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Citation for published version (APA):

Han, D., Kim, N. H., Kim, J. S., Yin, Y., Koo, J-W., Cho, J., Lee, S., Kläui, M., Swagten, H. J. M., Koopmans, B., & You, C-Y. (2016). Asymmetric hysteresis for probing Dzyalohsinskii-Moriya interaction. *Nano Letters*, 16(7), 4438–4446. <https://doi.org/10.1021/acs.nanolett.6b01593>

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DOI:
[10.1021/acs.nanolett.6b01593](https://doi.org/10.1021/acs.nanolett.6b01593)

Document status and date:
Published: 15/07/2016

Document Version:
Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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Asymmetric Hysteresis for Probing Dzyaloshinskii–Moriya Interaction

Dong-Soo Han,[†] Nam-Hui Kim,^{‡,§} June-Seo Kim,[†] Yuxiang Yin,[†] Jung-Woo Koo,[†] Jaehun Cho,[‡] Sukmook Lee,[‡] Mathias Kläui,[§] Henk J. M. Swagten,[†] Bert Koopmans,[†] and Chun-Yeol You^{*,‡,||}

[†]Department of Applied Physics, Center for NanoMaterials, Eindhoven University of Technology, PO Box 513, 5600 MB Eindhoven, The Netherlands

[‡]Department of Physics, Inha University, Incheon 22212, Republic of Korea

[§]Institut of Physics and Graduate School of Excellence Materials Science in Mainz, Johannes Gutenberg-Universität Mainz, 55099 Mainz, Germany

^{||}Department of Emerging Materials Science, DGIST, Daegu 42988, Republic of Korea

Supporting Information

ABSTRACT: The interfacial Dzyaloshinskii–Moriya interaction (DMI) is intimately related to the prospect of superior domain-wall dynamics and the formation of magnetic skyrmions. Although some experimental efforts have been recently proposed to quantify these interactions and the underlying physics, it is still far from trivial to address the interfacial DMI. Inspired by the reported tilt of the magnetization of the side edge of a thin film structure, we here present a quasi-static, straightforward measurement tool. By using laterally asymmetric triangular-shaped microstructures, it is demonstrated that interfacial DMI combined with an in-plane magnetic field yields a unique and significant shift in magnetic hysteresis. By systematic variation of the shape of the triangular objects combined with a droplet model for domain nucleation, a robust value for the strength and sign of interfacial DMI is obtained. This method gives immediate and quantitative access to DMI, enabling a much faster exploration of new DMI systems for future nanotechnology.

KEYWORDS: Dzyaloshinskii–Moriya interaction, spintronics, nanomagnetism, asymmetric hysteresis loop, chiral magnet, perpendicular magnetic anisotropy

Recently, the interfacial Dzyaloshinskii–Moriya interaction (DMI)^{1–3} has been of great interest because of its intriguing physical role in the stabilization of nanometer-sized chiral spin configurations in perpendicularly magnetized multilayers, such as a Neel-type domain wall (DW)^{4–10} and a nanomagnetic skyrmion,^{11–23} both very promising candidates for future information-processing devices with high-energy efficiency^{6,12,14,20} and ultrahigh-density storage capability.^{12,13} In particular, searching materials or systems providing strong DMI has become a central focus in current research, since it has been theoretically predicted¹³ that room-temperature magnetic skyrmions may only be stabilized^{15,21,22} by substantial DMI. To tackle the challenge met in the quest for large DMI systems, it is indispensable to develop a simple and accurate way of quantifying DMI. In this context, several experimental attempts have been made to estimate the sign and/or magnitude of the effective DMI coefficient D through different measurement techniques. For out-of-plane magnetized systems, for instance, a number of ways to quantify the DMI have been suggested, mostly requiring imaging of magnetic domain walls^{5,24} and based on the analysis of domain wall propagation^{6–10,25–27}

under the application of current or magnetic field. The asymmetric field-induced growth of reverse domains is the most widely used approach in this class of materials. However, this method has been found to entail serious drawbacks to quantify the DMI, as other effects, such as local stray field variations or anisotropy gradients, can also lead to an asymmetric displacement, and analysis is hampered by possible additional effects resulting from wall spin structure deformations during the propagation. Furthermore, several asymmetric wall propagation observations^{28–30} cannot be described with this model based on a pure DMI, and it has been suggested that chiral damping,³¹ in addition to DMI, might play a key role. Even taking into account chiral damping, many of the observations²⁶ using domain wall displacement can still not be described, showing that a reliable quantitative determination of the phenomenon of DMI using *dynamic methods* is still tenuous.

Received: April 18, 2016

Revised: June 13, 2016

Published: June 27, 2016

Figure 1. Schematic illustrations of magnetization configurations within a microstructured ferromagnetic thin film with an interfacial DMI of $D > 0$. The microstructured ferromagnetic thin film consists of a square (a) and a triangle shape (b) which involve the lateral 2-fold rotatory symmetry and asymmetry around the z -axis, respectively. Panels c and d represent the relative orientation between the local magnetizations at each left and right sideways facing surface and the in-plane field $\mathbf{H}_x$. The $\mathbf{m}_R$ and $\mathbf{m}_L$ indicates the local magnetization at the right and left side, respectively. The superscripts in each U and D represent the upper and lower side of the triangle structures, respectively.

Alternatively, measurements of D via nonreciprocal spin waves^{32–40} have been recently realized in both in-plane and out-of-plane magnetized systems. Despite successful experimental demonstration of DMI by these approaches, it is still practically challenging to measure the magnetization dynamics in the ns or GHz regime, which requires unique measurement tools with high-spatial (wave vector) and/or time (frequency) resolution. Furthermore, the accuracy of this approach is also contested, as asymmetric anisotropies at the two interfaces of the ferromagnetic materials (FM) can lead to similar asymmetries of the dispersion relation,⁴¹ making the interpretation for the DMI determination nontrivial. The technical complexities as well as issues with the reliability and robustness of these approaches are another hurdle to be overcome, impeding a rapid development of this exciting research field. Hence, in spite of the increasing interest to DMI-related phenomena, so far very limited experimental efforts have been devoted to extract DMI in films of (sub) nanometer thickness. It is clear that to develop new materials, systems, and devices, the main bottleneck is the reliable determination of the DMI. So there is an obvious urgent need to develop a novel *static method* that is not susceptible to chiral damping or asymmetric anisotropies and that would allow for a straightforward and versatile determination of both the sign and magnitude of DMI.

In this work, we present the observation of a significantly shifted, asymmetric magnetic hysteresis loop manifested by interfacial DMI in laterally asymmetric microstructures. More specifically, in triangular-shaped ultrathin ($\sim$ nm) magnetic films patterns, where the lateral 2-fold rotation symmetry with respect to the z -axis perpendicular to film plane is broken, we

demonstrate a significant shift of the magnetic hysteresis along the magnetic field axis under the application of an additional static in-plane field, closely resembling the shifted “exchange-biased” hysteresis^{42,43} due to coupling a ferromagnetic film with an antiferromagnetic material. This loop shift in our asymmetric triangular microstructures is found to reverse sign when changing the polarity of the in-plane field as well as when D changes sign, and moreover, it strongly depends on the strength of the in-plane field, all suggesting that a *unique straightforward* measurement of the DMI in nanoscale is feasible. Indeed, from a well-established half-droplet^{29,44,45} model used to explain preferential nucleation at one of the sides of an unstructured nanomagnetic thin film, the shifted hysteresis loop is shown to directly yield the sign and strength of DMI. Moreover, the robustness and successful quantification of DMI is further substantiated by a systematic variation of the triangular shape of these structures, yielding shift in the hysteresis almost independent of the object size. This new tool is, thus, providing a fascinating prospect for a swift exploration of materials for nanoelectronics relying on engineered interfacial DMI.

In a confined system like a nano- or microstructured magnetic thin film, DMI is known to allow for a boundary condition^{13,44,46} of $2A\partial_{\mathbf{m}}/\partial_{\mathbf{n}} = D(\mathbf{z} \times \mathbf{n}) \times \mathbf{m}$, where $\mathbf{n}$ is unit vector normal to the side surface, $\mathbf{z}$ unit vector normal to film interface, $\mathbf{m}$ local magnetization orientation, D the Dzyaloshinskii–Moriya coefficient, and A exchange stiffness. Consequently, the magnetic configuration on the sideways facing surfaces (edges) of perpendicularly magnetized thin film structure experiences an effective magnetic field, which tend to pull the local magnetization parallel or antiparallel to the surface normal vector $\mathbf{n}$ depending on the sign of D and on the

Figure 2. Serial snapshot images of local magnetic domain structure during its magnetization switching under the indicated in-plane H_x and out-of-plane field H_z , as captured by the polar Kerr microscope in square (a) and triangle (b) patterns of Pt/Co/Ir. The left and right column in each panel of (a) and (b) represent the switching from down-to-up (D-U) and up-to-down (U-D), respectively. The yellow square indicates the microstructured magnetic thin films. The gray and black colors in the structure represent negative and positive magnetizations along the z -axis.

sign of the out-of-plane component of magnetization, M_z . This leads to an opposite tilted orientation of local magnetization on the opposite sides of the structure (see Figure 1a) over a length scale of the order of the domain wall width, which is typically a few nanometers. These chiral magnetic configurations at the sideways facing surfaces in a Pt/Co/AlO_x magnetic thin film have recently been revealed by a pioneering experiment by Pizzini et al.,⁴⁴ demonstrating chirality-induced asymmetric nucleation of magnetic domains when applying an in-plane bias field, $\mathbf{H}_x = \pm H_x \hat{x}$. According to their work, when the in-plane magnetic field is applied to the system, the chiral magnetic configurations at sideways facing surfaces give rise to a difference in energy barrier between the two opposite sides. Therefore, the nucleation process is found to be induced on one specific side where the Zeeman energy barrier is lowered when the tilt of the local magnetizations along the x -axis is parallel to $\mathbf{H}_x$.

A remarkable and useful aspect in the asymmetric switching mechanism is that the nucleation field is quantitatively related to the amount of Zeeman energy, which strongly depends on both the strength of H_x and the orientation of local magnetization near the sideways facing surfaces relative to $\mathbf{H}_x$. In this regard, one would intuitively expect contrasting magnetic hysteresis loops within two different systems: one with laterally symmetric left and right sides and the other with antisymmetric sides, where the relative orientation of local magnetizations between two opposite sides are distinct. Figure 1a–b, for instance, schematically illustrates the magnetization distribution for $D > 0$ in two different structures with and without symmetric left and right side edges, respectively. In Figure 1c–d, their local magnetizations $\mathbf{m}_L(\mathbf{m}_R)$ at the left (right) side relative to each positive and negative H_x are highlighted. In a square structure, when magnetization reversal occurs, the nucleation side dominating the switching process changes depending on the polarity of M_z and H_x : for up-to-down (U-D) switching, the nucleation is initiated at the left

(right) side for a positive (negative) H_x , vice versa for down-to-up (D-U) switching. However, in this case, the local magnetization on one side with a specific M_z shows an equivalent configuration as compared to the opposite side for an inverted M_z . As a result, the nucleation fields for both U-D and D-U switching should be exactly the same (see Figure 1c). This is in striking contrast to the case when switching a triangular object where the lateral 2-fold rotation symmetry with respect to the z -axis perpendicular to film plane is broken. In this case, although the nucleation side also changes upon reversing the polarity of M_z , it experiences a different Zeeman energy at two opposite sides due to asymmetric orientation of local magnetizations at two opposite sides, yielding different nucleation fields for U-D and D-U (see Figure 1d). Consequently, one can intuitively expect an essentially *asymmetric magnetic hysteresis loop* in the triangle, where it is shifted along the out-of-plane field axis under the application of the additional static in-plane field. We note that such an asymmetrically shifted hysteresis loop is a telltale sign of a symmetry-breaking interaction: usually magnetism is time-reversal invariant meaning that hysteresis loops are always symmetric around zero field (positive and negative fields have symmetric effects and are energetically degenerate). By applying an in-plane field, this degeneracy is not lifted, thus still symmetric loops can be expected. Only effects, such as *exchange bias*, can lift this degeneracy and are thus symmetry-breaking,^{42,43} and this effect relies on uncompensated spins at the interface between a ferromagnet and an antiferromagnet leading to an effective unidirectional bias field. However, we here show that the specific combinations of the device symmetry, the field symmetry, and the symmetry breaking induced by the DMI result in a shifted hysteresis loop reminiscent of exchange bias and thus indicative of an effective bias field for the reversal, which is only slightly appreciated so far.

Figure 3. Magnetic hysteresis loops under the indicated in-plane bias field as measured in (a) Pt/Co/Ir and (b) AlO_x/Pt/Co. The symbols represent average experimental data measured in a single pattern over 10 times, and solid lines indicate the least-squares fit to $2(1/(1 + \exp((\mu_0 H_x - A)/B))) - 1$, where A and B are the fitting parameters to be determined. The top and bottom panels in each column of (a) and (b) represent the result from the square and triangle pattern, respectively. $|H_C/H_0|$ versus H_x as obtained from Pt/Co/Ir (c), AlO_x/Pt/Co (d), and micromagnetic simulations for $D = +2$ (e) and 0 mJ/m² (f). The symbols in experimental cases represent data and solid lines are the least-squares fit to eq 1. The top and bottom panels at each column of c, d, e, and f are the results from square and triangle pattern, respectively.

As a proof-of-concept and experimental demonstration of the feasibility of our approach, we carried out measurements on samples of Si(substrate)/Ta(4 nm)/Pt(4 nm)/Co(1.2 nm)/Ir(4 nm) and Si(substrate)/Ta(10 nm)/AlO_x(2.5 nm)/Co(1.15 nm)/Pt(4 nm) (hereafter denoted as Pt/Co/Ir and AlO_x/Co/Pt, respectively), with a strong perpendicular magnetization anisotropy (details, see [Supporting Information I and II](#)). To implement symmetric and asymmetric left and right sideways facing edges, the magnetic thin films were microfabricated into two patterns, namely, into a square and a triangle. (See [Methods](#) and [Supporting Information III](#).) Magnetic hysteresis loops were monitored with a wide-field polar Kerr microscope applying a magnetic field of $\mu_0 H_z = -26.5$ mT into the out-of-plane direction to saturate the magnetization and subsequently swept toward the opposite z -direction at a rate of 1 mT/s. During the measurement, an additional static in-plane bias field was subjected along the x -axis to initiate the chirality-induced asymmetric nucleation breaking the lateral 2-fold symmetry. We would like to further note that square patterns, which should exhibit symmetric switching fields, were measured prior to triangle patterns, thereby excluding any possible artifacts which might arise from an unintentional misalignment of the in-plane magnet or exchange bias effects, which affect all geometries equally.

To validate the chirality-induced asymmetric nucleation process in our system—an essential prerequisite for the measurement of the DMI-shifted magnetic hysteresis loop—we first imaged magnetization dynamics close to the switching field. [Figure 2](#) shows representative serial snapshots of instantaneous magnetic domain structures in the two different patterns as measured in a Pt/Co/Ir sample during its U-D and D-U switching (see [Supporting Information IV](#) for AlO_x/Co/Pt). Here, the yellow-lined region indicates the microstructured magnetic thin film, and the bright and dark contrasts inside the structure represent negative and positive saturation of magnetizations, respectively. Two remarkable features are clearly seen in [Figure 2](#). First, the nucleation side dominating the reversal

process changes according to its switching polarity, clearly revealing chirality-induced asymmetric switching behavior as evidenced in previous work.⁴⁴ For D-U switching, the nucleation of magnetic domains is initiated at the right side, while it starts at the left side for U-D switching for $\mu_0 H_x = +120$ mT in both the triangular and squared patterns. Accordingly, one can immediately deduce that our Pt/Co/Ir sample stabilizes a left-handed chiral configuration with a moderate DMI, as in Pt/Co/AlO_x.^{15,34,36,44} Second, the nucleation field indeed differs depending on the side where the switching process is initiated in the triangle, whereas it is almost the same in the square. In the square pattern, the nucleation of magnetic domains appears at around $\mu_0 H_x \approx 2.9$ mT, irrespectively of the switching polarity. By contrast, it exhibits a significant and large difference (~ 1 mT) in the nucleation fields between the D-U and U-D switching in the triangular pattern. Here we note that statistically distributed small differences in the nucleation field between D-U and U-D switching were also observed in the squares due to the stochastic nature of domain nucleation as evidenced from repeatedly sweeping the magnetic field. However, these stochastic fluctuations are much smaller than the shifts in the triangles, where only systematic differences in nucleation fields were found upon repeatedly sweeping the field. In addition, the chiral switching was also confirmed upon reversing the polarity of H_x . In both patterns, completely opposite switching behavior was found for a negative H_x , supporting the chiral and asymmetric switching behavior in our samples (see the [Supplementary Movies](#), demonstrating the dynamics of the nucleation and switching process).

To be able to quantitatively determine the interfacial DMI, we now discuss the magnetic hysteresis loops for these two distinctive geometries as is shown in [Figure 3a–b](#). For Pt/Co/Ir, when no in-plane field is present, the square and triangular patterns show clear perpendicular magnetization with a coercivity $\mu_0 H_z \approx 17.5$ mT and 18.4 mT, respectively. When the in-plane field is additionally introduced, i.e., in this case $|\mu_0 H_x| = 100$ mT, the square pattern exhibits a symmetric loop

Figure 4. (a) Schematic drawing of the triangle-shaped ferromagnetic thin film with indicated the vertical length of $5\ \mu\text{m}$ and the angle γ . The triangle with the different angle $\gamma = 22.5^\circ$, 30° , and 37.5° were measured. Angular dependence of $|H_C/H_0|$ versus H_x in Pt/Co/Ir (b) and AlO_x/Co/Pt (c). The symbols represent average experimental data measured in 2–5 different patterns, but with the nominally same shape and solid lines represent the least-squares fit to eq 1.

with reduced coercive fields of $|\mu_0 H_x| \approx 3.5\ \text{mT}$, irrespective of the polarity of H_x . In contrast, for the triangles indeed an asymmetric hysteresis loop with a positive coercive field $\mu_0 H_C^+ \approx +7.7\ \text{mT}$ and a negative coercive field $\mu_0 H_C^- \approx -2.9\ \text{mT}$ is observed for $\mu_0 H_x = +100\ \text{mT}$. When reversing the in-plane field direction, i.e., $\mu_0 H_x = -100\ \text{mT}$, the loop is shifted into the opposite direction, perfectly in agreement with the concept of DMI-induced tilt of the local edge magnetization. Evidently, we successfully demonstrated asymmetric hysteresis loop with a significant horizontal offset of $|\mu_0 \Delta H_C| \equiv |\mu_0 H_C^+ + \mu_0 H_C^-| \approx 3.95\ \text{mT}$ corresponding to $\Delta H_C/H_0 \approx 21\%$ for $|\mu_0 H_x| = 100\ \text{mT}$ in the triangular-shaped Pt/Co/Ir sample.

For AlO_x/Co/Pt, similar features to the results from Pt/Co/Ir are seen (see again Figure 3). Interestingly we find for the reversed shifts of the magnetic hysteresis loops: at $\mu_0 H_x = +60\ \text{mT}$, the magnetic hysteresis loop is biased to the left, but it is shifted to the right for $\mu_0 H_x = -60\ \text{mT}$. Again, as the reduction or increase of Zeeman energy at the sideways facing surfaces is directly related to the sign the DMI, this contrasting response can be fully accounted for by the opposite tilt of local magnetization due to a different sign of D between the two sample stacks. For AlO_x/Co/Pt, the platinum on top of the magnetic film is believed to play a dominant role in determining the sign of the DMI in this system, whereas Pt/Co/Ir includes it at the bottom of the magnetic film, suggesting an inverted contribution of Pt to the effective DMI⁴⁷ (see Supporting Information IV for more detailed information).

In Figure 3c–f, the variation of H_C as a function of several in-plane magnetic fields H_x is plotted. To be able to compare all system in the same scale and to extract the DMI (as discussed later on), the absolute values of H_C for U-D and D-U are normalized to H_0 , the coercivity without an in-plane field. The decrease of $|H_C/H_0|$ with increasing $|H_x|$ is seen over the entire field range due to the reduction in energy barrier for the magnetization reversal by the external field. In agreement with Figure 3a–b, for square patterns, symmetric features with respect to $H_x = 0$ are measured regardless of the switching polarity for both two samples. On the other hand, asymmetric curves are evidenced again in the triangles. For the case of Pt/Co/Ir, rapid (slow) decrease in H_C/H_0 with increasing $|H_x|$ for negative (positive) H_x is measured for D-U, and vice versa for U-D. By contrast, for AlO_x/Co/Pt, due to the suggested inverted sign of D , obviously an opposite trend is seen in Figure 3d. To support the key aspects of our experimental data, we have performed micromagnetic simulations⁴⁸ as shown in Figure 3e–f (see Method and Supporting Information V). As seen in the Figure 3e, for $D = 2\ \text{mJ/m}^2$, the general features of micromagnetic simulations are in excellent agreement with the

experimental data, except for the quantitative value in H_C/H_0 , which may originate from the discrepancy in material parameters and finite temperature effects in the experimentally measured samples. One of the most remarkable features here is that for $D = 0$, even in the asymmetric triangular structure, H_C versus H_x shows completely symmetric behavior with respect to the $H_x = 0$. Hence, we can conclude that the measured asymmetric hysteresis loops are purely originating from the presence of a DMI-induced tilt of the local magnetization, making these shifted loops a unique and straightforward fingerprint for the existence of the DMI. Moreover, we like to emphasize that the possibility of an oxidation-induced exchange bias effect to explain our data is excluded: it has been experimentally demonstrated that, at the interface between Co/AlO_x, overoxidized Co may lead to an antiferromagnetic CoO and exhibit asymmetric hysteresis loops.⁴⁹ Although the shift in hysteresis loop from the exchange bias could occur in our system, it should be irrespective of the in-plane field and the shape of the microstructure; i.e., this would show up as a shifted hysteresis for $H_x = 0$ and for all geometries including square patterns. However, the measured shift in hysteresis loops shows a very specific dependence on the in-plane field sign and strength as well as the asymmetry in the patterned shape, inconsistent with an exchange bias effect.

As the asymmetric magnetic hysteresis loops are attributed to the relative orientation between local magnetizations near the sideways facing surfaces and the in-plane field, the shift of the hysteresis should necessarily be strongly dependent on the geometric angle of the pattern, i.e., the angle γ indicated in Figure 4a. To examine the dependence of H_C on the geometric angle of the triangle, we measured H_C versus H_x for three different cases $\gamma = 22.5^\circ$, 30.0° , and 37.5° . Here, H_C was chosen as the average of two switching fields of U-D and D-U for oppositely oriented in-plane fields. In all our samples, we found that the amount of asymmetry between positive and negative in-plane fields increases when making the triangle sharper, i.e., for smaller γ (Figure 4b–c). This can be qualitatively explained in terms of the projection of the local magnetizations near the side edge onto the in-plane field axis, which increases with an increasing angle as $\sin(\gamma)$. Not only do these result additionally confirm the proof-of-concept of our tool for probing DMI, it also allows for a more accurate estimate on the strength of DMI, which will be discussed next.

In order to quantitatively address the measured angle dependence and explore the correlation between experimental data and D , we discuss the results on the basis of a well-established half-droplet model,^{44,45,50} which describes the nucleation field of a magnetic domain at the side edge under

Table 1. Comparison of Qualitative D_s with Other Techniques^a

materials system	D_s (pJ/m)	method	A (pJ/m)	ref
Pt(4)/Co(1–2)/AlO _x (2)	1.43	Brillouin light scattering	X	34
Pt(3)/Co(0.6)/AlO _x (2)	1.32	chirality-induced asymmetric nucleation	16	44
Ta(3)/Pt(3)/Co(0.6–1.2)/AlO _x (2)/Pt(3)	1.72	Brillouin light scattering	X	36
Ta(4)/Pt(4)/Co(1.4–2)/AlO _x (2)/Pt(3)	2.20	Brillouin light scattering	X	37
Ta(4)/AlO _x (1.95)/Co(1–1.4)/Pt(4)	–1.15	Brillouin light scattering	X	Supporting Information
Ta(4)/AlO _x (1.95)/Co(1.15)/Pt(4)	–1.63	asymmetric hysteresis	10	
Pt(3)/Co(0.7)/Ir(0–1.3)/Pt(1)	–0.56 to 0.84	asymmetric magnetic domain growth	16	27
SiN/Pt(10)/Co(0.6)/Pt(1)/[Ir(1)/Co(0.6)/Pt(1)] ₁₀ /Pt(3) ^b	1.15	imaging with a STXM	10	22
Ta(4)/Pt(4)/Co(0.8–1.6)/Ir(4)	1.61	Brillouin light scattering	X	Supporting Information
Ta(4)/Pt(4)/Co(1.2)/Ir(4)	2.05	asymmetric hysteresis	10	
Ta(4)/Ir(4)/Co(1.25–3)/AlO _x (2)	0.84	Brillouin light scattering	X	38

^aThe values in parentheses represent the nominal thickness of the layers in nanometers. The values of the exchange stiffness A , presented in the table, are based on the assumptions made in the literature. “X” in the column indicates that A is not used for extracting D . ^bIn the multilayer structure of [Ir/Co/Pt]₁₀, the thickness of a single Co layer is used to obtain $D_s = Dt_{\text{FM}}$.

the application of both in-plane and out-of-plane fields (see [Methods](#) and [Supporting Information VI](#)). Taking into account the geometric angle of a triangle, the normalized coercive field $H_{C,L}$ and $H_{C,R}$ which stands for the left- and right-side dominated nucleation process, respectively, can be written as

$$H_{C,L}/H_0 = \sigma^2(H_x)/[\sqrt{1 - (H_x/H_K)^2}] \quad (1)$$

$$H_{C,R}/H_0 = \sigma^2(H_x \sin(\gamma))/[\sigma^2(0)\sqrt{1 - (H_x/H_K)^2}]$$

where the domain wall energy under the certain in-plane field with DMI is given as $\sigma(H_x) = \sigma_0[\sqrt{1 - (H_x/H_K)^2} - (H_x/H_K + 2D/\sigma_0)\arccos(H_x/H_K)]$.⁴⁴ (see [Supporting Information VI](#)). Here, H_K is the perpendicular uniaxial anisotropy field and σ_0 domain wall energy in the absence of an in-plane field and DMI, which is simply given by $\sigma_0 = 4\sqrt{AK_{\text{eff}}}$ with K_{eff} the anisotropy resulting from the combined interfacial and shape anisotropies. We note that this analytical model is assuming that the Bloch wall profile partially introduced by the in-planed field parallel to the domain wall, is negligible for moderate DMI strengths (as shown in the [Supporting Information VI](#)). Then, H_C/H_0 can be simply expressed in terms of the strength and sign of H_x and the projection of the local magnetizations on the in-plane field axis—governed by $\sin(\gamma)$ —consistent with our earlier qualitative discussion. The best-fit curves to the experimental data using the above equations are shown in [Figure 4c–d](#) by solid curves. The material parameters for M_s and H_K are taken from superconducting quantum interference device-vibrating sample magnetometer (SQUID-VSM) data as performed in unstructured Pt/Co/Ir and AlO_x/Co/Pt thin films. The exchange constant is assumed to be $A = 10$ pJ/m corresponding to the value used in a similar structure.²² The analytically calculated data are generally in good agreement with experimental results, implying the adequacy of the half-droplet model for the present analysis, although some deviation from the equation is allowed due to our simplified approximation made in [eq 1](#). Moreover, the fact that we can simultaneously describe our data for all four structures with different sharpness (defined by γ) by a single unique value of D strongly supports the reliability of the method. From such a simultaneous nonlinear least-squares fits to the results for all angles, we can extract $D = 1.69 \pm 0.03$ mJ/m² and -1.43 ± 0.06 mJ/m² in Pt/Co/Ir (thickness of Co $t_{\text{Co}} = 1.2$ nm) and AlO_x/Co/Pt ($t_{\text{Co}} = 1.15$ nm), respectively, which corresponds to interfacial DMI constant $D_s = Dt_{\text{Co}} = 2.03 \pm 0.04$ pJ/m and

-1.62 ± 0.07 pJ/m, respectively. Here we note that the simultaneous fit for all angles can give a much more accurate and reliable estimation on D_s , as compared to fitting results for a single angle case (we found up to ~15% variations in the estimate of D_s between different angles). We also note that the estimated error in the (simultaneous) fit value is found to be less than 5%, but further systematic errors could arise from the uncertainty in A , which we did not separately address in the current work. With two different exchange stiffnesses, $A = 5$ and 15 pJ/m, which is in the range covered by other reports,^{10,11} we yield an approximately 25% deviation in the values for D as compared to $A = 10$ pJ/m. [Table 1](#) summarizes D_s as measured for similar samples through different techniques. For the case of AlO_x/Co/Pt sample, the magnitude of D_s shows generally good agreement with those from other techniques, even considering the systematic errors from the uncertainty in A . For the Pt/Co/Ir sample, rather diverse values for D_s are found probably due to structural (interfacial) differences between samples used for each measurement.

In order to better access the reliability of the estimated D 's, we also independently performed BLS measurement with the same material systems of Ta(4)/Pt(4)/Co(t_{Co})/Ir(4) and Ta(10)/AlO_x(2.5)/Co(t_{Co})/Pt(4) (nominal thickness in nm), where t_{Co} varied from 0 to 2 nm in a wedge shape (see [Supporting Information VIII](#)). From various thicknesses of Co, we obtained $D_s = 1.61 \pm 0.14$ pJ/m, and -1.15 ± 0.12 pJ/m in Pt/Co/Ir, and AlO_x/Co/Pt, respectively, which are quite comparable to our determined values, implying that our quantification on D is reliable in the present analysis and for this system, where the BLS method has been investigated thoroughly including possible artifacts from asymmetric interface anisotropies. Note that D_s estimated in Pt/Co/Ir from both measurement techniques is slightly lower than $D_s = 2.20 \pm 0.02$ pJ/m in Pt/Co/AlO_x as measured in our previous study. Interestingly, this is in contrast to the theoretical prediction⁴⁷ as well as experimental data.^{22,27} In these studies, it is suggested that the effective DMI of Pt/Co/Ir is enhanced with respect to Pt/Co/AlO_x, as the sign of D at Ir/Co is opposite to that for Pt/Co. In other words, Co sandwiched between Pt and Ir with an opposite sign of D at each interface is expected to yield an increase of the total effective D . Our conflicting result could arise from many possible origins. One could speculate the enhancement of D is due to the interface between Co/oxide. It has been theoretically predicted¹⁵ that a Co/oxide interface could also give a significant additional

contribution to the effective DMI. From recent *ab initio* calculations, the magnitude of D at Ir/Co interfaces⁴⁷ has been predicted to be very small and comparable to the value as predicted at the Co/oxide interface.¹⁵ The large D in Pt/Co/ AlO_x then, can be understood when a larger additional contribution from Co/ AlO_x interfaces arises as compared to the Pt/Co interfaces. Another possibility is that the D at the Ir/Co interface has the same sign as that at Pt/Co, in contrast to previous results.^{22,27} Indeed, recent experiments on Ir/Co/ AlO_x performed by BLS³⁸ have shown that Ir/Co/ AlO_x has the same sign of D as compared to Pt/Co/ AlO_x . The origin of this effect has not been fully understood, and obviously, different sample stacks and growth conditions may strongly influence the interfacial quality which in turn should be reflected in changes of the DMI.

In the present study, although only triangular-shaped microstructures were used as a prototype to demonstrate asymmetric magnetic hysteresis, we would like to note that this effect is also obtainable by introducing other shapes with a lateral asymmetry, such as a pentagon or a half-circular object. More importantly, our micromagnetic simulations performed for various lateral sizes of the triangles reveal that the shift in hysteresis loops is insensitive to the size of microstructures (see Supporting Information V), at least in a range of structures with lateral dimensions around 1–2 μm . This shape and size independence emphasizes the strength of our DMI probe in the possibility of handling a variety of microstructures including larger patterns for fast and reliable access. However, one should note that, in the latter case, the nucleation should predominantly take place at sideways facing edges instead of at local sites within the microstructure⁴⁴ or at the sharp edge point of the triangular structure. This requires sufficiently clean materials with sharp enough edges to make a meaningful analysis of DMI, which is rather standard in most deposition and processing of ultrathin magnetic films. Nevertheless, when dealing with considerable edge roughness, e.g., creating a different nucleation barrier at each side of the triangle, we experimentally observed a rather different effect on the coercivity, although still affected by the presence of DMI (see Supporting Information VII). Next, the asymmetric magnetic hysteresis loops were quasi-statically demonstrated by using a wide-field Kerr microscope. However, more standard laboratory tools for the measurement of magnetic hysteresis loops, such as SQUID, VSM, and polar MOKE equipped with two-dimensional magnets, are also applicable to measure DMI, provided that a large enough signal-to-noise ratio is achieved by introducing a sufficiently large two-dimensional array of asymmetric magnetic objects. Finally, we would like to emphasize again that the interfacial DMI originating from atomic interactions at (sub) nanometer scale can be measured in a macroscopic manner without the need for any imaging technique or measuring its dynamics, providing a more straightforward and faster exploration of the DMI.

In summary, we establish a novel robust, simple, and reliable approach to quantify the DMI in thin film structures. We successfully demonstrate a shift in magnetic hysteresis loops arising from the DMI, by introducing a lateral asymmetry in microstructures and applying an additional in-plane bias field. In our measurement, the presence of DMI and its sign can be straightforwardly determined in a robust manner by measuring the asymmetric behavior in switching fields, clearly depending on the polarity of the in-plane field and magnetization. Furthermore, with help of a half-droplet model, we are able

to estimate the strength of DMI in two different sample stacks of Pt/Co/Ir and AlO_x /Pt/Co, which corresponds to $D_s = 2.03 \pm 0.04$ pJ/m and -1.62 ± 0.05 pJ/m, respectively, which is quite comparable to one obtained from other reported techniques. Our results open a novel avenue to a rapid and reliable screening of materials and systems for DMI, and thus overcome the key obstacle to design devices with tailored DMI for ultimate topological stability and efficient manipulation as required for next-generation devices for nanoelectronics.

Method Section. Sample Fabrication and Characterization of Magnetic Properties. We prepared square- and triangle-shaped magnetic thin films on a silicon wafer coated with 100 nm thick SiO_2 . The magnetic thin films were deposited using DC magnetron sputtering with layer structures of Si(substrate)/Ta(4)/Pt(4)/Co(1.2)/Ir(4), and Si(substrate)/Ta(10)/ AlO_x (2.5)/Co(1.15)/Pt(4), where the number in parentheses denotes nominal thicknesses in nanometers. The AlO_x layer was obtained from plasma oxidation of Al layer as deposited on Ta. The plasma oxidation process was carried out for 10 min in an in situ chamber at a 0.1 mbar background pressure of oxygen and a power of 15 W. The AlO_x /Co/Pt sample was annealed at 250 °C for 30 min to introduce perpendicular magnetization. To improve pattern quality, we employed a PMMA/HSQ bilayer for a mask, prepared by the high-resolved electron beam lithography and polymer reactive ion etching (RIE), and Ar^+ ion milling is followed afterward. Magnetic properties were measured on unstructured continuous films with superconducting quantum interference device-vibrating sample magnetometry (SQUID-VSM). The saturation magnetizations of ~ 920 and 1490 $\text{emu}/\text{cm}^{-3}$ were measured for Pt/Co/Ir and AlO_x /Co/Pt, respectively. All of the films considered here exhibited perpendicular magnetic anisotropy (PMA), with their remanent state fully magnetized in the direction perpendicular to the substrate. The perpendicular uniaxial anisotropic field $\mu_0 H_K$ were measured around 0.7 and 0.45 T for Pt/Co/Ir and AlO_x /Co/Pt, respectively.

Magnetic Hysteresis Loops. Magnetic hysteresis loops were acquired using a wide-field polar Kerr microscopy equipped with two-dimensional magnets. For the acquisition of reliable data set, the measurement of magnetic hysteresis loop was repeated over 10 cycles for each pattern, and 2–5 different patterns but with the nominally same shape were used for the measurement, and average of the data was used in Figure 4. The coercive field of each hysteresis loop was automatically extracted from a homemade code fitting of the normalized MOKE signal to $2(1/(1 + \exp((H_z - A)/B))) - 1$, where A and B are the fitting parameters. To get rid of artifacts which might arise from an unintentional misalignment of the in-plane magnet, square patterns, which should exhibit the symmetric switching fields, were measured before the measurements on triangle patterns.

Micromagnetic Simulations. Micromagnetic simulations were carried out by using OOMMF code incorporating the interfacial DMI term. We used squares with lateral size of $L = 1$ μm and triangles with $L = 1$ μm and $\gamma = 30^\circ$ for the calculation. We also employed a unit cell size of $4 \times 4 \times 1.2$ nm^3 and the typical materials parameters for Co: saturation magnetization $M_s = 1090$ kA/m, exchange stiffness $A = 10$ pJ/m, and perpendicular magnetocrystalline anisotropy $K_u = 1.25$ MJ/m, and a damping constant $\alpha = 0.5$ to obtain rapid convergence for quasi-static investigations.

Analytical Model for the Extraction of D Value. For rigid quantitative analysis of the experimental data, we adopted a half-droplet model for the estimation of nucleation field (see [Supporting Information](#)). In the presence of DMI and an in-plane bias field H_x , the nucleation field at the edge can be expressed by

$$H_C = \frac{\pi t (\sigma_{DW})^2}{4\mu_0 M_S p k_B T \sqrt{1 - (H_{in}/H_K)^2}} \quad (2)$$

Here, σ_{DW} is domain wall energy density under the in-plane field and DMI, and t is thickness of magnetic thin film, μ_0 a magnetic permeability, M_S saturation magnetization, H_K in-plane saturation field, and $k_B T$ are Boltzmann constant and temperature. The p is defined by the switching time t simply expressed by $t = t_0 e^p$. In the experiments, the sweeping time of the out-of-plane field is fixed as 100 ms (~ 1 mT/s), and the switching time is typically in the order of $\sim$ ns; therefore, p can be approximated to ~ 18 . For a moderate DMI strength, the domain wall energy density σ_{DW} can be approximated into

$$\sigma_{DW} = \sigma_0 \left(\sqrt{1 - (H_{in} \cos \varphi / H_K)^2} + \left(H_{in} \cos \varphi / H_K + \frac{2D}{\sigma} \right) \left(\arcsin(H_{in} \cos \varphi / H_K) - \frac{\pi}{2} \right) \right) \quad (3)$$

where φ is azimuthal angle of the in-plane field with respect to the axis normal to domain wall.

The D is estimated from the least-squares fit through experimental data of all angles in [Figure 4b](#) to [eq 3](#), where $\varphi = \pi/2 - \gamma$.

Brillouin Light Scattering. The Brillouin light scattering spectra were independently performed. A p -polarized single longitudinal mode laser with a power of 300 mW and a wavelength of 532 nm was used as a light source. In order to measure Damon–Eshbach mode, an external static magnetic field was further applied parallel to the film surface. The backscattered light from the sample is focused and collected. The s -polarized light is passed through the interferometer and the photomultiplier tubes. All measurements are performed at room temperature.

■ ASSOCIATED CONTENT

■ Supporting Information

The Supporting Information is available free of charge on the [ACS Publications website](#) at DOI: [10.1021/acs.nanolett.6b01593](#).

Detailed sample preparation, characterizations, micro-magnetic simulations, analytical derivation of the angle-resolved droplet model, BLS measurement ([PDF](#))

Movie for the chirality-induced asymmetric switching in Pt/Co/Ir : a square under the positive ([AVI](#)) and negative ([AVI](#)) in-plane field

Movie for the chirality-induced asymmetric switching in Pt/Co/Ir : a triangle under the positive ([AVI](#)) and negative ([AVI](#)) in-plane field

Movie for the chirality-induced asymmetric switching in AlO_x/Co/Pt : a square under the positive ([AVI](#)) and negative ([AVI](#)) in-plane field

Movie for the chirality-induced asymmetric switching in AlO_x/Co/Pt : a triangle under the positive ([AVI](#)) and negative ([AVI](#)) in-plane field

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: cyyou@dgist.ac.kr.

Author Contributions

C.Y.Y. conceived the main idea. C.Y.Y., J.S.K., and D.S.H. contributed to the design of the experiment. D.S.H. performed the sample fabrication, measurements with the Kerr microscope and analysis on the experimental data with the help from Y.Y. and K.J.K. N.H.K., J.C., and J.S.K. conducted BLS measurement and relevant data analysis. C.Y.Y. and D.S.H. performed micromagnetic simulations and developed an analytical model. D.S.H. wrote the manuscript with review and input from the other authors. H.J.M.S., B.K., and C.Y.Y. supervised the study. All authors discussed the results and commented on the manuscript.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We acknowledge support from the research program of the Foundation for Fundamental Research on Matter (FOM), which is part of The Netherlands Organisation for Scientific Research (NWO), the National Research Foundation of Korea (Grant Nos. 2015M3D1A1070465 and 2014K2A5A6064900), the German Research Foundation (DFG via the joint Korean-German project KL1811/14 and the SFB TRR 173 Spin+X) and the EU (MultiRev ERC-PoC-2014 665672).

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