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Room-temperature creation and manipulation of skyrmions in MgO/FeNiB/Mo multilayers

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Magnetic skyrmions in multilayer structures are considered as a new direction for the next generation of storage due to their small size, strong anti-interference ability, high current-driven mobility, and compatibility with existing spintronic technology. In this work, we present a tunable room temperature skyrmion platform based on multilayer stacks of MgO/FeNiB/Mo. We systematically studied the creation of magnetic skyrmions in MgO/FeNiB/Mo multilayer structures with perpendicular magnetic anisotropy (PMA). In these structures, the magnetic anisotropy changes from PMA to in-plane magnetic anisotropy (IMA) as the thickness of FeNiB layer increases. By adjusting the applied magnetic field and electric current, stable and high-density skyrmions can be obtained in the material system. The discovery of this material broadens the exploration of new materials for skyrmion and promotes the development of spintronic devices based on skyrmions.

Keywords: magnetic skyrmion, MgO/FeNiB/Mo multilayers, electromagnetic coordinated manipulation, Lorentz transmission electron microscopy (LTEM)

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1. Introduction

In recent years, with the rapid development of emerging fields such as big data, artificial intelligence, and the Internet of Things, people have put forward higher and higher requirements for information storage technology. Researching new types of random access memory with high density, high speed, non-volatile, and low energy consumption is currently the most urgent and challenging task. In magnetic materials with broken structure-inversion symmetry, there will be magnetic skyrmions with several compelling attributes as prototype memory elements, namely their: nontrivial spin topology, small size and solitonic nature.[1] These characteristics make it possible to meet the above requirements and break through challenges such as Moore's law and superparamagnetic limit, and have great potential in future applications of electronic devices such as storage and logic.[2]

In order to realize the practical application of memory devices by using magnetic skyrmions, the key is to obtain magnetic skyrmions which are stable at room temperature and easy to be regulated. It is generally believed that skyrmions exist in non-centrosymmetric systems with chiral interactions[3–6] or multilayer film systems with interface symmetry breaking.[7–10] Among them, the magnetic skyrmions observed in the latter has many advantages, such as its stable existence near room temperature, easy adjustment of experimental parameters, good compatibility with devices, and the ability to achieve current regulation behavior, which makes it beneficial for practical applications.[1,11–17] People have conducted extensive researches in this area and found that stable skyrmion phase can be created by engineering the interfacial perpendicular magnetic anisotropy (PMA) of the multilayer film systems.[9,10,18] Our recent study shows that MgO/FeNiB/Mo-based heterostructures exhibit large PMA, which can be regulated by the thickness of the FeNiB layer.[19] This indicates that it is possible to find skyrmion in MgO/FeNiB/Mo system, which will undoubtedly add bricks to the skyrmion family.

In this work, a series of MgO/FeNiB/Mo multilayer structures were prepared by magnetron sputtering. When the thickness of FeNiB layer is 1.5 nm, PMA of the structure is moderate, and skyrmions can be observed under the effect of an external magnetic field. The creation of skyrmion in the structure was studied by Lorentz transmission electron microscope (L-TEM), and it was manipulated by adjusting the applied

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magnetic field and electric field. The discovery of this material broadens the exploration of new materials for skyrmion and promotes the development of spintronic devices based on skyrmion.

2. Experimental methods

The multilayer structures of substrate/MgO(2.5)/[MgO (2.5)/FeNiB(t_{FeNiB})/Mo(3.0)]$_{\times 7}$/Mo(5.0) (where "×7" denotes the layer repetition numbers, thickness in nm and FeNiB-layer thickness t_{FeNiB} varying from 1.4 nm to 1.7 nm) were fabricated using magnetron sputtering with a background pressure of $\sim 4\times10^{-8}$ Torr (1 Torr = 1.33322×10^{2} Pa). The structure is schematically shown in Fig. 1(a). The FeNiB films were sputtered from the composite target by symmetrically placing boron patches on $Fe_{85}Ni_{15}$ alloy targets. The film compositions are $Fe_{68}Ni_{14}B_{18}$ in atomic percent, determined by coupled plasmas atomic emission spectrometry (ICP-AES). The MgO layer were deposited by radio frequency sputtering, where the Mo layer was deposited by direct current magnetron sputtering. A 5-nm Mo cap was deposited to prevent the oxidation of the stack. The deposition pressure was kept at 3.5 mTorr for all the layers. Films were deposited on two types of substrates: 10-nm-thick Si_3N_4 membrane windows (for direct TEM observation) and simultaneously on thermally oxidized Si wafers with standard Hall bar shape (for magnetic measurements). Each Hall bar includes a 4.8 mm×0.5 mm channel and a perpendicularly placed 1.5 mm×0.3 mm channel. All the films were annealed at 400 °C for 30 min in a high vacuum furnace (3×10^{-7} Torr) without external magnetic field to enhance their PMA. The magnetic structures were observed by Lorentz transmission electron microscope (L-TEM: JEOL 2100F) with perpendicular magnetic field, which was introduced by increasing the objective lens current. The skyrmion manipulation behavior by electric current was conducted using a double-tilt electrical TEM holder with two electrical conducting blocks at two sides of the film. A current pulse with 150-μs pulse width was supplied by a source–measure unit instrument (Keithley 6221). The magnetic hysteresis (M–H) loops of the same Hall bar were measured using vibrating sample magnetometry (VSM). All the measurements were done at room temperature.

3. Results and discussion

In multilayer structures with PMA, Néel-type magnetic skyrmion is mainly involved due to the competition of the exchange interaction, the dipolar interaction, the Dzyaloshinskii–Moriya interaction (DMI) and the anisotropy.[12,20–23] There is experimental evidence that medium strength PMA is more likely to produce magnetic skyrmion than strong PMA.[11] For the convenience of discussion, the quality factor Q ($Q = K_{\mathrm{u}}/K_{\mathrm{d}}$) is used to identify different magnetic anisotropy,[9,24] where K_{u} is the dipole demagnetization energy, also known as the uniaxial anisotropy constant, and K_{d} ($K_{\mathrm{d}} = 2\pi M_{\mathrm{s}}^2$, M_{s} is the saturation magnetization) is the shape anisotropy constant. Therefore, materials with $Q > 1$ and $Q < 1$ correspond to PMA and IMA, respectively. In this work, we will pay comprehensive attention to the magnetic anisotropy of MgO/FeNiB/Mo system and its relationship with skyrmion.

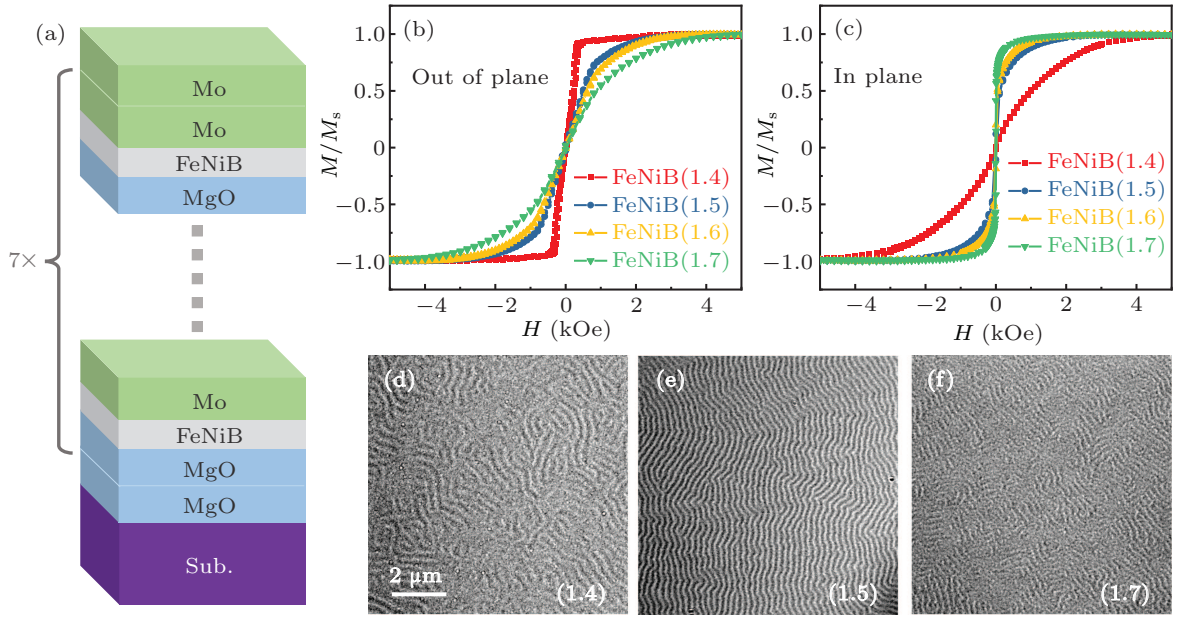


Fig. 1. The hysteresis loops and magnetic domain of the stacks of MgO(3)/FeNiB(t)/Mo(3) in different thicknesses of FeNiB. (a) Schematic multilayers made of seven repetitions of MgO/FeNiB/Mo trilayer. (b)–(c) The normalized out-of-plane and in-plane hysteresis loops of the stacks of MgO(3)/FeNiB(t)/Mo(3). (d)–(f) The magnetic domain of the samples while FeNiB thickness was 1.4 nm, 1.5 nm, and 1.7 nm, where the external magnetic field was absent. The scale bar is 2 μm.

By changing the thickness of FeNiB layer, PMA can be controlled in multilayer films. The normalized out-of-plane and in-plane hysteresis loops together with the corresponding magnetic domain morphology of the multilayers at room temperature are shown in Fig. 1. As shown in Figs. 1(b) and 1(c), as the thickness of FeNiB layer increases, the easy magnetization axis of the sample gradually rotates from out-of-plane to in-plane. For multilayers with thinner FeNiB layer thickness $t_{\mathrm{FeNiB}} = 1.4$ nm, significant PMA is observed from the magnetic hysteresis loops, with an effective perpendicular anisotropy field of about 4000 Oe (1 Oe = 79.5775 A·m^{-1}), and the quality factor $Q > 1$. When t_{FeNiB} increases from 1.4 nm to 1.5 nm, the perpendicular magnetic anisotropy energy decreases from 1.14×10^5 J/m^3 to -4×10^4 J/m^3, and the corresponding quality factor $Q \approx 1$. The saturated magnetization of FeNiB is around 1.16×10^6 A/m for all the samples, which was measured by VSM. As t_{FeNiB} continues to increase, the value of quality factor becomes less than 1.

Besides, in the absence of external magnetic field, the magnetic domain structure of the multilayer films grown on the Si_3N_4 membrane windows was observed by L-TEM [Figs. 1(d)–1(f)]. The sample behaves like a labyrinth domain when $t_{\mathrm{FeNiB}} = 1.4$ nm [Fig. 1(d)]. When t_{FeNiB} increases to 1.5 nm, it appears as a stripe domain and its density is smaller than that of 1.4 nm [Fig. 1(e)]. When $t_{\mathrm{FeNiB}} = 1.7$ nm, most of the magnetic domains are in-plane domains [Fig. 1(f)]. Then, a perpendicular magnetic field was applied *in situ* by L-TEM to observe the relationship between the domain structure and the external magnetic field. It is found that skyrmion can be observed in the stripe domain samples of $t_{\mathrm{FeNiB}} = 1.5$ nm under suitable external magnetic field. However, no matter how the external magnetic field is changed, skyrmion is unlikely to be observed in other samples ($t_{\mathrm{FeNiB}} = 1.4$ nm and $t_{\mathrm{FeNiB}} = 1.7$ nm). Therefore, the sample with $t_{\mathrm{FeNiB}} = 1.5$ nm was mainly studied.

Figure 2 depicts the magnetic domain patterns of MgO(2.5)/[MgO(2.5)/FeNiB(1.5)/Mo(3.0)]$_{\times7}$/Mo(5.0) multilayer films under different perpendicular external magnetic fields. The images of magnetic domain patterns were obtained using L-TEM. In the absence of an external magnetic field, the stripe domain along z direction is very stable [Fig. 2(a)]. With the application of perpendicular magnetic field, the density of stripe domain decreases [Fig. 2(b)]. When the perpendicular magnetic field increases to around 550 Oe, the stripe domains disappear and skyrmion with a size of about 200 nm is created [Fig. 2(c)]. When the perpendicular magnetic field continues to increase, the density of skyrmion decreases gradually (660 Oe) [Fig. 2(d)], and finally drops to 0, showing a single domain state (720 Oe) [Fig. 2(e)]. Subsequently, with the decrease of the perpendicular magnetic field (650 Oe), skyrmion reappears [Fig. 2(f)]. When the perpendicular magnetic field reduces to around 580 Oe, the density of skyrmion reaches the highest again [Fig. 2(g)]. As the perpendicular magnetic field continues to decrease, the skyrmion disappears gradually (390 Oe) [Fig. 2(h)] and the initial stripe domain reappears (0 Oe) [Fig. 2(i)]. Obviously, skyrmions in multilayer films can be reversibly created through the reciprocating changes of the external perpendicular magnetic field.

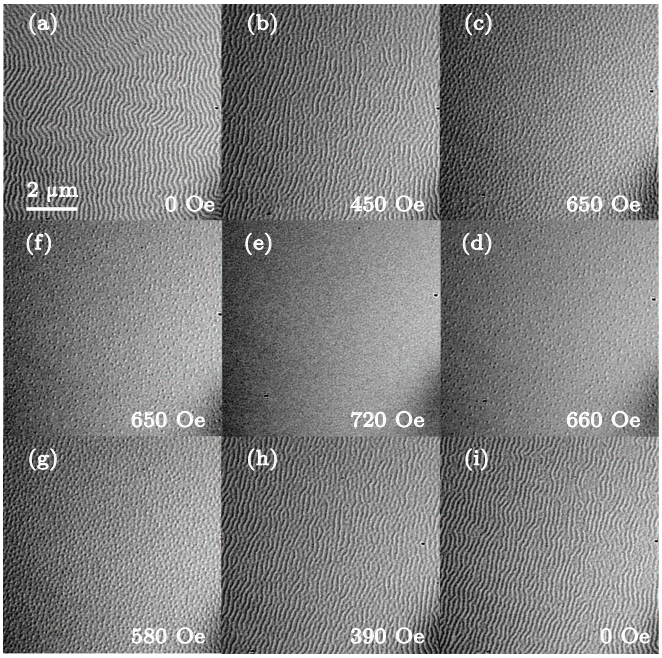


Fig. 2. Magnetic domain evolution of MgO(2.5)/[MgO(2.5)/FeNiB(1.5)/Mo(3.0)]$_{\times7}$/Mo(5.0) multilayer films under different perpendicular magnetic fields. 0 Oe (a), 450 Oe (b), 650 Oe (c), 660 Oe (d), 720 Oe (e), 650 Oe (f), 580 Oe (g), 390 Oe (h), and 0 Oe (i). The scale bar is 2 μm.

So far, it is certain that high-density skyrmion can be obtained in MgO/FeNiB/Mo multilayer structures. However, in practical applications, the manipulation of skyrmion is a necessary condition for achieving its practical use. As we all know, the application of magnetic field in information storage devices is a serious obstacle to the development of high-density and low-energy devices, and the realization of electrical control of skyrmion is the most critical step in the future application of skyrmion. Therefore, in this work, the effect of electric current on skyrmion is further studied. And the schematic geometry of the electric current application is shown in Fig. 3(a).

Firstly, the electric current is applied to the sample in the absence of magnetic field, and it was found that even if a large current was applied, the creation and manipulation of skyrmion cannot be achieved while ensuring the integrity of the device. Therefore, for MgO(2.5)/[MgO(2.5)/FeNiB(1.5)/Mo(3.0)]$_{\times7}$/Mo(5.0) multilayer films, in order to achieve electrical control, the application of a certain size of perpendicular magnetic field is necessary. When a perpendicular magnetic field of 450 Oe is applied, the magnetic domain structure of the sample remains striped [Fig. 3(b)]. When a current of 1.35×10^8 A/m^2 is applied to the sample, the stripe domains pinch off into slyrmions and generate the mixed skyrmion and stripe phase [Fig. 3(c)].

This is probably due to the introduction of current in the device. On the one hand, torque will be generated due to the spin Hall effect after the introduction of current in heavy metals, which will further affect the magnetic domain structure of the sample.[10,16] The inevitable defects in polycrystalline multilayer films help to pin the main body of the stripe domains, while the current applies spin Hall torque at the head and tail of the stripe domains.[10] On the other hand, the Joule thermal effect will be introduced to increase the temperature of the sample, which will reduce the magnetic anisotropy to a certain extent. As the current continues to increase, the number of skyrmion further increases, and the stripe domains gradually disappear [Figs. 3(d)–3(e)]. The skyrmion density is remarkably enhanced by this electromagnetic control compared with the skyrmions induced only by the magnetic field as shown in Fig. 2(c). It is worth mentioning that the current density to produce the high-density skyrmons are quite smaller than those in other multilayers.[10,25] However, when the current increases to 1.77×10^8 A/m^2, the skyrmion density will actually decrease [Fig. 3(f)]. The reason may be that the heat generated by the high current prevents skyrmion from being stable, while this does not cause damage to the device. When the current is reduced to 0, the initial stripe domains reappear [Fig. 3(g)]. Therefore, it can be concluded that the magnetic properties of MgO(2.5)/[MgO(2.5)/FeNiB(1.5)/Mo(3.0)]$_{\times7}$/Mo(5.0) multilayer films are highly tolerant to electric current. Meanwhile, the associated domain structures, including skyrmion, are not sensitive to current processes.

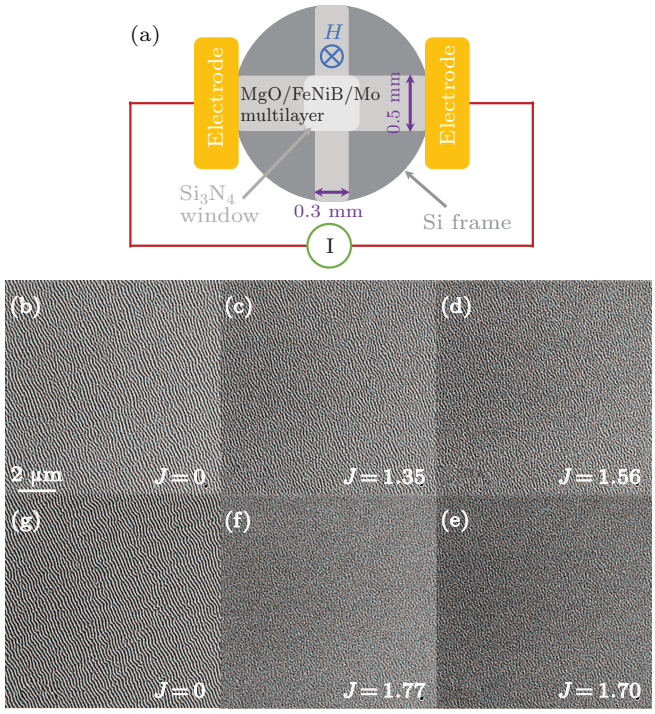


Fig. 3. The electric current manipulations of the MgO(2.5)/[MgO(2.5)/FeNiB(1.5)/Mo(3.0)]$_{\times7}$/Mo(5.0) multilayer films via applying a fixed magnetic field (450 Oe) at different currents. (a) The schematic geometry of the electric current application for 0 (b), 1.35 (c), 1.56 (d), 1.70 (e), 1.77 (f), and 0 (g) (in units of 10^8 A/m^2). The scale bar is 2 μm.

Subsequently, the electric current dependence of the skyrmion distribution under different fixed magnetic fields is systematically analyzed by Lorentz TEM to better understand the electromagnetic manipulation. In the case of a given current (0.64×10^8 A/m^2), the external perpendicular magnetic field is changed to observe the changes in magnetic domains in the sample, as shown in Fig. 4. In the absence of an external magnetic field, there are only stripe domains under the action of a simple current [Fig. 4(a)]. When the external perpendicular magnetic field increased to 450 Oe [the amplitude of the magnetic field is consistent with Fig. 2(b)], the number of stripe domains decreases and some skyrmions begin to occur. When the perpendicular magnetic field increases to around 550 Oe, the skyrmion density reaches the maximum, which is consistent with the situation where only the external magnetic field is applied without adding current. The magnetic field to form complete skyrmions is significantly reduced at a fixed current dentsity [Figs. 4(c) and 2(c)]. However, when the perpendicular magnetic field continues to increase (650 Oe), the skyrmion completely disappears [Fig. 4(d)]. This is different from the previous case of simply applying an external magnetic field (the skyrmion will not completely disappear until the external magnetic field increases to 720 Oe). The possible reason is that the application of the electric current changes the perpendicular anisotropy of the multilayer films, which in turn reduces the size of the external field that enables the steady state formation of the skyrmion in the sample. Subsequently, with the decrease of the perpendicular magnetic field (520 Oe), the high-density skyrmion reappears [Fig. 4(e)]. When the external magnetic field is removed, the magnetic domain returns to the original stripe domain, which once again shows that the above process is stable and repeatable.

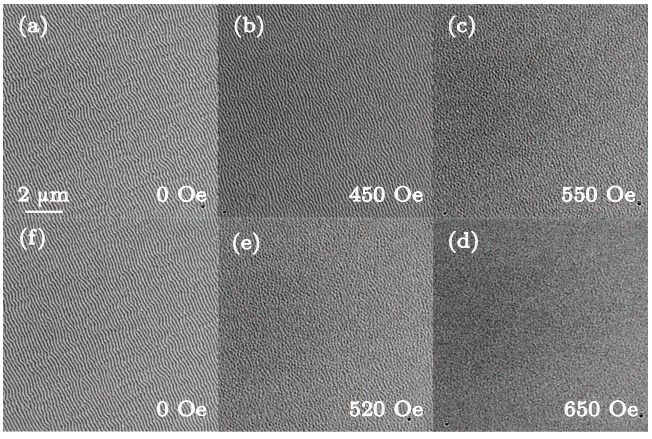


Fig. 4. The magnetic field manipulations of the MgO(2.5)/[MgO(2.5)/FeNiB(1.5)/Mo(3.0)]$_{\times7}$/Mo(5.0) multilayer films via applying a fixed electric current (0.64×10^8 A/m^2) at different perpendicular magnetic fields: 0 (a), 450 (b), 550 (c), 650 (d), 520 (e), and 0 (Oe) (f). The scale bar is 2 μm.

4. Conclusion and perspectives

In conclusion, in MgO(2.5)/[MgO(2.5)/FeNiB($t_{\rm FeNiB}$)/Mo(3.0)]$_{\times7}$/Mo(5.0) multilayer structures, after high-temperature vacuum annealing at 400 °C, the effective per-

pendicular anisotropy of the sample approaches zero when FeNiB thickness is about 1.5 nm. By adjusting the amplitude of the external perpendicular magnetic field, high-density skyrmions can be created, and when the external magnetic field is removed, it can return to the initial steady state, which proves that the regulation is reversible. Besides, it is found that the electric current can effectively reduce the magnetic field required for skyrmion formation in the system to some extent. The above results show that MgO/FeNiB/Mo-based heterostructures may provide a new material system for novel skyrmion-based spintronic devices.

Acknowledgements

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