Interfacial atomic layers for full emergence of interfacial Dzyaloshinskii–Moriya interaction

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Abstract
Interfacial phenomena play decisive roles in modern science and technology as the scale of the material shrinks down to a few atomic layers. Such minute nanostructures require a more comprehensive understanding beyond the conventional concepts of interfaces and interfacial phenomena generated at interfaces. From a series of a few-atomic-layer-thick magnetic films, we experimentally demonstrate that, contrary to the common notion, interfacial phenomena require a finite thickness for their full emergence. The layer-thickness dependences reveal that the interfacial Dzyaloshinskii–Moriya interaction (DMI) begins to appear with increasing thickness, and emerges completely at a thickness of 2–3 atomic layers, at which the magnitude is maximized. This result implies that the DMI is suppressed when the “bulk” layer adjacent to the interface is thinner than the threshold thickness. The existence of the threshold thickness indicates the need to refine conventional perspectives on interfacial phenomena, and imposes the lowest structural bound and optimum thickness to maximize interfacial effects for technological applications.

Introduction
The Dzyaloshinskii–Moriya interaction (DMI) has attracted much attention because of its peculiar properties for realizing chiral spin structures in magnetic systems with broken inversion symmetry1–10. These chiral spin structures, such as chiral magnetic domain walls3 and magnetic skyrmions9, have been extensively studied as the building blocks of emerging magnetic memory and logic devices3,4. A sufficiently large DMI is essential for highly efficient domain-wall motion via current-induced spin–orbit torques (SOTs)5,6, and for the topological protection of the skyrmion stability4. Hence, extensive efforts have been made to engineer a large DMI and understand its physical origin.

Interest in DMIs has exploded recently because of the discovery of large DMIs at the interfaces of magnetic multilayered systems5,11–13. By using these systems, the interfacial DMI strength can be engineered to be strong enough to demonstrate magnetic skyrmions at room temperature2. Beyond the original concept based on the atomic three-site anisotropic superexchange interaction with spin–orbit coupling8,9, recent studies have suggested more comprehensive mechanisms with different governing parameters, such as the work function difference at the interface11, orbital characteristics12,14, charge distribution and interface dipole moment15, and Rashba effect16,17. Although the most decisive parameters and mechanisms are still under debate, it is commonly accepted that the interfacial DMI has to be generated by broken inversion symmetries at the interfaces of heavy metals with large spin–orbit coupling9,13

Similar to other interfacial effects, it is commonly believed that the interfacial DMI arises at the interface itself, and thus, its strength average over the ferromagnetic layer is inversely proportional to the layer thickness. Therefore, a system with a thinner layer is expected to show a larger DMI strength, as observed by recent studies18–20. Such monotonic dependence has been
explained by Yang et al.\textsuperscript{21} based on the atomic-layer-resolved calculations of Pt/Co bilayers by suggesting that the interfacial DMI is generated mostly at the first nearest Co atomic layer to the interface, and thus, the DMI strength is inversely proportional to the layer thickness down to an atomic monolayer. In contrast, another atomic-layer-resolved calculation by Jia et al.\textsuperscript{22} predicted a significant contribution of the second nearest Co atoms to the DMI in Pt/Co bilayers, resulting in a nonmonotonic dependence of the DMI strength on the layer thickness. Both studies agree that the interfacial phenomena originate from a few atomic layers near the interfaces, but the detailed contributions of each atomic layer to the DMI with the refined DMI mechanisms are still under debate.

In general cases, even at ideal interfaces with abrupt structural boundaries, the crossover between different material properties requires a finite nonzero thickness, over which the interfacial phenomena are generated. Such a crossover thickness might arise from the interactions between the conduction and orbital electrons with a redistribution of the charge and dipole moment\textsuperscript{10,13,15}, or spin chiral mechanisms with the Rashba effect\textsuperscript{16,17}, resulting in off-nearest-site atomic interactions\textsuperscript{25} beyond the original concept of atomic interactions between the nearest sites.

Materials and methods

Sample fabrication

Two different series of films were prepared. The full-layer stacks of the films are 1.5-nm Ta/3-nm Pt/t\textsubscript{Co} Co/5-nm W/2-nm AlO\textsubscript{x} with different ferromagnetic Co-layer thicknesses (t\textsubscript{Co}) in the range from 0.7 to 1.2 nm for the former Pt/Co/W films and 1.5-nm Ta/3-nm Pt/t\textsubscript{Co} Co/0.6-nm Cu/5-nm W/2-nm AlO\textsubscript{x} with different t\textsubscript{Co} values in the range from 0.3 to 0.8 nm for the latter Pt/Co/Cu/W films. All samples were deposited by dc-magnetron sputtering on Si wafers with a 100-nm-thick SiO\textsubscript{2} layer. The lowermost Ta and uppermost AlO\textsubscript{x} layers are employed as seed and protection layers, respectively. The AlO\textsubscript{x} layers are formed by natural oxidation after depositing Al layers. To keep a similar film quality, the sputtering conditions were carefully maintained to be the same for all the films with an Ar working pressure of \~2 mTorr and sputtering power of \~10 W. To measure the maximum strength of the SOT efficiency (\varepsilon\textsubscript{0}) and DMI-induced effective magnetic field (H\textsubscript{DMI}), 20-\textmu m-wide and 350-\textmu m-long microwires were patterned by photolithography and ion-milling processes. To inject electric current into the microwires, 5/100-nm-thick Cr/Au electrodes were deposited by a lift-off process.

Sample characterization

The magnetic anisotropy field (H\textsubscript{K}) and saturation magnetization (M\textsubscript{S}) were measured by means of a vibrating sample magnetometer, where H\textsubscript{K} and M\textsubscript{S} were estimated by the saturation magnetic field and moment from hard- and easy-axis hysteresis loops, respectively. One can then obtain effective perpendicular magnetic anisotropy (K\textsubscript{eff}) from the relation K\textsubscript{eff} = \mu\textsubscript{0}M\textsubscript{S}H\textsubscript{K}/2. The measured M\textsubscript{S} and H\textsubscript{K} and estimated K\textsubscript{eff} are listed in Supplementary Table 1.

The values of H\textsubscript{DMI} and \varepsilon\textsubscript{0} were quantified by a measurement scheme\textsuperscript{11} based on the variation in the SOT efficiency (\varepsilon) with respect to the in-plane magnetic field (H\textsubscript{x}). In this scheme, a domain wall was first created in a magnetic microwire, and then, a domain-wall depinning field (H\textsubscript{dep}) in the direction perpendicular to the film plane) from the initial position was detected by sweeping the external magnetic field. By measuring H\textsubscript{dep} under the application of several different current densities (J), one can obtain \varepsilon via the relations \mu\textsubscript{0}H\textsubscript{dep}(J) = \mu\textsubscript{0}H\textsubscript{dep} + \varepsilon J\textsubscript{0}, where \mu\textsubscript{0}H\textsubscript{dep} is the depinning field without an electric current, and \varepsilon J\textsubscript{0} corresponds to the effective magnetic field (H\textsubscript{eff} in the direction perpendicular to the film plane) caused by the SOT. By repeating these procedures by sweeping H\textsubscript{x}, one can obtain an \varepsilon–\mu\textsubscript{0}H\textsubscript{x} plot, as shown in Fig. 1.

Under the application of an electric current, the damping-like SOT generates the effective magnetic field in the direction of \~\hat{m} \times \hat{s}, where \~\hat{m} is the unit vector of the magnetization inside the domain wall and \hat{s} is the unit vector of the spins associated with the SOT\textsuperscript{23}. For the case that \varepsilon is parallel to the x axis, \hat{s} is parallel to the y axis, and therefore, H\textsubscript{eff} along the z axis is proportional to the x component (m\textsubscript{x}) of the magnetization inside the domain wall, i.e., H_{\textsubscript{eff}z} \propto m\textsubscript{x} \hat{x} \cdot \hat{y}. Since \mu\textsubscript{0}H_{\textsubscript{eff}z} = \varepsilon J\textsubscript{0}, one can then obtain the relation \varepsilon \propto m\textsubscript{x}. By sweeping H\textsubscript{x}, the domain wall varies from one Néel-type configuration (m\textsubscript{n} = -1) to another (m\textsubscript{n} = 1) through a Bloch-type configuration (m\textsubscript{n} = 0). Therefore, the \varepsilon–\mu\textsubscript{0}H\textsubscript{x} plots exhibit the typical behavior of two saturation regimes with the Néel-type domain-wall configurations (\varepsilon = \pm \varepsilon\textsubscript{0}) across the Bloch-type domain-wall configuration (\varepsilon = 0). Since the Bloch-type domain-wall configuration appears when H\textsubscript{x} exactly compensates for H\textsubscript{DMI} (i.e., H\textsubscript{x} + H\textsubscript{DMI} = 0), the x intercept (i.e., \varepsilon = 0) indicates the magnitude of H\textsubscript{DMI} (black vertical line), as shown in Fig. 1. In addition, \varepsilon\textsubscript{0} is quantified from the saturation values in the Néel-type domain-wall configurations.

Results and discussion

Observation of quick DMI decay with decreasing Co-layer thickness

To demonstrate the existence of a finite thickness for DMI generation, we experimentally examined the DMI strength with respect to t\textsubscript{Co}. Figure 2 summarizes the measured values of H\textsubscript{DMI} (a) and \varepsilon\textsubscript{0} (b) obtained from the plot in Fig. 1 with respect to t\textsubscript{Co}. The measured H\textsubscript{DMI} and
$\varepsilon_0$ are listed in Supplementary Table 1. Due to the interfacial nature, it is expected that both the strengths of the DMI and SOT increase as $t_{Co}$ decreases via an increase in the surface-to-volume ratio.\textsuperscript{6,18,19,24,25} This expectation truly occurs for the case of the SOT, as seen in Fig. 2b, that $\varepsilon_0$ increases monotonically as $t_{Co}$ decreases. However, interestingly, a quite distinct behavior occurs for the case of the DMI. Figure 2a shows that $H_{DMI}$ quickly decays in the thinner $t_{Co}$ regime, whereas the typical interfacial behavior of the inverse proportionality appears in the thicker $t_{Co}$ regime. The critical thickness between these two regimes is found to be ~0.8 nm, where the maximum $H_{DMI}$ strength is achieved.

**Consideration of possible structural origins**

We first examined structural imperfections as the possible origins of this peculiar behavior. Among the possible structural imperfections, the discontinuous island growth of the Co layer is not observed in our films, but rather a continuous crystalline structure of the Co layer appears in a cross-sectional transmission electron microscope (Cs-TEM), which was used to confirm the degree of the continuity and roughness at the interface. The cross-sectional image visualizes a continuous crystalline structure of the Co layer, even for the thinnest case of the Co layer, as shown in Fig. 3a, and the signaling effective magnetic layer remains continuous for this series of films. If the Co layer is discontinuous (see Supplementary Fig. 1), then there is a possibility for a reduction of the DMI strength (see Supplementary Fig. 2) due to the decreasing number of atomic pairs between Co and the atoms of the

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**Fig. 1** Plots of $\varepsilon$ with respect to $\mu_0 H_x$ for the series of Pt/Co/W films with different $t_{Co}$ as indicated. The vertical error bars are the standard deviation of several repeated measurements. The black vertical lines indicate the antisymmetric center with $\varepsilon = 0$ for each curve. The curved solid lines guide the eyes.

**Fig. 2** Plot of $H_{DMI}$ and $\varepsilon_0$ with respect to $t_{Co}$ for the series of Pt/Co/W films. a Plot of $\mu_0 H_{DMI}$ with respect to $t_{Co}$. b Plot of $\varepsilon_0$ with respect to $t_{Co}$. The horizontal error bars correspond to the experimental accuracy of the Co-layer thickness, and the vertical error bars are the standard deviation of several repeated measurements.

**Fig. 3** Cross-sectional transmission electron microscopy image of the interfacial atomic layers. a Wide-range image of the Pt/Co/W film with the thinnest Co layer ($t_{Co} = 0.7$ nm). b Image magnified on the Co layer and its interfaces.
nonmagnetic material, as described in Supplementary Information. However, a closer look indicates smooth atomic-layer boundaries and clear contrast at the interfaces, as visualized in Fig. 3b, and thus, a reduced number of atomic pairs due to a discontinuous Co layer is unlikely the major origin of the quick $H_{\text{DMI}}$ decay.

Structural variation, such as the coherent-to-incoherent transition of crystalline structures, was also examined by the measurement of $K_U^{\text{eff}}$ with respect to $t_{\text{Co}}$. The measurement results (see Supplementary Fig. 3a) indicate that the coherent-to-incoherent transition occurs at approximately $t_{\text{Co}} \approx 1.2$ nm, which is very different from the critical thickness for quick $H_{\text{DMI}}$ decay. In addition, $K_U^{\text{eff}}$ exhibits a gradual thickness dependence that is dissimilar to the peculiar $H_{\text{DMI}}$ variation. Hence, the coherent-to-incoherent transition associated with the $K_U^{\text{eff}}$ variation is unlikely to share the origin of the quick $H_{\text{DMI}}$ decay.

To check whether the quick $H_{\text{DMI}}$ decay is possibly caused by a reduction of the magnetically effective thickness ($t_m^{\text{Co}}$) near the interface, we examined the reduced amount of $t_m^{\text{Co}}$ with respect to the deposited thickness $t_{\text{Co}}$. If W atoms penetrate into the Co layer, then they may alter the properties of the Co layer nearby, forming a reduced thickness ($\delta$) or the so-called dead layer. Even for the continuous Co layers, there may be the possibility of a reduced number of atomic pairs effectively by the penetration of W atoms. For this test, $M_S$ was measured by means of a vibrating sample magnetometer. Figure 4a shows the results by plotting $M_{\text{S}t_{\text{Co}}}$ with respect to $t_{\text{Co}}$. The linear dependence between $M_{\text{S}t_{\text{Co}}}$ and $t_{\text{Co}}$ manifests that most of the films have similar $M_S$ values, considering the magnetic layer volume from $t_m^{\text{Co}}$, as shown in other experiments with Pt/Co trilayer systems.

Then, as guided by the blue linear line of the best fit, the nonzero $x$ intercept indicates the amount of $\delta$, which is quantified as $\approx 0.26 \pm 0.01$ nm. Notably, $\delta$ is noticeably smaller than the critical thickness for the quick $H_{\text{DMI}}$ decay. In addition, the roughness/mixing does not exceed the effective magnetic layer thickness (approximately two atomic layers for this sample), as shown in Fig. 3. This situation is similar to the situation depicted on the right side in Supplementary Fig. 1a, with a continuous Co layer even if the W atom penetrates into the Co layer to some extent. Therefore, the results indicate that the mechanism associated with the $\delta$ formation is not directly responsible for the quick $H_{\text{DMI}}$ decay.

Analysis with the magnetically effective thickness

To obtain detailed insights into the origin more quantitatively, the DMI strength ($D$) is estimated by the relation $D = \mu_0 H_{\text{DMI}} M_S^2 \lambda$, where $\mu_0$ is the permeability constant and $\lambda$ is the domain-wall width. The measured $D$ values are listed in Supplementary Table 1. The values of $\lambda$ are estimated by the equation of the Bloch-type domain-wall width, i.e., $\lambda = (A_{\text{ex}}/K_{U0}^{\text{eff}})^{1/2}$ under the assumption that the exchange stiffness ($A_{\text{ex}}$) remains constant ($\approx 22$ pJ/m). We also confirmed that the possible variation in $A_{\text{ex}}$ with respect to ferromagnetic layer thickness does not change the main trend in the $D$ variation. Then, $D$ is analyzed in terms of $t_m^{\text{Co}}$ ($= t_{\text{Co}} - \delta$). The inverse proportionality between $D$ and $t_m^{\text{Co}}$ is examined by plotting $Dt_m^{\text{Co}}$ with respect to $t_m^{\text{Co}}$, as

![Fig. 4 Plot of $M_{\text{S}t_{\text{Co}}}$, $D_{\text{Co}}$, and $\delta_{\text{f}t_{\text{Co}}}$ with respect to $t_{\text{Co}}$ for the series of Pt/Co/W films. a Plot of $M_{\text{S}t_{\text{Co}}}$ with respect to $t_{\text{Co}}$ (upper label) and $t_{\text{Co}}$ (lower label). The blue line shows the linear fit. The $x$ intercept for $t_{\text{Co}}$ indicates $\delta$. b Plot of $D_{\text{Co}}^{\text{m}}$ with respect to $t_{\text{Co}}$. The blue and yellow shaded areas show the thicker and thinner $t_{\text{Co}}$ regimes, respectively. The blue horizontal line shows the average value over the thicker $t_{\text{Co}}$ regime. c Plot of $\delta_{\text{f}t_{\text{Co}}}$ with respect to $t_{\text{Co}}$. The blue horizontal line shows the average value over both regimes. In all the plots, the horizontal error bars are the experimental accuracy of the Co-layer thickness, and the vertical error bars are the standard deviation of several repeated measurements.](Image 332x331 to 512x697)
shown in Fig. 4b. This scaled plot is useful for decomposing the surface contribution \( (D_s) \) and the volume contribution \( (D_v) \) via the relation \( D_{\text{Co}} = D_s + D_{\text{vol}} \). Interestingly, the plot again exhibits two different regimes. In the thicker \( t_{\text{Co}}^m \) regime (blue area), \( D_{\text{Co}}^{m} \) shows a negligible variation with respect to \( t_{\text{Co}}^m \), indicating that \( D \) has a negligible volume contribution, i.e., \( D_v \approx 0 \). Therefore, the surface contribution is mostly responsible for the DMI, i.e., \( D_{\text{Co}}^{m} \approx D_s \). In addition, the saturation values of \( D_{\text{Co}}^{m} \) show that \( D_s \) is fully generated in this regime, resulting in the inverse proportionality between \( D \) and \( t_{\text{Co}}^m \).

On the other hand, in the thinner \( t_{\text{Co}}^m \) regime (yellow area), \( D_{\text{Co}}^{m} \) becomes smaller than the saturation value. The critical thickness between these two regimes is \( \approx 0.55 \text{ nm (} \approx 3 \text{ atomic layers)} \), which is consistent with the prediction based on the atomic-layer-resolved calculation by Jia et al.\(^{22} \). Notably, the other atomic-layer-resolved calculation by Yang et al.\(^{21} \) can be extended to explain such insufficient DMI generation for the case of an incomplete atomic interface formation with large vacancies (see Supplementary Information), although this case is unlikely for our films with reasonably clear atomic-layer boundaries, as shown in Fig. 3b. It is not easy to quantify the exact contributions of these mechanisms, but both cases suggest that the incomplete generation of the interfacial DMI might be a possible origin of the quick DMI decay in the thinner regime.

The distinct thickness dependences of the DMI and SOT provide another hint to understand the interfacial and bulk phenomena, and the possibility of the independent engineering of the DMI and SOT. In contrast to the DMI that should be generated near the interfaces, the SOT is known to have two sources: at the interfaces via the Rashba effect\(^{30} \) known as the source of the damping-like torque, and from the adjacent nonmagnetic layers via the spin-Hall effect\(^{24,31} \) known as the source of the field-like torque. The plot of \( \epsilon_0 t_{\text{Co}}^m \) with respect to \( t_{\text{Co}}^m \) shows that \( \epsilon_0 t_{\text{Co}}^m \) remains unchanged within the error bars over the whole thickness range. The slight reduction in \( \epsilon_0 t_{\text{Co}}^m \) with respect to \( t_{\text{Co}}^m \) might be attributed to the increase in spin current absorption in Co as \( t_{\text{Co}}^m \) becomes too thin\(^{32} \). This observation indicates that the SOT is less sensitive to interface formation in our samples, and thus, the damping-like torque is the main source of the SOT originating mainly from the spin-Hall current from the outer nonmagnetic layers in these films. Unlike the SOT in our samples, the DMI is sensitive to the thickness of the atomic layers in the vicinity of the interface, indicating that the quick DMI decay might be attributed not only to the interface characteristics, but also to incomplete DMI generation with Co layers thinner than the critical thickness.

**Confirmation with other series films without a reduction of the magnetically effective thickness**

To avoid any possible artifacts associated with the mechanisms of the \( t_{\text{Co}}^m \) reduction, another series of films with Pt/Co/Cu/W stacks was prepared with different \( t_{\text{Co}} \) values. In these films, a Cu layer (0.6 nm) was inserted at the Co/W interface to suppress intermixing between the Co and W atoms\(^{33} \). The same measurements were performed on Pt/Co/Cu/W samples, and the measured parameters are listed in Supplementary Table 2. The Co thickness dependence of \( K_U^{\text{eff}} \) in the Pt/Co/Cu/W samples is similar to that of the Pt/Co/W samples, as is also shown in Supplementary Fig. 3b. The same typical \( \epsilon \) variation with respect to \( H_x \) was observed from this series of films, as shown in Fig. 5.

Figure 6a shows a plot of \( M_s t_{\text{Co}} \) with respect to \( t_{\text{Co}} \). It is noteworthy that the \( x \) intercept of
thinner $t_{Co}^m$ regime below 0.4 nm, whereas the SOT remains unchanged over the whole $t_{Co}^m$ range. The same behaviors exist for both cases with and without the $t_{Co}^m$ reduction, which confirms that the peculiar thickness dependence of the DMI is not associated with the mechanisms of the $t_{Co}^m$ reduction.

In summary, we demonstrate the existence of a finite thickness for full DMI generation at interfaces from two different series of films Pt/Co/W and Pt/Co/Cu/W with various Co-layer thicknesses. Both series of films exhibit the same behavior of quick DMI decay with a decrease in the ferromagnetic layer thickness below a threshold thickness. Such a quick DMI decay has to be caused by insufficient DMI generation at interfaces with ferromagnetic layers that are thinner than the threshold. Therefore, the full emergence of the interfacial DMI requires a finite-threshold thickness, in contrast to the commonly accepted concept that the interfacial phenomena are generated exactly at the interfaces. Interestingly, both series of films provide a similar threshold thickness of ~2–3 atomic layers, signaling the possibility of common key mechanisms for DMI generation. These results provide a technological criterion to optimize the DMI strength with a maximum at the threshold thickness. In addition, the distinct behaviors between the DMI and SOT manifest that the DMI is generated in the vicinity of the interface, whereas the SOT is generated mainly by the spin-Hall effect from the outer nonmagnetic layers in our series of films. These distinct behaviors provide the perspective of a systematic study on the interfacial DMI and bulk SOT phenomena, providing a method for the independent tuning of the DMI and SOT for spintronic applications.

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Conflict of interest
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