Skyrmion Stabilization at the Domain Morphology Transition in Ferromagnet/Heavy Metal Heterostructures with Low Exchange Stiffness

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Herein, the experimental observation of micrometer-scale magnetic skyrmions at room temperature in several Pt/Co-based thin film heterostructures designed to possess low exchange stiffness, perpendicular magnetic anisotropy, and a modest interfacial Dzyaloshinskii–Moriya interaction (iDMI) is reported. It is found both experimentally and by micromagnetic and analytic modeling that a low exchange stiffness and modest iDMI eliminates the energetic penalty associated with forming domain walls in thin films. When the domain wall energy density approaches negative values, the remnant morphology transitions from a uniform state to labyrinthine stripes. A low exchange stiffness, indicated by a sub-400 K Curie temperature, is achieved in Pt/Co, Pt/Co/Ni, and Pt/Co/Ni/Re structures by reducing the Co thickness to the ultrathin limit (<0.3 nm). Similar effects occur in thicker Pt/Co/Ni,Cu1−x structures when the Ni layer is alloyed with Cu. At this transition in domain morphology, skyrmion phases are stabilized by small (<1 mT), perpendicular magnetic fields, and skyrmion motion in response to spin–orbit torque is observed. While the temperature and thickness-induced morphological phase transitions observed are similar to the well-studied spin reorientation transition that occurs in the ultrathin limit, the underlying energy balances are substantially modified by the presence of an iDMI.

1. Introduction

In recent years, there has been an intense study of the magnetic textures known as skyrmions, given that their quantized, winding-like spin structure leads to novel behaviors that are attractive from both fundamental and applied viewpoints. Particularly, the unique spin topology of skyrmions lends them a degree of topological stability and offers avenues toward their efficient motion—making them potentially relevant in developing next-generation data storage and novel computation schemes. Previous work has shown that while it is possible to stabilize skyrmion phases through an intricate balance of the ferromagnetic (FM) exchange, dipolar, and anisotropy energies of a material system, the Dzyaloshinskii–Moriya interaction (DMI)—an antisymmetric exchange interaction that favors the orthogonal canting of neighboring magnetic spins—often plays a crucial role. For example, the DMI associated with the noncentrosymmetric B20-type crystal structure was central to the first experimental observation of a skyrmion phase.

The interfacial DMI (iDMI) that develops at the interface between thin FM layers and nonmagnetic heavy metals (HMs) with significant spin–orbit coupling also assists with the formation of skyrmion phases. There have been numerous reports on multi-repetition FM/HM heterostructures with perpendicular magnetic anisotropy (PMA), where a large iDMI assists with the formation of skyrmion phases, and the multiple repetitions of the unit structure increases the total volume of magnetic material, leading to enhanced skyrmion stability. Additionally, there have been several reports of skyrmion phases in single magnetic layer HM/FM/HM systems with modest iDMI, where the FM layer thickness was chosen to be large enough that the system is close to experiencing a spin reorientation transition (SRT), and consequentially has a low effective PMA characterized by \( K_{eff} = K_u - \frac{\mu_b M_s^2}{2} \), where \( K_u \) is the intrinsic PMA constant and \( \frac{\mu_b M_s^2}{2} \) reflects the shape anisotropy of a thin film. The domain wall energy density \( \sigma \) in thin film systems with iDMI is often approximated as \( 4\sqrt{A_{ex} K_{eff} - \pi |D| \lambda} \), where \( A_{ex} \) and \( |D| \) are the exchange stiffness and magnitude of the iDMI energy density, respectively. Thus, by lowering \( K_{eff} \) while maintaining \( |D| \) or increasing \( |D| \) while maintaining substantial \( K_{eff} \), it is possible to reduce \( \sigma \), the energy barrier to forming multidomain states, including the skyrmion phase.

In this work, we explore the approach of reducing \( A_{ex} \) to lower the energy of forming domain walls to facilitate transitions to multidomain and skyrmion phases in ultrathin films while maintaining a reasonable \( K_{eff} \) and modest \( D \). Given that the domain wall width is estimated as \( \lambda = \sqrt{A_{ex}/K_{eff}} \), lowering \( A_{ex} \) while maintaining a significant \( K_{eff} \) to obtain skyrmion phases has the added benefit of maintaining a lower domain...
wall width than would be expected from lowering $K_{\text{eff}}$. We observe skyrmion phases at room temperature in several FM/HM heterostructures designed to have a low $A_{\text{ex}}$, including Pt/Co/Pt, Pt/Co/NiCu/Pt, Pt/Co/Ni/Pt, and Pt/Co/Ni/Re, with either one or two repeats of the unit structures listed. For the Pt/Co/Pt, Pt/Co/Ni/Pt, and Pt/Co/Ni/Re samples, the exchange stiffness is lowered through the use of ultrathin (<0.3 nm) Co layers.[27,28] Similar properties were obtained in a thicker Pt/Co/NiCu/Pt sample by increasing the proportionality of Cu to Ni in the alloy layer.[29,30] Concomitant to these changes in the static magnetic properties, we find that as the calculated domain wall energy density diminishes toward negative values, there is a dramatic transition in the domain morphology. We observe the stabilization of labyrinthine stripe domains at remanence and field-induced skyrmion phases within a narrow window of environmental conditions (i.e., temperature or applied magnetic field). By exploiting spin–orbit torques generated when passing an electrical current through the sample, skyrmion motion is observed. Using micromagnetic and analytic modeling, we demonstrate the strong relationship between the exchange stiffness and the equilibrium domain morphology in thin films with modest iDMI and significant PMA. Additionally, we show how the associated changes in the energy landscape allow for the stabilization of field-induced skyrmion phases within a framework of understanding like that of previous material approaches,[19–23] but through control of an unconventional parameter—the exchange stiffness.

2. Experimental Results

We begin by discussing experimental results for the [Pt/Co]$_2$ samples. Information regarding the composition of the samples discussed herein is provided in the Experimental Section. While the Pt/Co/Pt structure is nominally symmetric, previous work has shown that structural asymmetry between the Pt–Co and Co–Pt interfaces can lead to the development of PMA and a modest iDMI.[31,32] From measurements of the domain expansion velocity as a function of in-plane (IP) field for a [Pt/Co(0.35 nm)]$_2$ sample, we observe asymmetries that are typically associated with the “left-handed” iDMI thought to originate from the lower Pt–Co interface[32] and estimate a $|D|$ of $\approx 0.14$ mJ m$^{-2}$. The details regarding this measurement of $D$ from domain growth asymmetry measurements[24,33] can be found in Note S1 and Figure S1 in the Supporting Information. Given that it has been shown that $|D|$ scales with the exchange strength,[34,35] we take these results as an upper estimate for $|D|$ in samples with $t_{\text{Co}} < 0.35$ nm, as we will subsequently show that $A_{\text{ex}}$ roughly scales with $t_{\text{Co}}$ in the ultrathin limit.

In agreement with previous studies, the saturation magnetization as a function of temperature $M_s(T)$ plots provided in Figure 1a show a strong dependence on the Co thickness $t_{\text{Co}}$ and that the Curie temperature $T_C$ decreases as $t_{\text{Co}}$ is reduced into the ultrathin limit.[27,28] In particular, Figure 1a demonstrates that the sample with $t_{\text{Co}} = 0.32$ nm is thermally stable at room temperature, as $T_C$ is well above 300 K. However, decreasing $t_{\text{Co}}$ to 0.24 nm suppresses
the room temperature $M_s$ value and the $M_s(T)$ curve indicates a $T_C$ of $\approx 365$ K. To first-order, this reduction in $T_C$ is indicative of a reduced $A_{ex}$, particularly when $T$ is near $T_C$. However, it should be noted that the proportionality of the relationship between $T_C$ and $A_{ex}$ may be complicated in the ultrathin limit.[36] Further decreasing $t_{Co}$ to 0.2 nm suppresses $T_C$ to near room temperature. While it is anticipated that the uniformity of the Co layers may be compromised in the ultrathin limit (when the magnetic layer thickness approaches a single monolayer), previous reports have shown that well-defined FM behavior is observed even with submonolayer FM coverage, provided the FM layer is abutted by materials susceptible to proximity-induced magnetism (e.g., Pt, Pd).[27,28]

In Figure 1b, we show room temperature ($T = 296$ K) polar magneto-optic Kerr effect (MOKE) hysteresis loops for $[Pt/Co]$ samples with different $t_{Co}$. As expected for FM/HM heterostructures with PMA, we find that films with $t_{Co} \geq 0.26$ nm have square hysteresis loops with full remanence. However, reducing $t_{Co}$ to 0.25 or 0.24 nm, sheared polar MOKE loops are observed—suggesting a multidomain state at remanence in these samples. Further reducing $t_{Co}$ to a nominal thickness of 0.23 nm, the polar MOKE loop has zero remanent magnetization and a hard-axis-like character. As Figure 1a demonstrates that $T_C$ decreases dramatically in this regime of $t_{Co}$, this may potentially indicate a paramagnetic-like character to this sample or an in-plane magnetization. To understand how these changes in the static magnetic properties impact the room-temperature domain morphology, we have collected polar MOKE images at zero field and $T = 296$ K (Figure 1c–f). Before capturing an image, the following field history protocol was employed: First, the sample magnetization was saturated in the negative perpendicular field direction ($-M_z$). The magnetic field was then gradually increased toward each sample's respective positive coercive field ($+H_C$) before removing the magnetic field. For samples with $t_{Co} \geq 0.26$ nm, large circular magnetic domains were observed at remanence—as is expected for thin films with PMA. In agreement with the polar MOKE hysteresis loops, a labyrinthine stripe domain pattern with a sporadic density of circular domain textures was observed at remanence when $t_{Co}$ was reduced to 0.24 nm. Further reducing $t_{Co}$ to 0.23 nm, the featureless remanent state may indicate an in-plane magnetic anisotropy, a paramagnetic-like character to this sample, or that the domain morphology is smaller and/or fluctuating faster than our polar MOKE system can resolve. Overall, Figure 1c–f demonstrates that the domain morphology evolves dramatically with Co thickness in the limit of ultrathin Co.

In Figure 2a,b, we provide more detailed, temperature-dependent magnetometry data for the $[Pt/Co(0.24 nm)]_2$ sample. From out-of-plane (OOP) hysteresis loops collected in the temperature range of $294 K \leq T \leq 302 K$ (Figure 2a), the extreme temperature sensitivity of this sample’s magnetic properties can be seen. For $T \leq 292 K$, square hysteresis loops are observed, as is typical...
for thin films with PMA. As the temperature is increased over the range 294 K \(\leq T \leq 302\) K, the hysteresis loop becomes progressively more sheared—suggesting a multidomain remanent state. For \(T \geq 302\) K, the hysteresis loops take on a hard axis-like character (e.g., low remanence and low hysteresis). Thus, the evolution of the OOP hysteresis loops with increasing temperature is similar to the evolution with decreasing Co thickness shown in Figure 1b. The corresponding IP hysteresis loops, collected in the same temperature range over which the OOP loops exhibited such dramatic changes, are shown in Figure 2b. Surprisingly, we find that the IP saturation field is relatively large, giving an anisotropy field (estimated from the saturation field) of \(\mu_0 H_k = 500\) mT that does not appreciably change with temperature—suggesting that the temperature dependence of \(K_{\text{eff}}\) is scaling with \(M_s\). The magnetometry data collected at \(T = 302\) K shown in Figure 2a,b demonstrate that both the OOP and IP hysteresis loops exhibit zero remanence and little to no hysteresis. This evolution in magnetic response with temperature, including the formation of stripe domains, is similar in many respects to that observed during the SRT that occurs in the limit of ultrathin FM layers, when the intrinsic PMA equals the shape anisotropy (i.e., when \(K_{\text{eff}} = 0\)). However, above the SRT temperature, a finite remanence and a low saturation field are expected in the IP loops—behavior that is quite different from our observations. A further comparison of our results to the previously studied SRT is provided in Section 3.

In Figure 2c–f, we show polar MOKE images of the \([\text{Pt}/\text{Co}(0.24\ \text{nm})]_2\) sample as the perpendicular magnetic field strength \(\mu_0 H_z\) was quasi-statically decreased toward negative saturation (\(-M_r\)) at \(T = 296\) K. As mentioned previously, a labyrinthine domain pattern with a sporadic population of circular features is present in this sample when \(\mu_0 H_z = 0\). As in other systems whose static magnetic properties are extremely sensitive to temperature, we observe that the remanent domain pattern experiences a significant degree of thermal fluctuation when observed over several seconds, particularly for the \([\text{Pt}/\text{Co}]_2\) sample (see Video S1 in the Supporting Information). When a sufficiently strong \(\mu_0 H_z\) is applied, some stripe domains collapse into circular features with a characteristic diameter of \(\approx 3\) nm. Further increasing \(\mu_0 H_z\) causes all the stripe domains to collapse, forming a disordered ensemble of circular textures. A video of this evolution in domain morphology with \(\mu_0 H_z\) is provided in Video S2 in the Supporting Information. While this field-induced stripe-to-skyrmion transition can be observed at room temperature (\(T = 296\) K) in the \([\text{Pt}/\text{Co}(0.24\ \text{nm})]_2\) sample, we note that by heating the \([\text{Pt}/\text{Co}(0.26\ \text{nm})]_2\) sample (i.e., the sample whose room temperature domain morphology is shown in Figure 1c) slightly, similar behavior is observed (see Figure S2 in the Supporting Information).

To confirm that these field-induced circular features are magnetic skyrmions with chiral domain walls, the features’ response to applied electrical currents was determined. While we have not performed a precise quantification of the spin-Hall angle, Slonczewski-like torque, and field-like torque associated with the spin-Hall effect in our samples, previous reports have shown that Pt/FM/Ta-type structures exhibit a net positive spin-Hall angle, which dictates a positive polarization to the spin-current injected into the FM layers. As such, if our skyrmions possess a left-handed Néel chirality (as inferred from the

Figure 2. a–d) Polar MOKE images (collected at \(T = 296\) K) depicting the motion of several skyrmions in the \([\text{Pt}/\text{Co}(0.24\ \text{nm})]_2\) sample before and after a current density of \(j_e = 7.6 \times 10^9\) A m\(^{-2}\) was passed through the sample. Panels (a) and (b) correspond to +\(M_r\) domains (stabilized in \(\mu_0 H_z = -0.6\) mT), whereas panels (c) and (d) depict −\(M_r\) domains (stabilized in \(\mu_0 H_z = +0.6\) mT). e) Velocity of skyrmions along the x-axis (\(v_x\)) in the \([\text{Pt}/\text{Co}(0.24\ \text{nm})]_2\) sample as a function of \(j_e\). The velocities shown in panel (e) were calculated from wide-field polar MOKE images using techniques discussed in the Experimental Section. The error bars correspond to the standard deviation in velocity of all tracked skyrmions in motion over 60 s. The directionality of \(v_x\) and \(j_e\) are defined relative to the \(\hat{x}\) vector shown in panel (a). Data shown in Figure S1 in the Supporting Information, they should move in the same direction as the applied electrical current, regardless of the efficiency of the spin-Hall effect. While skyrmions are known to exhibit a deflection transverse to the applied current axis when driven—a phenomenon known as the skyrmion Hall effect—this behavior is muted in the limit of the low driving currents necessitated by the extreme sensitivity of our samples to temperature and magnetic fields.

Despite these limitations, we find that the circular features stabilized by \(\mu_0 H_z\) in the \([\text{Pt}/\text{Co}(0.24\ \text{nm})]_2\) sample move in the direction of the conventional current density \(J_z\) regardless of the domain polarity, as demonstrated in Figure 3 (Videos S3 and S4,
Considering these findings in the [Pt/Co] samples in the limit of ultrathin $t_{Co}$, we now examine how modifying the HM and FM layers to increase the structural inversion asymmetry (and hence, the total iDMI) impacts the morphological phases accessible in the ultrathin limit. We highlight two other material systems: [Pt/Co(0.24 nm)/Ni/Re]$_2$ and [Pt/Co(0.24 nm)/Ni]$_2$ (thicknesses in nm)—all featuring interfaces (Pt–Co, Ni–Re, and Ni–Pt) thought to contribute to an additive iDMI acting on the FM layers due to their compositional asymmetry (i.e., in addition to an asymmetry in interfacial quality, as in the [Pt/Co] samples).[49,50] While the Co layers of the two structures mentioned above were grown as wedges (using the approach detailed in the Experimental Section), we indicate the thickest $t_{Co}$ at which a $\mu_0H_z$-induced skyrmion phase was observed at $T = 296$ K. The remanent state of the [Pt/Co/Ni/Re]$_2$ sample shown in Figure 4a succinctly demonstrates the compositional sensitivity of the domain morphology in the ultrathin limit, as the stripe domain size dramatically decreases when $t_{Co}$ is varied by as little as $\approx 0.06\%$ (see Section 5 for information regarding the calculation of the thickness gradient).

We emphasize that while Figure 4a is reminiscent of previous studies that showed a strong link between film thickness and domain periodicity on account of proximity to a SRT,[15–18] the static characterization does not indicate the presence of the SRT in our FM/HM heterostructure samples near room temperature, given the sizeable in-plane saturation field is indicative of significant PMA.

From the higher-magnification polar MOKE images shown in Figure 4b,d, it can be seen that the [Pt/Co/Ni/Re]$_2$ and [Pt/Co/Ni]$_2$ samples exhibit a labyrinthine stripe pattern in zero field at room temperature ($T = 296$ K). In line with the [Pt/Co]$_2$ samples, temperature-dependent magnetometry of these samples indicate $T_C$ values below 400 K and OOP hysteresis loops extremely sensitive to variations in temperature in the vicinity of 300 K. Much like the [Pt/Co]$_2$ samples, the stripe domains formed at remanence in the [Pt/Co/Ni/Re]$_2$ and [Pt/Co/Ni]$_2$ samples transform into circular textures when a small perpendicular field is applied. By observing these features’ response to spin–orbit torques (using the process previously described for the [Pt/Co]$_2$ samples), it is possible to classify these features as skyrmions with fixed Néel chiralities. Figure 4c,e show polar MOKE images collected at the $\mu_0H_z$ value where each sample exhibits its densest skyrmion phase.

Thus far, the materials systems discussed have shared the commonalities of an ultrathin (<0.3 nm) Co layer producing a low $A_{ex}$ (inferred from a low $T_C$). However, if a low $A_{ex}$ is responsible for the observed domain morphologies, it should be possible to attain similar results in systems with somewhat thicker FM layers. Previous works have shown that alloying Ni with Cu can lower $T_C$ relative to that of elemental Ni and that this trend is linear with respect to Cu concentration.[29,31] Based on this, we fabricated a Ta(2)/Pt(5)/Co(0.4)/Ni$_{40}$Cu$_{60}$(0.9)/Ta(3) sample (thicknesses in nm) to explore the role of exchange stiffness by optimizing the Cu–Ni ratio. From magnetometry data collected near room temperature (Figure 5a), it can be seen that the [Pt/Co/NiCu]$_2$ sample’s OOP hysteresis loops are extremely sensitive to temperature; the square loop present at 285 K becomes more sheared as the temperature is increased to 290 K, and becomes hard axis-like when the temperature is
increased to 295 K. These findings are similar to those shown for the [Pt/Co(0.24 nm)]2 sample in Figure 2a.

Much like the [Pt/Co]2 samples with ultrathin Co layers, the IP hysteresis loops of the [Pt/Co/NiCu]1 sample do not change significantly in the vicinity of room temperature (Figure 5b); given that the sign of $K_{eff}$ does not change over the relevant temperature range, this does not indicate the presence of a SRT. The $M_s(T)$ curve shown in Figure 5c (generated from hysteresis loops collected over a wide range of temperatures) indicates a $T_C$ of ≈360 K for the [Pt/Co/NiCu]1 sample. From polar MOKE images collected at $T = 290$ K, it can be seen that the [Pt/Co/NiCu]1 sample exhibits a labyrinthine stripe domain pattern at remanence near room temperature, with a characteristic domain width of ≈2 µm (Figure 5d). When an ≈0.04 mT perpendicular field is applied, the labyrinthine remanent state is transformed into an ensemble of circular textures (Figure 5e) that display the spin–orbit torque induced dynamics emblematic of skyrmions as seen in the previously discussed samples with ultrathin Co layers—indicating that the energetic considerations that give rise to the remanent labyrinthine morphology and field-induced skyrmion phase are not contingent on a reduced dimensionality to the FM layers.

3. Discussion

Thus far, the temperature dependence of the OOP hysteresis loops, the suppressed $T_C$ values, and the labyrinthine domain morphologies observed at remanence in our samples draw strong parallels to extensive, past studies of the SRT.[15,16,18] Particularly, the thickness sensitivity exhibited in Figure 4a is heavily reminiscent of previous studies of a Cu/Fe/Ni structure that was found to exhibit a SRT when the Fe thickness was reduced to ≈1.25 monolayers.[18] While the original understanding of the SRT was developed using systems that have little or no iDMI, recent reports have shown that the iDMI can assist in raising the temperature at which the SRT occurs and facilitates a direct transition between the ferromagnetic PMA and paramagnetic states for sufficient $D$.[52] However, despite the many similarities between our HM/FM heterostructures and systems that undergo a SRT, our work is differentiated by the fact that $K_{eff}$ does not approach zero when the labyrinthine stripe phase becomes the remanent state in our samples (whether by adjusting the FM layer thickness or the sample temperature)—a key point that rules out a traditional SRT at the thicknesses and temperatures relevant to our samples. Given that the harbinger of a skyrmion phase in our samples has been the observation of a labyrinthine stripe phase at remanence, the treatment of the SRT, while related, may not fully be applicable.

To better understand the factors that give rise to skyrmion phases in thin films with significant PMA, modest iDMI, and low $A_{ex}$, we have performed micromagnetic and analytic modeling. For our models, a $M_s$ of 250 kA m$^{-1}$, a first-order uniaxial anisotropy constant $K_{1}$ of 675 kJ m$^{-3}$ (corresponding to

Figure 5. For the [Pt/Co/NiCu]1 sample: a) out-of-plane and b) in-plane magnetometry data collected in the temperature range of 285 K ≤ $T$ ≤ 295 K. c) Saturation magnetization $M_s$ over the temperature range 250 K ≤ $T$ ≤ 360 K. Polar MOKE images collected at $T = 290$ K in perpendicular magnetic fields of 0 and −0.04 mT are shown in panels (d) and (e), respectively.
A schematic depiction of the uniform (left) and two-domain (right) states used in the micromagnetic simulations. Blue (red) regions correspond to $+M_s$ ($-M_s$) magnetic orientations. b) Micromagnetic calculations of the difference in energy density $\Delta \varepsilon$ between the uniform and two-domain states (red) and the analytic domain wall energy density $\sigma$ (blue) as a function of iDMI energy density $D$. For each initial state, $\Delta \varepsilon/\sigma$ has been normalized to the value when $D = 0$. Data are shown for $A_{ex} = 2.5 \text{ pJ m}^{-1}$ (squares) and $A_{ex} = 10 \text{ pJ m}^{-1}$ (stars). c) Analytic modeling of the expected domain size as a function of the magnetic film thickness $t_{film}$ for selected $D$ values. The dashed line indicates the magnetic layer thickness of the [Pt/Co/NiCu]$_1$ sample used in our experiments. d) $\varepsilon$ as a function of $\mu_0 H_s$ for the labyrinthine stripe and skyrmion phases. The material parameters used in the modeling are stated in Section 3 of the main text.

$$K_{eff} = K_u - \frac{\mu_0 M_s^4}{2} = 28.25 \text{ kJ m}^{-3} \text{ in the thin film geometry},$$

and a film thickness $t_{film}$ of 1.5 nm were used to approximate the [Pt/Co/NiCu]$_1$ sample. While an $A_{ex}$ of $10 \text{ pJ m}^{-1}$ is often used when modeling Pt/Co and Pt/Co/Ni-based systems, this generally assumes that $T_0$ is closer to the bulk value of Co.[53–55] To reflect the fact that $T_0$ of the [Pt/Co/NiCu]$_1$ sample is $\approx 75\%$ lower than that of bulk Co, we use an $A_{ex}$ of $2.5 \text{ pJ m}^{-1}$ in our calculations. The iDMI energy density $D$ was varied between 0 and 1 mJ m$^{-2}$. All simulations were performed at $T = 0 \text{ K}$.

In the thin-film limit, it is well known that the relative balance between the demagnetization energy and the energy penalty associated with forming a domain wall (which depends on $M_s$, $K$, $A_{ex}$, and $D$) determines whether the film will break into domains in zero field and the corresponding domain periodicity.[56–59] To understand this energetic competition in the [Pt/Co/NiCu]$_1$ sample (which can generally be extended to describe the samples with ultrathin Co layers), we first performed a simple micromagnetic calculation of the difference in energy density $\Delta \varepsilon$ between the two-domain state with a single domain wall and the uniform magnetic state (i.e., $\Delta \varepsilon = \varepsilon_{\text{two-domain}} - \varepsilon_{\text{uniform}}$) as a function of $D$. This calculation of $\Delta \varepsilon$ allows for a micromagnetic estimation that can be compared to the analytical expression for the domain wall energy density $\sigma = 4\sqrt{A_{ex}K_{eff}} - \pi |D|$. A schematic depiction of the magnetic states considered is provided in Figure 6a. The $\varepsilon$ associated with each state was determined after allowing the domain state to relax from the respective initial state to the minimum energy configuration. From the $\Delta \varepsilon(D)$ plot provided in Figure 6b, it can be seen that the uniform state has a lower $\varepsilon$ at lower $D$—indicating that the energetic penalty associated with breaking into domains/forming a domain wall outweighs the associated reduction in dipolar energy.

As shown in Figure 6b, at a relatively modest value of $D$ ($=0.28 \text{ mJ m}^{-2}$), $\Delta \varepsilon(D)$ becomes negative, suggesting that the energetic penalty to forming a domain wall has been diminished and the energy landscape is more conducive to the formation of multidomain states. If instead, an $A_{ex}$ of 10 pJ m$^{-1}$ is used—as is often the case when describing Pt/Co-based systems—Figure 6b also shows that the root of $\Delta \varepsilon(D)$ is shifted toward $D$ values which are generally only feasible in material systems optimized to have a large iDMI or a reduced $K_{eff}$. Calculations of the domain wall energy density $\sigma = 4\sqrt{A_{ex}K_{eff}} - \pi |D|$ under the same considerations—also shown in Figure 6b—show similar trends as the $\Delta \varepsilon(D)$ simulations. We note that accounting for the domain wall anisotropy energy density[60] in the net $\sigma$ only slightly modifies the curve for lower $A_{ex}$ as shown in Figure 6b.
While these results are most applicable to the [Pt/Co/NiCu]_1 sample, we show how these trends in $\Delta \varepsilon$ and $\sigma$ evolve with $A_{ex}$, $K$, and $D$ in greater detail in Figure S3 in the Supporting Information. In keeping with our experimental results, as $D$ is not expected to change dramatically when varying the temperature by several $K$, we argue that the reduction in $\Delta \varepsilon$ and $\sigma$ with temperature in our samples is primarily determined by $K_{eff}$ (which itself is scaled by $M_s$, not $\mu_0H_K$) and the inverse scaling of $A_{ex}$ with temperature. Indeed, $M_s$ of the [Pt/Co/NiCu]_1 sample changes significantly for small modifications in temperature within the relevant temperature range (Figure 5c). Thus, while the shift in the relative energy of the uniform magnetic state versus a multidomain state is driven by $K_{eff}$, a low $A_{ex}$ and modest $D$ permit this transition without $K_{eff}$ approaching zero (i.e., away from the SRT).

To understand how these energetic balances impact the equilibrium domain size in the [Pt/Co/NiCu]_1 sample, we have employed the analytic model of Ref. [63]. In Figure 6c, we show the equilibrium domain width predicted as a function of magnetic film thickness $t_{film}$ for the [Pt/Co/NiCu]_1 sample (using the same static parameters as those used in the micromagnetic modeling) for several values of $D$. For the lower $D$ values (0 and 0.26 mJ m$^{-2}$), a sample with $t_{film} \approx 1.5$ nm is predicted to exhibit nm-scale domains, which may be considered as a uniform magnetic state. This calculation agrees with the micromagnetic modeling, which suggested that it was energetically disadvantageous for a sample with $t_{film} = 1.5$ nm to break into domains at lower $D$. For $D = 0.28$ mJ m$^{-2}$ (i.e., close to the $D$ value where $\Delta \varepsilon$ and $\sigma$ become negative), the equilibrium domain size decreases exponentially with the film thickness in the vicinity of the $t_{film}$ values relevant to the [Pt/Co/NiCu]_1 sample. Knowing that $t_{film} = 1.5$ nm and the experimentally determined domain size is $\approx 2 \mu m$, the analytic model indicates that 0.28 mJ m$^{-2}$ is a reasonable estimation for $D$ in the [Pt/Co/NiCu]_1 film.

Taken in concert, the micromagnetic and analytic modeling suggest that a modest $D$ can enable the formation of a labyrinthine stripe phase at remanence in samples with low $A_{ex}$—behavior that is not typical to thin films with low $M_s$ and appreciable PMA. However, the appearance of a labyrinthine stripe phase at zero field does not necessarily guarantee the presence of a field-induced skyrmion phase. To understand the nature of the field-induced morphological transition between stripe domains and skyrmions, we have performed additional micromagnetic simulations, comparing the $\mu_0H_z$-dependence of $\varepsilon$ between the labyrinthine stripe and skyrmion phases at $T = 0$ K. For these calculations, the material parameters were the same as those employed when generating Figure 6b, using $D = 0.28$ mJ m$^{-2}$. The initial state of the simulation was set to a random magnetization pattern or a skyrmion lattice (for the labyrinthine stripe and skyrmion phases, respectively). At each $\mu_0H_z$ step in the simulation, the system was relaxed to its minimum total energy configuration. In line with the experimental results, comparing the $\varepsilon(\mu_0H_z)$ profiles of these two morphological phases (Figure 6d) demonstrates that the skyrmion phase becomes the ground state when $\mu_0H_z = 0.09$ mT, which is reasonably close to the experimental findings. While an energetic barrier must be overcome to transition between morphological states, finite-temperature atomistic simulations have shown that bringing the system closer to $T_C$ can shallow this barrier. Given that all the samples that exhibit skyrmion phases discussed herein have a $T_C$ less than 400 K, we argue that the energy barrier separating morphological phases may be easily overcome by the thermal energy present near room temperature.

We have yet to comment in detail on the magnetic state present when the hysteresis loops indicate PMA (Figures 1b and 2a,b), but polar MOKE contrast has disappeared and there is little or no hysteresis (Figure 1b,f). In the simplest scenario, the domain size in this uniform-appearing phase may have become too small to resolve with polar MOKE. In the context of the SRT, however, such observations have been interpreted as an indicator of a phase transition, and there has been extensive debate in the literature as to whether this consists of a transition from the ferromagnetic state to a paramagnetic "gap" state with PMA,[18,66] or a fluctuating stripe domain phase.[67–69] While our samples do not have the low $K_{eff}$ emblematic of the SRT, the static magnetic properties are similar in many regards; thus, further investigations with higher spatiotemporal resolution are needed to fully understand the nature of the temperature/thickness-induced domain morphology transformations observed in our samples. Regardless of the end result of the transformation, the modeling discussed above permits an understanding of the energetic balances responsible for skyrmion stabilization in our samples with low exchange stiffness, appreciable PMA, and modest iDMI.

4. Conclusion

In summary, we have experimentally characterized the domain morphologies present in a number of thin-film heterostructures with perpendicular magnetic anisotropy, modest interfacial Dzyaloshinskii–Moriya interaction energy density, and low exchange stiffness. We find that by lowering the exchange stiffness, the remanent domain morphology transitions from a uniform state to a labyrinthine stripe phase—drawing strong, yet nuanced parallels to previous work on the spin reorientation transition that occurs in the limit of vanishing perpendicular magnetic anisotropy. Furthermore, when a small perpendicular magnetic field is applied at this morphological transition, skyrmion phases become stabilized. Spin–orbit torque-induced motion is observed when an electrical current is passed through the samples. Micromagnetic and analytic modeling demonstrates that in the limit of low exchange stiffness, the presence of a moderate interfacial Dzyaloshinskii–Moriya interaction modifies the energetic balance of thin films, allowing for the formation of multidomain remanent states and a shallowing of the energy barrier separating the stripe and skyrmion ground states.

5. Experimental Section

Sample Fabrication: Samples of the structure $[\text{Co} (t_{Co})/\text{Pt} (1)]_2$, $[\text{Co} (t_{Co})/\text{Ni} (0.3)/\text{Re} (0.5)/\text{Pt} (0.5)]_2$, $[\text{Co} (t_{Co})/\text{Ni} (0.3)/\text{Pt} (1)]_2$, and $[\text{Co} (0.4)/\text{Ni}_{0.2}\text{Cu}_{0.8}(0.9)]$, (thicknesses in nm) were used in this study. In the text, these samples were referred to as the $[\text{Pt/Co}]_2$, $[\text{Pt/Co/Ni}]_2$, and $[\text{Pt/Co/NiCu}]_2$ samples, respectively. A Ta (2)/Pt (5) seeding layer and Ta (3) capping layer were used for all samples. The samples were grown using dc magnetron sputtering at ambient
temperature onto Si substrates with a 300 nm thick thermal oxide coating. A 3 mTorr partial pressure of Ar and a sputtering power of 50 W were used. The samples were rotated at a frequency of ~5 Hz during deposition. It was noted that all thicknesses stated in the manuscript were determined by taking the product of the calculated deposition rate (determined from X-ray reflectivity measurements of reference samples) and the deposition time. Through computerized control of the shutter timing, average thicknesses can be controlled with high accuracy. For some samples, the Co layers were grown as wedge-type structures using a slit placed on top of the Co sputtering source—allowing for a 14% thickness gradient to be achieved during a single deposition over a 4 cm long substrate. By measuring the film thickness at several locations along a thicker reference sample, the percent change in thickness per unit length was determined and used to quantify the thickness gradient indicated in Figure 4a. Some samples were patterned into 100 µm wide wires using conventional metal lift-off UV photolithography.

**Magnetic Characterization and Imaging:** Temperature-dependent magnetometry was performed in the OOP and IP geometries using vibrating sample magnetometry (VSM). When calculating volumetric parameters from the magnetometry data, it was assumed that any Pt layers were thick enough to saturate the magnetic skyrmions. MOKE images and hysteresis loops were obtained using an Evico Magnetics microscope, in which perpendicular magnetic fields were applied using an air coil. For some MOKE measurements, the sample temperature was varied using a Peltier chip. The sample temperature during MOKE imaging was determined using IR thermometry. For the MOKE images presented within, images of the sample when the magnetization was saturated were subtracted as a background. The skyrmion velocity data shown in Figure 3e was calculated from 60 s long, 50 µm × 175 µm polar MOKE videos using the APREX TRACK software package. By defining contrast, size, and shape thresholds to describe the skyrmions, APREX TRACK can track the motion of many individual skyrmions at once.

**Micromagnetic Modeling:** Micromagnetic modeling was performed using the MuMax3 solver,[22] employing a 5 µm × 5 µm × 1.5 nm geometry discretized into 2 nm × 2 nm × 1.5 µm cells. Periodic boundary conditions were used to account for ten repetitions of the simulation geometry within the film plane.

**Supporting Information**
Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**
The authors declare no conflict of interest.

**Data Availability Statement**
The data that support the findings of this study are available from the corresponding author upon reasonable request.

**Keywords**
Dzyaloshinskii–Moriya interactions, magnetic skyrmions, magnetic thin films

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