 256               | 5.00                                     | -1.26  | -1.21  | -1.14                                      |
| —    | //Ta 10/Pt 7/[Pt 1/Co 0.6/Al <sub>2</sub> O <sub>3</sub> 1] <sub>20</sub> /Pt 3 | 1344                           | 469           | 190               | 4.06                                     | 1.40   | 0.88   | 1.29                                       |
| VIII | //Ta 10/Pt 7/[Pt 1/Co 0.8/Al <sub>2</sub> O <sub>3</sub> 1] <sub>20</sub> /Pt 3 | 1373                           | 358           | 175               | 3.72                                     | 1.20   | 1.20   | 1.01                                       |
| —    | //Ta 10/[Al <sub>2</sub> O <sub>3</sub> 1/Co 0.6/Pt 1] <sub>20</sub> /Pt 7      | 1120                           | 332           | 131               | 4.79                                     | -1.91  | -1.93  | -1.94                                      |
| IX   | //Ta 10/[Al <sub>2</sub> O <sub>3</sub> 1/Co 0.8/Pt 1] <sub>20</sub> /Pt 7      | 1245                           | 228           | 131               | 4.33                                     | -1.76  | -1.78  | -1.69                                      |

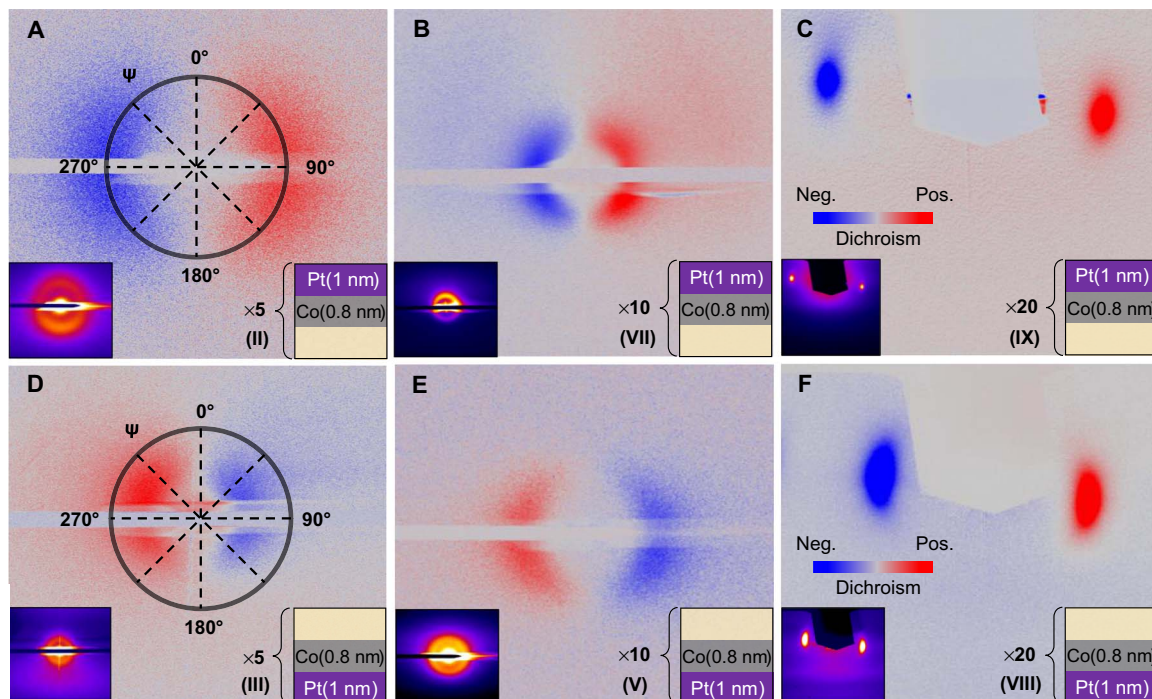
sense of rotation of the top layers of the stackings and thus the chirality of DWs as we recently demonstrated (30). For the present experiments, the energy is set at the  $L_3$  Co edge, and the angle of incidence of the x-rays has been chosen to be 18.5°, corresponding to the first diffraction peak of the multilayer. Under these conditions, x-rays are mainly sensitive to the first 15 nm, which corresponds to only four to five repetitions from the top of the multilayered structure that predominate in the measured XRMS signal (see Materials and Methods). Before performing the XRMS experiments, all the samples have been demagnetized to reach a magnetic ground state composed of either labyrinthine alternating up and down magnetized domains or parallel stripe domains depending on the chosen demagnetization procedure (see Materials and Methods).

In Fig. 2 (A to C), we display the XRMS dichroism patterns (diffraction patterns with unpolarized x-rays are shown as insets) recorded for the multilayers labeled (II), (VII), and (IX) (with 5, 10, and 20 repetitions, respectively). These three multilayers are the ones having the Pt layer on top of the FM ( $D < 0$ ; see Table 1). In the second line (see Fig. 2, D to F), the dichroism patterns are the ones obtained for the multilayers labeled (III), (V), and (VIII) (also with 5, 10, and 20 repetitions, respectively) that have the Pt layer below the FM ( $D > 0$ ; see Table 1). Note that, for the first two columns, the multilayers have been demagnetized with an oscillating out-of-plane field, leading to a labyrinthine domain configuration that generates rings in the diffraction patterns. On the contrary, for multilayers (IX) and (VIII) shown in Fig. 2 (C and F), the demagnetization procedure was done by applying an oscillating in-plane field, leading to parallel stripe domain configuration and resulting in diffraction spots appearing on both sides of the specular peak. The corresponding magnetic force microscopy (MFM) images are shown in section S2.

Important conclusions can be drawn from this series of XRMS patterns. First, because the dichroic signal is, in all cases, maximum along the 90° to 270° axis, it can be concluded that all the DWs in these dif-

ferent multilayers share the same type of DW in surface, corresponding to Néel DWs as we demonstrated recently (30), instead of tilted DWs in between Néel and Bloch configurations. Second, we observe that, for all samples with Pt on top of the FM (Fig. 2, A to C), a positive (red color in our convention) dichroism at 90° is found. This means that the top few layers exhibit a CW Néel DW chirality whatever the number of repetitions in the multilayers, which is the DW configuration that can be expected for this sign of the DMI. On the opposite, for the first two multilayers with Pt below the FM (Fig. 2, D and E), the dichroic signal is reversed (maximum positive at 270°). It indicates an opposite, CCW Néel DW chirality for 5 and 10 repetitions, again as expected from the sign of the DMI. A striking observation is then that the dichroism pattern recorded for the 20-repeats multilayer with Pt below the FM (shown in Fig. 2F) is again positive at 90° and thus indicates a CW Néel DW chirality in surface, which is opposite to what is expected for a positive DMI-driven chirality. As we explained previously, this apparent discrepancy is a direct fingerprint of the competition between interlayer dipolar fields and DMI: Because of the 20 repeats in the multilayer, the impact of the dipolar fields is large enough to impose a reversal of the chirality of the DWs in the top layers. Note that we are able to observe this dipolar field-induced chirality reversal only for sample (VIII) and not for multilayer (IX), because for  $D < 0$ , the DMI field is opposite to the in-plane component of the dipolar field in the bottom layers (see the magnetic configurations in Fig. 1, A to D), which is not probed by CD-XRMS. On the contrary, for  $D > 0$ , the reversal of the top layers dominates the CD-XRMS signal. As a consequence, for sample (IX), the local DW chirality reversal occurs in the bottom layers, not probed by the x-rays in our XRMS configuration, and thus, the reorientation is not observed. The chirality reversal does not occur for the 5- and 10-repeats multilayers because the strength of the dipolar fields relative to the DMI fields in these multilayers is not strong enough, as will be shown later. These experimental results demonstrate the dipolar field-induced twisting of DWs in magnetic multilayers, as described in the previous section.





**Fig. 2. CD-XRMS analysis of different multilayer stacking configurations.** Multilayers with  $D < 0$  and (A) 5 repetitions (sample II), (B) 10 repetitions (sample VII), and (C) 20 repetitions (sample IX) and with  $D > 0$  and (D) 5 repetitions (sample III), (E) 10 repetitions (sample V), and (F) 20 repetitions (sample VIII). The dichroism is normalized for each image and indicated by the color scale from blue (negative) to red (positive). Left insets are the corresponding sum [circularly left (CL) + circularly right (CR)] images evidencing the magnetic distribution ordering. Right insets present schemes of the studied stackings.

### Determination of $D$ with hybrid chiral DW textures

Being able to get a quantitative estimation of the interfacial DMI amplitude has been the subject of numerous studies in the last couple of years because this parameter is crucial to understand and control both the statics and dynamics of chiral DWs and/or skyrmions. Different approaches have been proposed. In single magnetic layers, the interfacial DMI has been experimentally accessed using Brillouin light spectroscopy (BLS) (27, 31), spin wave spectroscopy (32), chirality-induced asymmetric DW propagation (9, 33), or asymmetric magnetization reversal (34–36). However, these different methods appear to be not reliable in the case of interfacial DMI in multilayered systems. A first reason is that dipolar fields and magnetic couplings between layers shall influence significantly the spin wave propagation and thus the analysis of BLS spectra. Moreover, because dense labyrinthic magnetic domains are formed at low fields, they thus prevent the observation of the asymmetric reversal needed to estimate the DMI. This is why, in multilayered systems, analysis of domain spacing in the demagnetized state or as a function of the perpendicular magnetic field has been proposed to estimate the magnitude of DMI (7, 12, 37). This approach is based on the fact that the DW periodicity  $\lambda$  is the result of the balance between domain demagnetization energy and DW energy (38), the latter being dependent on  $D$ , as described above. By measuring the DW periodicity  $\lambda$  in demagnetized multilayers (in our case, using XRMS or MFM; see Materials and Methods and acquired images in section S2), it is possible to determine  $D$ .

We show here how the existence of hybrid chiral DWs induced by dipolar fields has a large impact on the evaluation of  $D$  in multilayers. To get an accurate estimation even in the presence of the hybrid chiral DW structures that we identified, our procedure is then to rely on the micromagnetic simulations with the complete geometry, as exemplified

above. We consider samples with the parallel stripe domains, and we vary the domain period in the simulation by changing the number of cells around the measured domain periodicity  $\lambda$  and then relax the system for different values of  $D$ . The extracted value  $D_{\text{full}}$  is  $D$  for which the energy density of the simulated system is minimum at  $\lambda$  (see section S3). The extracted  $D$  values for the different multilayers are listed in Table 1, in which we also compare  $D_{\text{full}}$  with the values of  $D$  estimated with the  $K_{\text{eff}}$  (39) and  $(\Delta, \lambda, \psi)$  (15) models (see section S3). Although all models give consistent values for pure Néel DWs, we note significant differences as soon as at least one layer has a reversed chirality.

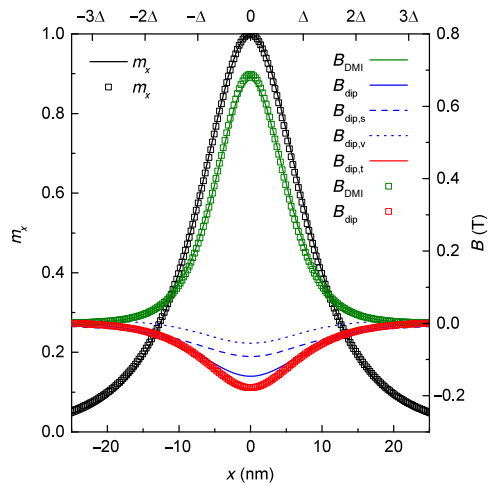
We emphasize that the measurement of the domain periodicity  $\lambda$  is error-free from the size of the MFM imaging probe (40) and that the sensitivity to local defects is reduced because of the averaging effect of imaging over sample areas much larger than  $\lambda$ . Thus, we believe that the most straightforward means to quantify the DMI in multilayers remains so far to find it from the measurement of  $\lambda$  in the ground state. Finally, another interesting aspect of this approach using the periodicity of stripe domains is that aligning parallel stripe domains with varying directions allows measurement of the DMI along different directions, which can give information about the anisotropy of DMI in materials having a crystalline structure that allows different DMI vectors along their different crystalline axes. We did not find anisotropic DMI in our samples.

### Criterion for dipolar field-induced twisting of DW chirality

In the following, our objective is to establish a simple criterion describing the occurrence of twisted chiral DWs in magnetic multilayered systems. Considering the simplified case of parallel domains where DWs in all layers are Néel, we can study the stability of this pure Néel configuration. To this aim, we find under these assumptions the horizontal

components of the dipolar- and DMI-induced fields inside the top layer for  $D > 0$ . We then compare the magnitude of the average action of both these fields on the DW profile,  $\mathcal{A}_{\text{dip}}$  and  $\mathcal{A}_{\text{DMI}}$ , over the total width of the DW (see section S4 for the details of the calculations). As an example, we show the deduced field profiles (green and blue lines for DMI fields and interlayer dipolar interaction fields, respectively, right scale) for our multilayer (III) [Pt(1)/Co(0.8)/Ir(1)]<sub>5</sub> in Fig. 3, together with the DW horizontal magnetization component  $m_x(x)$  profile (black curve, left scale). The dipolar interaction field  $B_{\text{dip}}$  (solid blue line) is obtained by summing surface (dashed blue line) and volume (dotted blue line) magnetic charge contributions. The small difference between the total field  $B_{\text{dip,t}}$  and the dipolar interaction field with the other layers  $B_{\text{dip}} = B_{\text{dip,s}} + B_{\text{dip,v}}$  comes from the horizontal component of the self-demagnetizing field of the top layer itself, which has no influence on the reorientation and which we do not include in the comparison of field actions. These analytically calculated field and magnetization profiles match the ones obtained from micromagnetic simulations (shown as squares of the corresponding colors in Fig. 3) under the assumptions mentioned just above.

When  $|\mathcal{A}_{\text{dip}}| > |\mathcal{A}_{\text{DMI}}|$ , for  $D > 0$  ( $D < 0$ ), the top layer (bottom layer) in-plane component of the magnetization inside the DW will reverse because of dipolar fields, so that a pure Néel DW throughout the whole stack is definitively not stable, whereas, for  $|\mathcal{A}_{\text{dip}}| < |\mathcal{A}_{\text{DMI}}|$ , Néel DWs of the same chirality can be stabilized in all layers. We can thus predict whether the DWs will reorientate or not, as shown in Fig. 4. The areas filled with a uniform color correspond to  $|\mathcal{A}_{\text{dip}}| < |\mathcal{A}_{\text{DMI}}|$  in which case pure Néel-like DWs are stabilized. The color indicates the preferred DW chirality imposed by the DMI. When this condition is not fulfilled, that is,  $|\mathcal{A}_{\text{dip}}| > |\mathcal{A}_{\text{DMI}}|$ , corresponding to the gradient areas, the DW reorientation into flux-closure DWs leading to the stabilization of hybrid chiral DWs shall occur. The twisting is more and more

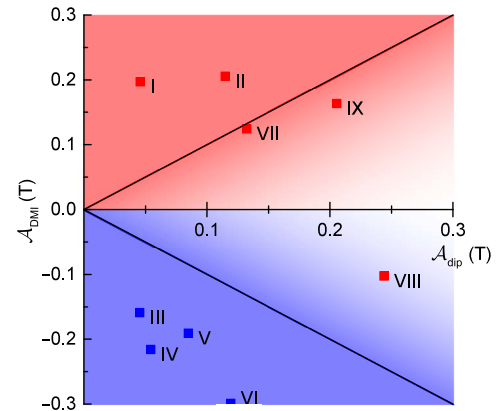


**Fig. 3.** DW profile (black, left scale) and analytical model for estimation of DMI (green, right scale) and dipolar (red, right scale) fields for the top layer of (III) [Pt(1)/Co(0.8)/Ir(1)]<sub>5</sub>. The squares are the result of micromagnetic minimization of the energy, while the lines are the fields obtained from the model. The dashed and dotted blue lines are, respectively, surface and volume charge contributions to the interaction dipolar field (solid blue line). The red line is the total dipolar field from the model (obtained by adding the intralayer demagnetizing field to the interlayer interaction dipolar field). The parameter  $\Delta$  has been adjusted for the magnetization arctan analytical profile to fit the micromagnetic profile.

pronounced when  $|\mathcal{A}_{\text{dip}}|$  becomes stronger relative to  $|\mathcal{A}_{\text{DMI}}|$ , which is signified by the progressively lighter background color. To compare these predictions with our experiments, we also include in Fig. 4 the experimental observation of the DW chirality of the top layers determined by CD-XRMS in all our different multilayers. The colors of the squares labeled (I) to (IX) indicate the top-surface DW sense of rotation, blue corresponding to CCW and red corresponding to CW chiralities. As described above, the DW surface chirality for multilayer (VIII) is found in agreement with the prediction, that is, the corresponding square is red in a blue gradient area, meaning that the observed chirality is CW, due to dipolar fields, whereas the DMI favors a CCW chirality. We further notice that, for multilayers (VII) and (IX), the DW reorientation that shall occur in the bottom layers is not observed by our enhanced surface-sensitive technique. This can be seen in Fig. 4, as the corresponding squares are red in a red gradient area. We believe that these predictions are important as they allow us to easily obtain for a given set of magnetic parameters  $M_s$ ,  $K_u$ ,  $A$ , and multilayer geometry, an approximation of the threshold DMI value  $D_u$  that ensures a unique DW or skyrmion chirality inside the whole stacking. We will see in the last section that knowing whether twisting of the magnetization chirality occurs or not is crucial as the DW or skyrmion dynamics are strongly modified depending on their actual spin textures.

#### Consequence on dynamics of skyrmions in multilayers

Stackings of ultrathin magnetic layers have been the subject of a large research effort recently for a demonstration of room temperature stabilization of small magnetic skyrmions. Moreover, it has already been shown (12, 21, 41, 42) that the interfacial SOTs can be used to efficiently move these skyrmions. As aforementioned, the existence of a complex spin texture through the different magnetic layers of a stacking due to the competition between interlayer dipolar fields and DMI fields is not only expected for DWs (and observed as we have demonstrated here) but also should be equivalent for skyrmions in multilayers, as it has been also pointed out in other very recent works (16, 18). Columnar skyrmions stabilized in multilayers, which is the favorite strategy to make them stable at room temperature, have a twisting of their chiral

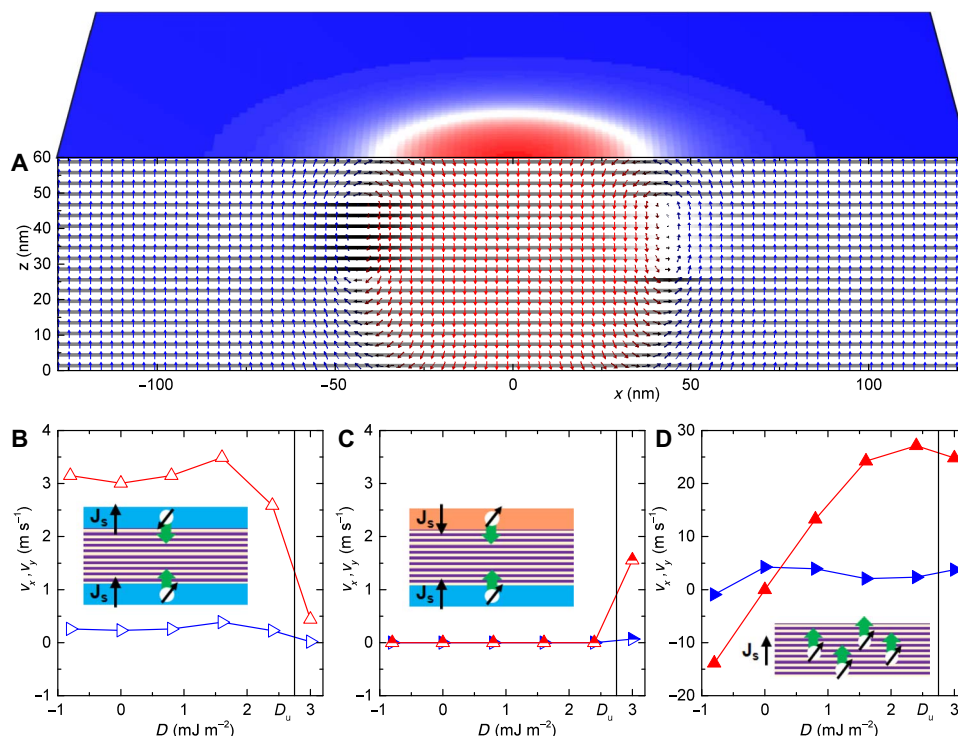


**Fig. 4.** Diagram comparing  $\mathcal{A}_{\text{dip}}$  and  $\mathcal{A}_{\text{DMI}}$  for each multilayer that has been characterized by CD-XRMS. When  $|\mathcal{A}_{\text{dip}}| < |\mathcal{A}_{\text{DMI}}|$ , pure Néel DWs are stabilized, and red (blue) indicates CW (CCW) chirality. The gradient areas correspond to  $|\mathcal{A}_{\text{dip}}| > |\mathcal{A}_{\text{DMI}}|$  with more and more pronounced reorientation into flux-closure DWs. Colored squares indicate the chirality as it has been observed by CD-XRMS for each sample labeled by roman numbers, where red stands for CW chirality and blue stands for CCW chirality.

spin texture through their thickness that strongly alters their dynamics. However, to our knowledge, this crucial issue has never been properly addressed so far. We show in Fig. 5A an example of the actual skyrmion profile in a multilayer of structure  $[X(1)/Co(1)/Z(1)]_{20}$  with  $D = 0.8 \text{ mJ m}^{-2}$ , obtained by micromagnetic modeling similar as before (see Materials and Methods). Equivalent to what happens for DWs, the skyrmion profile exhibits a progressive reorientation from CCW Néel to Bloch through its thickness and finally to CW Néel chirality in the topmost magnetic layers. Moreover, the skyrmion diameter also evolves depending on the layer position in the stack, being larger in the central layers and smaller in the external layers. These skyrmions with Néel caps were recently predicted and thoroughly modeled by micromagnetic simulations for amorphous, 50-nm thick Gd/Fe layers (16). With our findings, we demonstrate that the stabilization of these hybrid chiral skyrmions occurs in multilayered stackings of ultrathin magnetic and nonmagnetic layers with strong interfacial DMI. In our multilayers, the different magnetic layers are exchange-decoupled because of Pt/Ir and Pt/Al<sub>2</sub>O<sub>3</sub> spacers, which enhance reorientation effects as compared to exchange-coupled bulk materials such as Gd/Fe (16). In the following, we show that, through a precise engineering of the interfacial spin torques in stacked multilayers, new possibilities can be anticipated for achieving good control of the current-driven skyrmion dynamics.

To this aim, we have simulated the current-induced dynamics of isolated hybrid chiral skyrmions in an extended multilayer similar to the one shown in Fig. 5A for different values of the DMI and different spin injection geometries. Note that we only consider here damping-like

torques originating from vertical spin currents, which can be provided, for example, by the spin Hall effect and a current flowing along the  $x$  direction. The resulting skyrmion velocities are reported in Fig. 5 (B to D). In Fig. 5B, we first present the case in which the torque is applied only in the first bottom and the first top layers and the injected spin polarization is opposite in these two layers. This can occur when the multilayer is enclosed between two layers of the same heavy metal. We find that both the longitudinal and transverse velocities remain constant for moderate values of  $D$  [with transverse velocity larger than longitudinal velocity, as it is already known, because of the gyrotropic motion of the topological skyrmions (19)] but then drop to zero when  $|D| > D_u$ , corresponding to the critical minimum value of  $D$ , above which skyrmions with a unique chirality across all layers are stabilized. The velocity drops because skyrmions of opposite chiralities are driven in opposite directions for identical polarizations of injected spins, but in the same direction for opposite spin injections. As a consequence, the driving forces on the bottom CCW and top CW Néel skyrmion layers add up in the range of  $D$  for which the chirality twist is present and cancel out for a uniform skyrmion chirality. In Fig. 5C, we present the results for the opposite case where the injected spins have identical polarizations in the bottommost and topmost layers, for example, with the multilayer enclosed between heavy metals of opposite spin Hall angles. In that case, we find that the skyrmion motion is completely canceled up to  $D_u$  because the hybrid complex spin texture leads to opposite skyrmion chiralities in bottommost and topmost layers. The motion occurs only for  $|D| > D_u$ , in which case top- and bottom-layer chiralities and injected spins are identical. Finally, for a uniform current



**Fig. 5. Micromagnetic simulations of the dynamics of hybrid chiral skyrmions.** (A) Cut view of the simulation volume for a  $[X(1)/Co(1)/Z(1)]_{20}$  multilayer with  $D = 0.8 \text{ mJ m}^{-2}$ . Arrows point in the direction of the magnetization, and  $m_z$  is given by the color of the arrows from red (−1) to blue (1), while  $m_y$  is displayed by the color of the grid from black (−1) to white (+1). The  $m_z$  component in the top layer is represented in perspective view by the color from red (−1) to blue (1). (B to D) Skyrmion velocities for different values of  $D$  and geometries. Right/up pointing blue/red triangles stand for horizontal/transverse velocity components, obtained for (B) opposite injection in bottommost/topmost layers, (C) identical injection in bottommost/topmost layers, and (D) uniform injection. The injection geometry is depicted by the inset in each case.



injection (for example, with a Pt layer adjacent to the bottom of each ferromagnetic layer; Fig. 5D), we find that the transverse ( $v_y$ ) velocity is roughly proportional to  $D$  up to  $D_u$  (note the reduced velocity in cases of only external layer injection as compared to uniform injection within all layers, as fewer spins are injected in total). Again, it is because skyrmions of opposite chiralities are driven in opposite directions, which implies that the global motion is related to the balance of CCW and CW Néel skyrmion layers. Even if  $D$  does not directly affect the velocity of a given structure, as  $D$  controls the amount of layers with CCW and CW Néel orientations, it also controls the overall velocity. We also notice that, in this case, the driving forces compensate at  $D = 0$ , leading to a zero global  $v_y$ . The small  $v_x$  component arises from the Bloch part of the hybrid chiral skyrmion. The last case presented in Fig. 5D corresponds to the experimental studies of skyrmion dynamics reported by Woo *et al.* (12) and Litzius *et al.* (20) and provides a simple explanation why a very large (and hard to achieve) interfacial DMI is mandatory to achieve fast skyrmion motion in stacked multilayered systems in comparison to the case of single layers or multilayers with few repeats. In our multilayers, the intermediate Pt layers are thinner than the spin diffusion length. In this case the torques would be efficient only in the external layers, as is the case in Fig. 5B [multilayers (I) to (III), (V), and (VIII), enclosed between Pt layers] or in Fig. 5C [multilayers (IV), (VI) to (VII), and (IX), enclosed between Ta and Pt layers]. The high value of  $D_u \approx 2.8 \text{ mJ m}^{-2}$  for 20 repetitions of X/Co(1 nm)/Z is above all reported values in multilayers. This shows that pure Néel skyrmions in these structures are very unlikely to occur. However,  $D_u$  can be lowered to realistic values in 20 repetitions of a thinner ferromagnetic layer such as in Pt/Co(0.6 nm)/AlO<sub>x</sub> or in structures for which  $M_s$  is lower. Finally, this series of simulations thus provides guidelines on how to design multilayers with many repetitions hosting skyrmions and how to engineer the interfacial spin torques to achieve a fast motion when hybrid chiral skyrmions are present in these multilayers.

## DISCUSSION

The presented skyrmion dynamics simulations open new perspectives for the engineering of multilayered magnetic materials. Using these techniques, it will be possible to better control DW and skyrmion motion with SOTs in asymmetric multilayers and even in most common PMA symmetric stacks, such as Pt/Co and Pd/Co, which exhibit only low DMI or even no significant DMI. Notably, injecting spin Hall effect spin currents from the same material below and above the multilayer will lead to motion, as the chirality of the structure reverses from bottom to top. Moreover, as we have shown with the system displayed in Fig. 1, the balance between the number of layers and the value of  $D$  in the structure actually controls the position of the Bloch DW within the multilayer. It is thus possible to control the overall chirality of the structures by engineering the base multilayer repetition number, thus tuning the ratio of CCW Néel, Bloch, and CW Néel DWs. This would allow control of the direction of the motion in the case of a uniform spin current injection, as well as the chirality-related properties of DW and skyrmion motion. Moreover, our simulations of current-induced motion show the dynamical stability of the twisted chiral skyrmions as they are robust even under current injection, which can thus be used for spin current-induced motion.

With the help of our description, it is possible to understand and predict the occurrence of twisting of DW and skyrmion chiralities in multilayers. Notably, the common picture of a constant internal tilt inside the DW or constant chirality inside the skyrmion and through

the different layers is revealed not to be valid. We have experimentally demonstrated this reorientation effect with a surface-sensitive x-ray diffraction technique, allowing us to observe the surface chirality of the DW configurations. These complex structures strongly modify the way the DMI should be quantified in magnetic multilayers. These results highlight the importance of advanced engineering of spin-orbit-related interfacial properties, combining PMA, SOT, and DMI in multilayered systems to promote the stabilization and the fast dynamics of ultrasmall skyrmions at room temperature, which are needed for many types of potential applications.

## MATERIALS AND METHODS

### Sample fabrication and characterization

All series of multilayers were grown by dc and radio frequency magnetron sputtering at room temperature on thermally oxidized silicon substrates, after deposition of a Ta or Pt buffer, as described in Table 1, and capped with Pt ( $\geq 3 \text{ nm}$ ) to prevent oxidation. We checked by x-ray reflectivity analysis that the quality of the interfaces in the multilayers is independent of the number of repetitions (up to 20). The saturation magnetization  $M_s$  and the effective perpendicular anisotropy field  $H_{\text{eff}} = H_K - M_s/2$  of each multilayer were obtained by SQUID (superconducting quantum interference device) and alternating gradient magnetometry measurements.

### Demagnetization procedure and MFM imaging

We first compared the domain periodicity in the labyrinthine and aligned parallel domain configurations (obtained after out-of-plane and in-plane demagnetization procedures, respectively), measured by MFM (see the Supplementary Materials) in lift mode for our multilayer (VIII) [Pt(1)/Co(0.8)/AlO<sub>x</sub>(1)]<sub>20</sub>. For both in-plane and out-of-plane demagnetization procedures, the maximum oscillating field was set higher than the anisotropy field in the multilayers, that is,  $B \geq 1.2 \text{ T}$ . Then, the field was set to oscillate between positive and negative values with a decay factor of 0.5% until 0.5 mT was reached. For the in-plane demagnetization procedure, the multilayer was slightly tilted ( $\sim 5^\circ$ ) with respect to the field to favor multiple reversals of the magnetization everywhere to get closer to a parallel stripe ground state. The extraction of the mean domain periodicity  $\lambda$  after Fourier transform revealed that it can differ significantly, by up to 20%, between the two demagnetization procedures. All rigorous models of magnetic domains in multilayers had, however, been derived for parallel stripe domains (38, 43, 44). Even if the difference in the domain width for the two demagnetized state configurations may be very small (15) in the ideal case, this deviation precluded the use of perpendicularly demagnetized domains for the estimation of the DMI, as only the values derived from the parallel stripe domains periodicity provided consistent results. The multilayers were imaged with Asylum Low Moment tips in a double-pass, tapping mode, followed by a lift mode at 20-nm height, at room temperature.

### XRMS measurements

XRMS experiments were carried out at the SEXTANTS beamline of the SOLEIL synchrotron. The diffraction was measured under reflectivity conditions for CL and CR polarizations of the incident x-ray beam. The photon energy was set at Co  $L_3$  edge (778.2 eV) using the RESOXS (resonant soft x-ray scattering) diffractometer. The diffracted x-rays were imaged on a square charge-coupled device (CCD) detector covering  $6.1^\circ$  at the working distance of this study. All the images were geometrically corrected along the  $Q_x$  direction to account for the projection



effect related to the photon incidence angle of  $18.5^\circ$ . The sum of the images obtained with CL- and CR-polarized light gave rise to a diffraction pattern around the specular beam (which was blocked by a beamstop to avoid saturation of the CCD) in the reciprocal plane ( $Q_x, Q_y$ ). Denoting  $I_{CL}$  and  $I_{CR}$  the intensities collected by the CCD for CL- and CR-polarized incident lights, the CD of the scattering signal is defined as  $(I_{CL} - I_{CR})/(I_{CL} + I_{CR})$ .

### DW profiles—Micromagnetic simulations

To find the profiles of DWs in the stripe domain configuration without making assumptions on their shape, we performed micromagnetic simulations with the MuMax3 solver (45) in a 3D mesh accounting for the full geometry of the multilayers. The simulation volume was  $\lambda \times 32 \text{ nm} \times Np$ , respectively, in the  $x$ ,  $y$ , and  $z$  directions. Two DWs separating up, down, and up domains were initialized at  $-\lambda/4$  and  $\lambda/4$ . Periodic boundary conditions inclusive of the periodic stray fields calculated for  $64 \times 64$  identical neighbors in the  $x$  and  $y$  directions were introduced. The  $x$  cell size was 0.25 nm for the whole XRMS multilayer series. The  $z$  cell size was 0.2 nm, or 0.1 nm, when required by the values of layer spacing  $p$  and thickness  $t$ . Simulations were performed at 0 K.

### Skyrmion velocities—Micromagnetic simulations

To find the potential current-induced velocities for different skyrmion chirality conditions and spin current geometries, we performed micromagnetic simulations in the full geometry for the multilayer  $[X(1)/Co(1)/Z(1)]_{20}$ . Here, Co(1) was chosen to reduce the simulation grid size. The simulation volume was  $256 \text{ nm} \times 256 \text{ nm} \times Np$ , respectively, in the  $x$ ,  $y$ , and  $z$  directions. Periodic boundary conditions inclusive of the periodic stray fields calculated for  $3 \times 3$  identical neighbors in the  $x$  and  $y$  directions were introduced. All cell sizes were 1 nm. Before current was applied, the configuration was relaxed from an initial Bloch skyrmion with CCW wall internal magnetization. The current density was then modeled as a fully polarized (along  $y$ ) vertical spin current of current density  $J = 2.5 \times 10^{10} \text{ A m}^{-2}$ . The Gilbert damping  $\alpha$  was set to 0.1, and the out-of-plane field was set to  $B = 200$  or  $300 \text{ mT}$  (for  $D > D_w$ ). Because of this required increase of external field to confine the skyrmion to a stable circular shape for high  $D > D_w$ , the skyrmion size was reduced and thus velocity as well (19), without affecting the discussed quantitative behavior. Simulations were performed at 0 K.

### SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at <http://advances.sciencemag.org/cgi/content/full/4/7/eaat0415/DC1>

Section S1. Detailed description of the hybrid chiral spin textures found in simulations  
Section S2. Details of the determination of DMI with complete micromagnetic simulations  
Section S3. MFM on the different samples

Section S4. Domain wall fields—Analytical derivation

Fig. S1. Detailed version of Fig. 1.

Fig. S2. MFM images of domain structures in studied multilayers.

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