Enhancement of zero-field skyrmion density in [Pt/Co/Fe/Ir]$_2$ multilayers at room temperature by the first-order reversal curve

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ABSTRACT
Magnetic skyrmions are novel topological spin textures on the nanoscale, and significant efforts have been taken to improve their zero-field density at room temperature (RT). In this work, we reported an approach of improving zero-field skyrmion density in [Pt/Co/Fe/Ir]$_2$ multilayers at RT by using the first-order reversal curve (FORC) technique to obtain information on the irreversible or reversible behaviors in the magnetization switching process. It was found from the FORC diagram that the magnetization reversal mechanism can be characterized into three stages: (1) reversible labyrinth stripe domains expanding or shrinking stage; (2) irreversible stripe domains fracturing stage; and (3) irreversible skyrmion annihilation stage. Furthermore, the zero-field skyrmion density can be highly improved by choosing reversal fields from the irreversible stripe domains fracturing stage. The highest skyrmion density was approached according to the maximum FORC distribution $\rho$. Our results have established the FORC measurement as a valuable tool for investigating magnetic multilayers of high skyrmion densities.

I. INTRODUCTION
Magnetic skyrmions are topologically protected quasi-particles with non-trivial spin texture which could serve as information carriers in novel spintronic devices, such as magnetic racetrack memory, logic gates, spin-torque oscillators, and artificial synapses/neurons.1–5 In recent years, the existence of skyrmions in magnetic/ heavy-metal multilayers with perpendicular magnetic anisotropy at room temperature (RT) has been realized resulting from the competition between Dzyaloshinskii–Moriya interaction (DMI), dipolar interaction, and exchange interaction.6–10 However, it is still challenging to approach high skyrmion densities without the assistance of an external magnetic field at RT. To overcome this obstacle, lots of work have been done.11–17 for instance, generating high density skyrmions from the labyrinth domains at zero-field in Pt/Co/Ta multilayers using an electromagnetic coordinated method14 and a scanning local magnetic field.15

The first-order reversal curve (FORC) technique has been proven to be an effective method in probing the magnetization reversal mechanism.18–27 This approach collects a larger dataset compared with the complete magnetization hysteresis (M-H) loop, from which information about the distribution of switching fields and interaction fields for all of the domains that contribute to the M-H loop and the irreversibility of magnetization switching can be derived, such as the magnetization switching process during complete reversal in Co/Pt multilayers.28 An open question is that whether the zero-field skyrmion density can be highly improved via analyzing the process of magnetization reversal or not. A thorough understanding of the magnetization switching processes involved in skyrmion-hosting materials must first be obtained.

In this work, we demonstrated a mechanism for improving zero-field skyrmion density in [Pt/Co/Fe/Ir]$_2$ multilayers at RT by using the FORC technique. The magnetization switching process
displays a distinctive characteristic of irreversible and reversible behaviors from the FORC diagram. By increasing the field from zero to saturation, a predominant reversible behavior is first observed due to the labyrinth stripe domains expanding or shrinking in width without changing their topology. As the field increases, the stripe domains irreversibly fracture into multiple skyrmions. Finally, the skyrmions annihilate irreversibly approaching saturation. It is found that the zero-field skyrmions density is higher by choosing a reversal field from the irreversible stage than the reversible stage. The highest skyrmion density was achieved from the reversal field corresponding to the maximum FORC distribution value. Our findings are of great significance for the successful application of skyrmions for practical applications.

II. EXPERIMENTAL METHOD

The multilayer film stack of Ta(5)/Ir(2)/(Pt(1)/Co(0.5)/Fe (0.4)/Ir(0.9))2/Ta(2) (the numbers in parentheses are the nominal layer thickness in nanometer) was deposited on a thermally oxidized silicon wafer using direct current magnetron sputtering techniques at RT. A base pressure of 1 × 10−7 Torr or better is reached before sputtering is initiated. Argon gas was used during the sputtering process at 2.0 mTorr for all materials except Co that was sputtered at 3.0 mTorr. The deposition rates for Ta, Pt, Co, Fe, and Ir were 0.62, 0.68, 0.26, 0.30, and 0.37 Å/s, respectively. The bottom Ta layer was used as an amorphous and adhesive underlayer. The additional Ir underlayer was added to provide a similar interface for the repeating layers. Additional capping layer of Ta was used as a protective layer against oxidation.

The out-of-plane \( M-H \) loop and FORCs of the \([Pt/Co/Fe/Ir]_2\) multilayers were measured by the MagVision Kerr microscopy system in the polar mode with magnetization imaged simultaneously. To remove the non-magnetic background signal, differential imaging was performed. Due to the longer measurement durations needed for FORC, piezo-motors were used to compensate for the sample drift.

III. RESULTS

A full \( M-H \) loop under an out-of-plane magnetic field \( H_z \) was first measured using the polar magneto-optical Kerr effect (MOKE) at RT as shown in Fig. 1. The red and blue solid lines represent the ascending and descending branches, respectively. The insets show the MOKE images that correspond to the out-of-plane magnetization configurations at selected fields as labeled by triangles along the ascending branch of the \( M-H \) loop. A circular region with a diameter of 26 μm was prepared from the measured Kerr images.

FIG. 1. Evolution of the out-of-plane magnetization configurations in \([Pt/Co/Fe/Ir]_2\) multilayers at RT. The red and blue solid lines represent the ascending and descending branches of the \( M-H \) loop, respectively. Insets are differential polar-MOKE images showing the field evolution of magnetic domains at selected fields as labeled by triangles along the ascending branch of the \( M-H \) loop. A circular region with a diameter of 26 μm was prepared from the measured Kerr images.

The estimated diameter of skyrmions in image 2 is about 400–500 nm. Second, these skyrmion-like domains expand in a non-symmetrical manner and become into short labyrinth stripes with increasing \( H_z \) (image 3). After that, the short stripes lengthen continuously in random directions (image 4) and reach an up/down magnetization balanced remanent configuration at zero field (image 5). In the un-polarized part, the magnetic domains change roughly in an inverse process of the aforementioned process. The long stripes first shorten with the field increases from zero and approach a complex configuration comprised of both stripes and skyrmions (image 6). Then, the stripes continuously shrink and also break into skyrmions simultaneously, and the changes become more obvious with increasing external field (images 7 and 8). Image 9 shows a pure skyrmion configuration with all the stripes disappear. Finally, the skyrmions gradually annihilated and a positively saturated state arrives when the field is larger than 15 Oe (image 10). To be noted is that there is hardly any skyrmions in the magnetization configuration at zero-field as shown in image 5 of Fig. 1. As shown in Fig. 1, the skyrmions in image 2 spontaneously nucleate, while the skyrmions in image 9 result from the breakup of the labyrinth domains. The different skyrmion formation mechanisms decide the skyrmion densities, and the skyrmion density in image 9 is much larger than that in image 2. An open question is that whether the zero-field skyrmion density can be enhanced if we reverse field from a non-saturation field to zero field. To verify this assumption, we adopt the FORC measurement to analyze the irreversible or reversible behaviors in the magnetization switching process.
To improve the skyrmion density at zero-field, the FORC technique was adopted to investigate the magnetization reversal characteristics of the [Pt/Co/Fe/Ir]$_2$ multilayers, in particular, the details of the reversible and irreversible behaviors of the magnetization switching process with the history of the field $H_z$. The FORC measurement can be carried out along both the ascending branch and the descending branch of the $M$-$H$ loop. In view of the symmetrical reversal processes along the two branches, in this work, we did the FORC measurement in the manner as defined in Fig. 2(a). First, a positive field of $H_{sat}$ large enough to saturate the sample, was applied, and then $H_z$ decreases to the selected fields of $H_R$ [red dots in Fig. 2(a)], from which the field increases to $H_{sat}$. These minor $H_{sat}$-$H_R$-$H_{sat}$ $M$-$H$ loops form the whole FORCs together. Figure 2(b) shows the measured FORCs with the reversal fields $H_R$ selected in a constant interval of 3 Oe, in which the outer boundary delineates the major $M$-$H$ loop. The insets show the zoomed-in views of two portions of the FORCs, which indicate the reversible and irreversible regions of magnetization. To better understand the minor $H_{sat}$-$H_R$-$H_{sat}$ $M$-$H$ loops, the FORCs are usually analyzed by a corresponding FORC diagram. The magnetization can be traced out from the FORCs of Fig. 2(b) as a two-variable function $M(H_R, H_z)$ in the $(H_R, H_z)$ plane as shown in Fig. 2(c). The FORC distribution function, a FORC diagram, can be calculated by

$$
\rho = \frac{1}{2} \frac{\partial^2 M(H_R, H_z)}{\partial H_R \partial H_z} \Delta H_R.
$$

Figure 2(d) is the contour map plotting of $\rho$. The FORC diagram reveals the occurrence of reversible and irreversible changes in the reversal process: $\rho = 0$ for the pure reversible changes, while $\rho \neq 0$ for irreversible changes. To understand the reversible and irreversible switching processes, we mainly focused on the third quadrant of the FORCs diagram since the selected reversal fields $H_R < 0$. According to the values of $\rho$ in Fig. 3(a), we divide the FORC diagram into three regions as marked by A, B, and C, respectively. Region A (0 Oe $\leq -H_R \leq 7$ Oe) is a nearly featureless region with the value of $\rho$ close to zero. Here, the value of $\rho$ is nearly zero and which implies a reversible magnetization change corresponding to the FORC as the top inset in Fig. 2(b). The magnetization reversal processes are reversible with the width of the stripe domains shrinking with field $H_z$ and the number of domains keeping unchanged. In contrast, in region B (7 Oe $\leq -H_R \leq 11$ Oe), the value of $\rho$ changes highly with the presence of a maximum at $(-H_R, -H_z) = (9.6$ Oe, $9.6$ Oe). The field-dependent variation of $\rho$ indicates the existence of an...
irreversible behavior during magnetization switching, which corresponds to a non-overlapped part of the FORCs as the bottom inset in Fig. 2(b). The irreversible process corresponds to the fission of stripe domains into skyrmions. Finally, in region C (11 Oe ≤ -HR ≤ 15 Oe), the HR close to the negative saturation field, a variation of ρ was observed with the presence of a negative valley and a positive peak at H = −10.0 and −8.5 Oe, respectively. The pair of negative–positive peaks indicates the annihilation of skyrmions.

We measured polar-MOKE images of the magnetic domain structures at three reversal fields HR = −4, −10, and −13 Oe as shown in the left column of Figs. 3(b)–3(d). The three HR were selected from the aforementioned three regions. The images of the magnetic domains at the corresponding remanent states by increasing the external fields from HR to zero are shown in the right column of Figs. 3(b)–3(d), respectively. It is found that the number of skyrmion at HR located in region B is much larger than that at HR in regions A and C. In the meanwhile, the number of skyrmion at the remanent state corresponding to HR located in region B is also higher than that corresponding to HR located in regions A and C.

The dependence of skyrmion densities at Hz = HR and Hz = 0 on HR is plotted in Fig. 3(e); the skyrmion density is defined as the counted skyrmion numbers divided by the image area. It is found that the zero-field skyrmion density first increases when the HR increases from region A to B and approaches a maximum value of 0.05 μm⁻² where the skyrmion density at the corresponding HR are also the largest. Then, the zero-field skyrmion density decreases with the HR increasing from region B to C. To further correlate the degree of irreversibility quantified by ρ from the FORC diagram with the enhancement of zero-field skyrmion density at remanent, we calculated the maximum ρ value ρmax for each HR as open squares in Fig. 3(e). The HR-dependent variation of ρmax exhibits a similar trend to that of skyrmion densities, i.e., first increasing to a maximum value in region B and then decreasing from there when the HR increases from region B to C. Furthermore, the maximum value of the ρmax and the zero-field skyrmion density appears at a very similar HR. Therefore, the improvement of zero-field skyrmion density can be directly correlated to the maximum ρ value in the FORC diagram. To maximize the zero-field skyrmion density, we can choose a reversal field HR corresponding to the maximum value of ρ in the Hsat–HR–0 process.

IV. CONCLUSION

In summary, our MOKE and FORC measurements reveal three distinct magnetization switching mechanisms in [Pt/Co/Fe/Ir]2 multilayers: (1) reversible stripe domains expanding and shrinking; (2) irreversible fission of stripe domains into skyrmions; and (3) irreversible skyrmions annihilation. We have found an effective method to improve the zero-field skyrmion density using the FORC technique: the skyrmion density at remnant can be highly enhanced by choosing the reversal fields corresponding to

![Figure 3](https://example.com/fig3.png)

**FIG. 3.** Magnetization reversibility and irreversibility characterized by domain topology modification. (a) FORC diagram divided into three regions according to the degree of reversible or irreversible behaviors. (b)–(d) Polar-MOKE images (the side length is 20 μm) of the magnetic domain structures measured at selected reversal fields H_R = -4, -10, and -13 Oe corresponding to aforementioned three regions [left column, open circles in (a)] and subsequently measured at the corresponding remanent states [right column, open squares in (a)]. (e) Skyrmion density measured at selected reversal fields H_R (green solid line with symbols of triangles) and subsequently measured at the remanent states when field changed from H_R to 0 (origin solid line with symbols of dots). The maximum values of the FORC distribution ρ measured at different reversal fields represented by the blue solid line with symbols of open squares.
high FORC distribution $\rho$ values. Therefore, the FORC technique is an effective method for skyrmionic application at zero field.

**AUTHORS’ CONTRIBUTION**

M.M. and C.C.I.A. contributed equally to this work.

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**DATA AVAILABILITY**

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

**REFERENCES**


