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Creation of a thermally assisted skyrmion lattice in Pt/Co/Ta multilayer films

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Néel-type magnetic skyrmions in multilayer films have recently attracted significant attention due to their stability at room temperature and low threshold for current-driven motion, offering the potential for the construction of high-speed and high-density spintronic devices. However, to date, research studies reported in the literature have rarely examined the effect of temperature on the formation and behavior of Néel-type skyrmions. Here, we investigate the effect of the temperature on the creation of a skyrmion lattice in [Pt/Co/Ta]₁₀ multilayer samples, using in-situ Lorentz transmission electron microscopy. By imaging the magnetization reversal process from a positive (negative) to a negative (positive) saturation, we find that the skyrmions can be created by nucleation from a ferromagnetic state and by breaking the labyrinth domains under certain external fields. More importantly, we demonstrate that the density of skyrmions in the multilayers not only depend on the external magnetic field, but also depend on the temperature and the thermal history of the materials. Published by AIP Publishing. https://doi.org/10.1063/1.5053983

Magnetic skyrmions are stable swirling spin textures that have recently attracted tremendous interest due to their topologically protected structure, small size, and unique current-induced dynamics.¹⁻⁵ These extraordinary properties have recently attracted tremendous interest due to their stability at relatively low temperatures.³,⁴,¹³⁻²⁴ To create skyrmions in bulk materials are of Bloch-type and can only survive at relatively low temperatures.³,⁴,¹³⁻²⁴ Most skyrmions in bulk materials are of Bloch-type and can only survive at relatively low temperatures.³,⁴,¹³⁻²⁴ To create skyrmions that are stable at room temperature and, therefore, suitable for industrial applications, various [heavy metal (I)/ferromagnet/heavy metal (II) (or oxide)]ₙ heterostructured multilayer films have been developed.²⁵–³³ The Dzyaloshinskii-Moriya interaction (DMI) originating from the strong spin-orbit coupling of interfacial atoms neighboring the magnetic layer plays a key role in the formation of the Néel-type skyrmions.²⁵⁻³³ Néel-type skyrmions have been observed in a wide range of thin-film stacks by the interplay of DMI, dipole interaction, perpendicular magnetic anisotropy (PMA), exchange interaction, and the external magnetic field.²⁵–³⁵ Their mechanism of formation and how to control the density of the skyrmions in such films have also been investigated.⁸,²⁹,³¹,³⁵ Increasing the density of the skyrmions can generally be achieved by decreasing the perpendicular anisotropy Kₐ by tuning the thickness of the ferromagnetic layer,³⁰,³³,³⁶ or by increasing the DMI with the proper choice of heavy metals.²⁷,³³ Research studies reported in the literature have also shown that, for multilayers, the electrical current,³⁷ in-plane magnetic field,³⁸ and the disorder³⁸ can also facilitate the creation of skyrmions.

We recently discovered that skyrmion lattices could also be directly written by the magnetic force microscopy (MFM) tips, at a zero magnetic field.³⁹

The columnar-shaped skyrmions created in thick multilayers have a much larger volume than those in a thin trilayer, which make them more stable and resistant to thermal fluctuations at high temperatures.⁴,⁵,²⁷,²⁹,³¹–³³,³⁹ However, it is still not well known whether skyrmions can survive at higher temperatures and how the temperature affects the creation of skyrmions. In this work, we investigate the magnetization reversal behaviors in [Pt/Co/Ta]₁₀ multilayer samples, at different temperatures, using in-situ Lorentz transmission electron microscopy (L-TEM). We find that the density of skyrmions increases substantially with increasing temperature. We also created high-density skyrmion lattices in samples with a strong PMA, by the assistance of high temperature, whereas no skyrmion lattice can be created at room temperature.

The multilayer stacks of Ta(5 nm)/[Pt(3 nm)/Co(2 nm)/Ta(2 nm)]₁₀ were deposited by using DC magnetron sputtering at room temperature. The basic properties of the multilayers were investigated by measuring hysteresis loops within a wide range of temperatures, from 5 K to 600 K, using a SQUID magnetometer with the magnetic field applied parallel and perpendicular to the film [see results in supplementary material, Figs. S1(a)–S1(c)]. Figure 1(a) shows the normalized out-of-plane and in-plane hysteresis loops measured at 300 K. At this temperature, the behavior of the loops clearly indicates that the sample is PMA. Reversal behaviors of the multilayer’s magnetization are subsequently studied at different temperatures, using in-situ L-TEM, with either a cooling in-situ holder or a heating one. Unless stated otherwise, all L-TEM images are taken at a...
defocus of 8.64 mm and a tilt angle of 30°. Figures 1(b)–1(f) show five representative L-TEM images taken while sweeping the magnetic field from the positive saturation field to the negative saturation, at 300 K. The succession of images in this figure clearly shows that, when the magnetic field is decreased from the positive saturation to 560 Oe [Fig. 1(b)], some isolated skyrmions, as well as snake-like strip domains, emerge from the ferromagnetic state. We call this process a “nucleating process.” As the field continues to decrease to 980 Oe, the density of skyrmions in a material increases when the magnetic field is further increased, the skyrmions get annihilated, gradually, and the sample finally reaches the saturated, ferromagnetic state in the opposite direction.

Previous studies found in the literature have shown that the density of skyrmions in a material increases when increasing the material parameter \( \kappa \), where \( \kappa = \pi D/4\sqrt{AK_{\text{eff}}} \), \( D \) is the DMI constant, \( A \) is the exchange stiffness, and \( K_{\text{eff}} = \mu_0H_sM_s = K_u = \mu_0M_s^2/2 \) is the effective perpendicular anisotropy. It is also well known that the temperature dependence of the parameters can be expressed as scaling relations: \( A(T)/A(0) = (M_s(T)/M_s(0))^\gamma \), \( D(T)/D(0) = (M_s(T)/M_s(0))^\beta \), \( K_{\text{eff}}(T)/K_{\text{eff}}(0) = (M_s(T)/M_s(0))^\lambda \).41–48 Figures 1(g) and 1(h) show the temperature dependence of the saturation magnetization \( M_s \) of the Co layers and the effective perpendicular anisotropy field \( H_k \) (or the in-plane saturation field). As expected, one can see in this figure that both \( M_s \) and \( H_k \) decrease monotonically, when increasing the temperature. By fitting the calculated \( K_u \), we obtain a \( \gamma \) value of approximately 2.4, in the case of our sample [see supplementary material, Fig. S1(d)]. We use \( \kappa \approx \beta \approx 1.5 \) (as calculated in Ref. 42) and find that \( \kappa \) increases with an increase in temperature.

To validate the above argument, i.e., that the skyrmion density increases with increasing temperature, we further investigate the magnetization reversal process, using L-TEM, for temperatures ranging from 97 K to 513 K. Figure 2(a) shows L-TEM images taken at 97 K, 453 K, and 513 K, when the density of the skyrmions reaches its maximum, in both the nucleating and breaking processes. It can be clearly seen, in Fig. 2, that the nucleation of skyrmions, from a ferromagnetic state, is very difficult to achieve at low temperatures, and only a few nucleated skyrmions can be observed at 97 K. However, many more skyrmions are being created in the breaking process, at a field of \( -980 \) Oe.38 Interestingly, when the temperature increases, we find that the density of skyrmions dramatically increases, in both nucleating and breaking processes. Figure 2(b), which shows the temperature dependence of the skyrmion maximum densities, strongly indicates that the thermal fluctuations play a critical role in the formation of skyrmions. Moreover, we find that the absolute values of the magnetic fields at which the maximum density of the skyrmions occurs decreases from 710 Oe to 530 Oe in the nucleating process and from 980 Oe to 580 Oe in the breaking process, as the temperature increases with an increase in temperature.
increases from 97 K to 513 K. In Fig. 2(e), we summarize the phase diagram of the temperature and the magnetic fields dependence of the magnetic domain configurations observed when a perpendicular field is reduced from 1000 Oe to −1500 Oe, for temperatures ranging from 97 K to 513 K. The yellow and red areas represent the states of the skyrmions in the nucleating process and the breaking process, respectively. The green area includes two states: one is the mixed state of skyrmions and snake-like domains, and the other is the labyrinth domain state. In Fig. 2(e), one can clearly see that the field required to stabilize skyrmions in both the nucleating and the breaking processes is dependent on the temperature, in a weak manner, for low temperatures ranging from 97 K to 253 K. At high temperatures from 300 K to 513 K, the field decreases significantly. Moreover, the range of magnetic field values for which skyrmions exist also increases with increasing temperature, particularly in the nucleating process.

To get a deeper, clearer understanding of the effect of temperature on the formation of the skyrmion lattice, we study the evolution of the magnetic structures with temperature, at a constant external field. To this end, a strong and positive magnetic field is first applied to saturate the sample, and then decreased to 580 Oe, at 300 K. L-TEM imaging shows a few isolated skyrmions and snake-like structures emerging from the ferromagnetic state [Fig. 3(a)]. By keeping other parameters unchanged, we increase the temperature to 513 K, gradually. The L-TEM images were then taken at different temperatures, during the temperature sweeping [Fig. 3(a)]. We find that the snake-like domains break into skyrmions, with increasing temperature, and that more and more new skyrmions nucleate gradually, from the ferromagnetic state, as shown in images taken at 443 K and 513 K. Interestingly, we find that the skyrmion lattice observed at 513 K remains unchanged, even after the temperature is being decreased, back to 300 K. To examine whether the properties of the sample have changed, after the heating process, we measured the reversal behavior again at 300 K. Observations reveal a very similar reversal behavior [Figs. 1(b)–1(f)]. These results enable us to confirm that a cycling temperature between 300 K and 513 K does not significantly alter the properties of the sample. We then repeat the experiment, for the same temperature cycle, but using a smaller external magnetic field. Our experimental procedure, in this case, involves applying first a strong and positive field to saturate the sample, and then decreasing the field to 520 Oe, at 300 K. L-TEM images taken during the thermal cycles, 300 K → 513 K → 300 K, are shown in Fig. 3(b). We find that, after the field has been decreased to 520 Oe at 300 K, an increasing number of snake-like structures appear in the sample, and that a high density skyrmion lattice state is obtained in the film at a temperature of 513 K. This density is much higher than that obtained at room temperature (Fig. 1). Another interesting result is the ability of the high-density skyrmion lattice to survive, even at a field of 520 Oe (at 300 K) after cooling down from 513 K. This field of 520 Oe is significantly smaller than the 600 Oe field required to stabilize skyrmions in the sample, at room temperature. We also find that a few skyrmions have a tendency to extend to a snake-like structure, as indicated by the red dotted ellipses [Fig. 3(b)]. This can be interpreted as follows: an isolated skyrmion will extend to a longer structure when the magnetic field is lower than the critical field; when a skyrmion is part of a lattice of with very high density, the repulsive forces from neighboring skyrmions will enhance its stability.37,39

After investigating the effect of the temperature on the skyrmion nucleating process, we then perform similar studies for the formation of skyrmions in the breaking process. To do so, we saturate the film again with a positive magnetic field, at 300 K. We then gradually change the external field to a negative field of −580 Oe, the point at which the long labyrinth domains start to break into small pieces, as shown in Fig. 3(c). When temperature is increased, all stripe domains transform to skyrmions, gradually, as shown in the image obtained at 408 K. A highly dense lattice of skyrmions forms at 513 K. Note that the density of the skyrmions at 513 K is much higher than that obtained in the nucleating process at the same temperature [Fig. 3(a)]. This is because the creation of skyrmions is relatively easy in the breaking process than in the nucleating process. Similarly, we find that all the skyrmions remain stable during the cooling phase, down to 300 K.

To investigate the effects of the temperature on the multilayer film of stronger PMA, we deposit a [Pt(3 nm)/Co(1.6 nm)/Ta(2 nm)]10 multilayer sample. The hysteresis loops were measured for a 5 K–850 K temperature range. Figure 4(a) shows the normalized hysteresis loops measured at room temperature (see more details for the M-H loops in supplementary material, Fig. S2). The $\gamma$ value obtained for this sample is ≈2.95; we also find that both $M_s$ and $H_k$ decrease with increasing temperature, as shown in Fig. 4(b). Based on the curves presented in Fig. 4(b), we expect the Curie temperature, for this sample, to be around 650 K. More importantly, the value of $H_k$ obtained for the [Pt(3 nm)/Co(1.6 nm)/Ta(2 nm)]10 multilayer reaches as high as 7.2 kOe, at room temperature, a value much higher than that observed in the [Pt(3 nm)/Co(2 nm)/Ta(2 nm)]10 multilayer [Fig. 1(b)]. Due to the strong perpendicular anisotropy, the width of the domain can be as large as 810 nm, at a zero external magnetic field, and no skyrmion can be created at room temperature, neither in the nucleation process nor in

![FIG. 3. In-situ L-TEM observation of the skyrmion creation process by increasing the temperature from 300 K to 513 K and go back to room temperature (300 K) with a constant external field of (a) −580 Oe, (b) −520 Oe, and (c) 580 Oe.](image-url)
the breaking process\textsuperscript{29,30,49} (see detailed reversal behaviors in supplementary material, Fig. S3). The next step of our investigations involve heating the sample and changing the magnetic field from a negative saturation field to a positive saturation field, during which we observe the reversal of the magnetization. Figure 4(c) shows L-TEM images of the sample at a zero external field (first row) and when the density of skyrmions reaches its maximum, at 323 K, 593 K, and 633 K, by tuning the applied external magnetic field in the breaking process (second row). The width of the domain, at a zero external magnetic field, decreases significantly when temperature is increased, as shown in the first row of Fig. 4(c). To quantify our results, we measured the widths of the domains at different temperatures and plotted the results as a function of temperature [Fig. 4(d)]. When increasing the temperature from 300 K to 633 K, the domain width decreases almost linearly, from approximately 810 nm to about 140 nm. When increasing the temperature further, the labyrinth domains gradually become thinner and eventually fade out at 673 K, as shown in the first row of Fig. 4(c). The evolution of images from 633 K to 673 K reflects the transition from a ferromagnetic state to a paramagnetic state, as the temperature approaches the Curie temperature. Since there is no spontaneous magnetization in a paramagnetic state where the magnetic spins point randomly in the space, the contracts induced by magnetic domains in the Lorentz-TEM image disappear. At temperatures lower than 553 K, no skyrmion is formed, at any magnetic field, as shown by the image taken at 323 K [Fig. 4(c)]. More details are provided in supplementary material, Fig. S3. We find that an increase in the temperature leads to a decrease in the width of the domain, and more and more skyrmions are formed gradually. Here, we mainly focus on the creation of the skyrmions in the breaking process. When the temperature is increased to 593 K, some isolated skyrmions appear under a 400 Oe magnetic field, as observed in the second row of Fig. 4(c). At 633 K, a lattice of skyrmions is eventually formed, with an applied external field of 410 Oe. In Fig. 4(d), we plot the densities of the skyrmions obtained at several elevated temperatures (the stars). Although neither a labyrinth domain nor any isolated skyrmion is observed for temperatures higher than 673 K (the Curie temperature), a high density of skyrmions gradually emerges when the sample is cooled down in a constant, a small magnetic field of 290 Oe (the field-cooling process) to 300 K, and the corresponding L-TEM images become clearer and clearer as the temperature decreases. The last image in the second row of Fig. 4(c) shows the lattice of skyrmions formed after a field-cooling process, from 673 K to 300 K, under a relatively small magnetic field of 290 Oe. We then perform more field-cooling experiments of the sample, from different initially high temperatures down to 300 K, with a magnetic field of 290 Oe. The values of the density of the skyrmions observed at 300 K, after the field-cooling process, are plotted in Fig. 4(d) (squares). In this figure, we observe that the density increases when we increase the initial high temperature of about 650 K up to 733 K, reaching saturation for values as high as $12 \mu m^2/C_0^2$. This suggests that the sample reaches a real paramagnetic state at 733 K. Based on the results reported here, we can confidently say that the high-density, uniformly distributed lattice of skyrmions can be created much more easily through the field cooling process, with a much smaller external magnetic field, particularly for high anisotropy materials.

To conclude, our investigations of the magnetization reversal behaviors in $[\text{Pt}/\text{Co}(1.6 \text{ nm})/\text{Ta}]_{10}$ and $[\text{Pt}/\text{Co}(2.0 \text{ nm})/\text{Ta}]_{10}$ multilayer samples, at different temperatures, show that
the thermal effect plays a critical role in the creation of skyrmions. We also reveal that a high density, uniformly distributed lattice of skyrmions is more easily created through field cooling processes. These results represent key findings for the practical applications of skyrmion lattices when working within a broad range of temperatures. It may even be possible to use local heating to facilitate the creation or annihilation of skyrmions in memory or logic devices and interesting applications that we may further investigate in the future.

See supplementary material for the hysteresis loops within a wide range of temperatures and the fitting of the parameter \( \gamma \) for the two samples and reversal behaviors of the \([\text{Pt}/\text{Co}(1.6 \text{ nm})/\text{Ta}]_{10}\) sample at room temperature.

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