Formation and magnetic-field stability of magnetic dipole skyrmions and bubbles in a ferrimagnet

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Abstract

We study the magnetic domain pattern in a ferrimagnetic CoTb thin film using Lorentz transmission electron microscopy. The evolution of domain patterns is captured under different temperatures and external fields. Magnetic dipole skyrmions and bubbles can be created simultaneously by increasing the temperature of the sample and applying appropriate out-of-plane magnetic fields. By tuning the external magnetic fields, it is found that the skyrmions are annihilated at a higher magnetic field compared to bubbles. Moreover, we also find that the in-plane magnetic field influences the formation and shape of skyrmions and bubbles. Further micromagnetic simulations are also consistent with the experimental observations. Our findings provide insights into the magnetic-field stability of skyrmions.

Magnetic skyrmions are topologically protected spin textures with fascinating properties, including topological protection, small size, and ease of movability under low-driving current density¹⁻³. They are thus considered as potential information carriers for building next-generation high-performance storage⁴, logic⁵, and nano-oscillator devices⁶. Skyrmions were first observed in bulk magnetic materials with broken inversion symmetry⁷⁻¹¹, where the bulk-type Dzyaloshinskii-Moriya interaction (DMI) favors a Bloch-type chiral spin texture. Soon after, skyrmions were also observed in magnetic heterostructures¹²⁻¹⁸, where they were stabilized by the interfacial-type DMI. Because of the mature technology of film fabrication and their compatibility with today's semiconductor manufacturing, the study on skyrmion in magnetic thin films receives tremendous attentions.

The spins of a skyrmion wrap around the unit sphere once and give rise to a unit winding number (topological charge)². Under the continuum limit of ferromagnetic moments, skyrmion's topology prevents its evolution into a topologically trivial state¹⁹⁻²¹, e.g. a ferromagnetic state. Unlike skyrmion, a magnetic bubble with a topological charge of zero, which is also a circular-shaped spin texture, is topologically distinct from a skyrmion. The magnetic bubble might be less stable than skyrmion since its transition to the ferromagnetic state is not topologically protected. Therefore, it is interesting to directly compare the magnetic field stability of skyrmions and bubbles within a magnetic system that they can coexist.

In this work, we study the dipole skyrmions with unit topological charge and bubbles with zero topological charge in a CoTb ferrimagnet. The reason for choosing

ferrimagnetic systems is its advantages of reduced skyrmion size²², suppressed skyrmion Hall effect²³⁻²⁵, and high-speed skyrmion motion²². The magnetic field dependence of the magnetic domain pattern is studied using a Lorentz transmission electron microscopy (L-TEM). It is found that both skyrmions and bubbles can be simultaneously stabilized by heating up the sample. Thus, the magnetic-field stability of skyrmions and bubbles can be directly compared by tuning the external magnetic field. Further simulations are also employed to understand the experimental results.

Co₉₀Tb₁₀ (CoTb) thin films with 25 nm thickness are co-sputtered on Si/SiO₂ wafers for morphologic and magnetic characterization and on Si₃N₄ substrates for magnetic domain structure study, respectively. The growth power for Co and Tb is 80 W and 20 W, respectively. The growth pressure is 2.6 mTorr. A 4 nm-thick Pt layer is subsequently grown to prevent the CoTb layer from oxidation. Figure 1(a) shows the layer structure of the sample. The high-resolution transmission electron microscope (HRTEM) image shows that the CoTb layer has an amorphous structure (see Fig. 1b), which is further verified by the fast Fourier transformation (FFT) pattern shown in the inset of Fig. 1(b). Figure 1(c) displays the elemental mapping conducted by scanning transmission electron microscopy (STEM) energy-dispersive X-ray (EDX) spectroscopy technique. The CoTb film exhibits a homogeneous distribution of Co and Tb elements.

Figure 1(d) shows the out-of-plane and in-plane hysteresis loop of the CoTb sample. Similar to the previous work^{25,26}, the sample exhibits a perpendicular magnetic anisotropy (PMA) at 300 K. The almost zero remnant magnetization is due to the

formation of multi-domain states at zero magnetic field, which we will discuss in detail later. Figure 1(e) shows the temperature dependence of saturated magnetization (M_s). Different from a conventional ferromagnet, M_s of our ferrimagnetic CoTb sample increases with increasing temperature within the measured temperature range. This behavior can be understood by considering the opposite magnetic moments of Co and Tb and their different temperature dependences. In the studied CoTb sample, the Co composition is dominant and the measured magnetization is the net magnetization of Co subtracting Tb. With increasing temperature, the magnetization of Tb decreases faster than that of Co, and thus the net magnetization increases with temperature^{26, 27}. As the composition ratio of Tb is very low, it cannot fully compensate the magnetization of Co even at very low temperatures, which explains why the compensation point is not observed in the whole temperature range.

We then study the magnetic domain patterns using a FEI Titan Cs Probe TEM in Lorentz mode (the Fresnel imaging mode) at 300 kV, and all the images are obtained at defocus of 600 μm . The out-of-plane magnetic field can be applied and tuned by varying the current passing through the microscope's objective lens. Figures 2(a)-2(c) show the magnetic domain of CoTb thin film for different out-of-plane magnetic fields at room temperature. At zero field, the film presents a typical labyrinthine domain pattern, which is consistent with the small remnant state observed in the hysteresis loop. With increasing the magnetic field, the field-favored domain grows in size and eventually evolves into a single domain. In the labyrinthine domain state, clear contrast variations from dark to bright or the reverse can be observed between the adjacent two domains. This is a result of the electron deflection in the film plane, which is caused by

the in-plane magnetization in the domain walls (DWs) due to Lorentz force. The distribution of the in-plane magnetic component can be analyzed using a phase reconstruction technique based on a transport of intensity equation (TIE)²⁸. Figure 2(d) shows the in-plane magnetic component for one typical area labeled in Fig. 2(a). It is clear to see the in-plane magnetic component of the DW that separates two perpendicularly magnetized domains. This DW has a Bloch-type structure – the typical feature of the perpendicularly magnetized thin film when the interfacial DMI is not considered²⁹. Although the interfacial DMI may originate from the CoTb/Pt interface and can facilitate a Néel-type DW²², its amplitude decreases with the magnetic layer thickness³⁰. Since the CoTb sample thickness is 25 nm, which is much thicker than that reported for inducing an observable effect²², we speculate that the interfacial DMI is not strong enough to form a Néel-type DW in the studied sample.

At room temperature, no skyrmion or bubble phase is observed in the sample during sweeping the out-of-plane magnetic field. Previous works have shown that the skyrmion phase can be obtained through tuning the PMA and it usually appears when the PMA is weak^{17, 31}. Following this guideline, we heat up the sample to tune the anisotropy of the sample. Figures 2(e)-2(f) show the magnetic field dependence of domain pattern at 373 K. The domain size at this temperature decreases compared to those at room temperature, which is likely related to the reduced PMA and increased thermal fluctuations²⁹. The domain size is further reduced when the temperature increases to 423 K, as shown in Figs. 2(g)-(i). We note that the sample no longer has PMA but has an in-plane anisotropy when the temperature is over 400 K (see

supplementary material). Different from the evolutions of the domain patterns at 300 K and 373 K, circular domains can be observed during the increase of the magnetic field. These circular domains are skyrmions and bubbles. We note here that the absence of strong DMI cannot prevent the formation of magnetic skyrmions. Appropriate magnetic properties can also lead to the dipole-field-stabilized skyrmion that has non-zero topological number^{32, 33}.

Typical spin structures of skyrmions (bottom) and bubbles (top) and the corresponding simulated L-TEM images are shown in Figs. 3(a) and 3(b). Due to the distinct spin textures, the L-TEM image of a skyrmion is either a bright or dark spot while the L-TEM image of a bubble possesses more complicated patterns with alternating dark and bright contrast. Another distinct difference is the shape. The skyrmion has a circle shape but the bubble has an elliptic shape. Both the skyrmions and bubbles can be observed in Fig. 2(i). When the external field increases to 1102 Oe at 423 K [Fig. 3(c)], the worm domains transform into bubbles and skyrmions. As the external field increases to 1502 Oe, all the bubbles disappear, only leaving the skyrmions [Fig. 3(d)], proving that skyrmion is more stable than bubbles. The shapes of the observed skyrmions and bubbles are highly consistent with the simulation results. Interestingly, it is noted that most of the long axes of bubbles are oriented in the same direction. This observation is related to the in-plane component of the magnetic field due to the misalignment of the magnetic field in the out-of-plane direction. As shown in Fig. 3(a), in order to maintain its topology, skyrmion has to form a swirl like spin texture. In contrast, such constraint is not applied to a bubble. The spin texture of a

bubble, especially the DW part, can be easily re-orientated according to the in-plane field direction in order to reduce the total energy. To verify this argument, we tilt the sample in two opposite directions and thus intentionally introduced two opposite in-plane magnetic field. In this case, the long axes of the bubbles are always aligned with the in-plane field axis. However, the contrast variation along the short axes is reversed ([see Figs. 3(e) and 3(f)], indicating the opposite in-plane magnetic component in the DWs of the bubbles.

To understand the bubble's shape and the stability of skyrmions and bubbles, we further perform micromagnetic simulations using Mumax3³⁴. The size of the simulated system is $1000 \times 1000 \times 25 \text{ nm}^3$ and the mesh size is $5 \times 5 \times 5 \text{ nm}^3$. The material parameters are listed as follows: saturation magnetization $M_s = 9.0 \times 10^2 \text{ emu/cc}$ and PMA constant $K_u = 4.1 \times 10^6 \text{ erg/cc}$ were obtained from the experimental measured hysteresis loops at 400 K (see supplementary material, Fig. S1). The temperature was set to 400 K and we assume exchange constant $A = 1.0 \times 10^{-6} \text{ erg/cm}$. The simulated both in-plane and out-of-plane hysteresis loops are very similar with the experimental results (see supplementary material). Figure 4(a) shows the evolution of the magnetic domain from labyrinthine domain at zero external field to skyrmions when increasing the out-of-plane magnetic field. It can be seen that when the out-of-plane field is 3.0 kOe, a mixed state of worm domains, skyrmions, and bubbles with random orientations are formed. When the field is increased to 3.3 kOe, all the worm domains transform into skyrmions and bubbles. The coexisting of skyrmion and bubbles similar to the experimental image [Fig. 3(c)] is observed. Further increasing the field to 4.0 kOe, all

the bubbles are annihilated and only the skyrmions survive, which agrees well with the experimental observations [Fig. 3(d)].

To further compare the magnetic-field stability of skyrmions and bubbles, we simulate the in-plane field-dependent energy of a skyrmion and a bubble as shown in Fig. 4(b). At zero in-plane field, we found that the skyrmion has a lower energy state than the bubble, revealing that the skyrmion is more stable than the bubble. When the in-plane field is larger than 310 Oe, the energy of skyrmion becomes larger than the bubble due to the large energy density at the left side where the magnetization direction is opposite with that of the in-plane field [see the middle figure at the right panel of Fig. 4(b)]. In this case, the bubble has lower energy since its shape is modified and reoriented towards the direction of the in-plane field. Further increasing the in-plane field to larger than 1.49 kOe, the skyrmion is no longer stable because the magnetization in the circular wall is forced to follow the field direction and hence the skyrmion transforms into a bubble. These simulations could explain our experimental observations that at zero tilt angle (0 in-plane components field), skyrmions are more stable, while under a tilt angle of 10° , where a small in-plane component of the field is induced, the magnetization direction at the boundary of bubbles is reoriented.

In conclusion, various magnetic domain states have been studied in an amorphous ferrimagnets CoTb alloy using L-TEM. The evolution of labyrinthine domains and the coexistence of magnetic skyrmions and bubbles have been observed by tuning temperature and out-of-plane external field. Further increasing the applied field in the coexisting phase, only skyrmions persist and all bubbles are annihilated, demonstrating

that skyrmions are more stable than bubbles under perpendicular field and tiny in-plane field, which is further corroborated by the micromagnetic simulations.

Supplementary Material

See the supplementary material for the characterization of magnetic properties at 400 K and corresponding micromagnetic simulations.

Acknowledgments

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Figure 1. Microstructure of the Si/SiO₂/CoTb(25)/Pt(4) sample. (a) The side-view of the TEM results. (b) HRTEM images of the CoTb thin film. The inset is the fast Fourier transformation of HRTEM. (c) STEM-EDX elemental mapping of the Si/SiO₂/CoTb(25)/Pt(4) sample showing clearly the homogeneous distribution and the sharp interface. (d) Out-of-plane and in-plane hysteresis loops of CoTb thin film. (e) Temperature dependence of saturated magnetization for CoTb thin film.

Figure 2. L-TEM images show the changes in the magnetic domain structure produced by thermal modulation and out-of-plane magnetic fields. (a)-(c) Evolution of domain patterns in the field of 0 Oe, 222 Oe, and 910 Oe at 300 K. (d) The zoom-in of the corresponding in-plane magnetic component in the red box in (a) which is obtained by TIE analysis of the L-TEM data. The colors and arrows show the direction and the magnitude of the in-plane component. The inset shows the color wheel. (e)-(f) Domain patterns captured at 500 Oe, and 1000 Oe when increasing the magnetic field at 373 K. (g)-(i) Domain patterns captured at 0 Oe, 566 Oe, and 700 Oe when increasing the magnetic field at 423 K.

Figure 3. Coexistence of magnetic skyrmions and bubbles in a high magnetic field. (a) The illustration of bubbles and skyrmions with opposite spin chirality corresponding to simulated L-TEM pattern in (b). (c) Coexistence of magnetic skyrmions and bubbles in 1102 Oe field. (d) Disappearance of bubbles in 1502 Oe field with only skyrmions. (e)-(f) The domain pattern for the tilt angles are 10° (e) and -10° (f), respectively. The contrast variation across the short axis is reversed, indicating the reversed in-plane magnetization in the DW, as shown in the (a) and (b). (g)-(h) The indications of the sample rotations correspond to the images in (e) and (f), respectively.

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Figure 4. Micromagnetic simulation results for the creation of skyrmions and magnetic bubbles and their energy. (a) The evolution of domain patterns with increasing out-of-plane field. Skyrmions and bubbles coexist in the field of 3.3 kOe, and in the 4.0 kOe only skyrmions exist. The dashed circles label the bubbles. (b) The energy difference of a skyrmion and a bubble with the increase of the in-plane field. At the lower in-plane field, skyrmion has lower energy than bubbles which means skyrmion is more stable. Right panel shows the corresponding energy density of the spin texture.

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