Controlled Switching of the Number of Skyrmions in a Magnetic Nanodot by Electric Fields

Zhipeng Hou,* Yadong Wang, Xiaoming Lan, Sai Li, Xuejin Wan, Fei Meng, Yangfan Hu,* Zhen Fan, Chun Feng, Minghui Qin, Min Zeng, Xichao Zhang, Xiaoxi Liu, Weisheng Zhao, Xingsen Gao, and Jun-ming Liu

Magnetic skyrmions are topological swirling spin configurations that hold promise for building future magnetic memories and logic circuits. Skyrmionic devices typically rely on the electrical manipulation of a single skyrmion, but controllably manipulating a group of skyrmions can lead to more compact and memory-efficient devices. Here, an electric-field-driven cascading transition of skyrmion clusters in a nanostructured ferromagnetic/ferroelectric multiferroic heterostructure is reported, which allows a continuous multilevel transition of the number of skyrmions in a one-by-one manner. Most notably, the transition is non-volatile and reversible, which is crucial for multi-bit memory applications. Combined experiments and theoretical simulations reveal that the switching of skyrmion clusters is mediated by the strain-mediated modification of both the interfacial Dzyaloshinskii–Moriya interaction and effective uniaxial anisotropy. The results not only open up a new direction for constructing low-power-consuming, non-volatile, and multi-bit skyrmionic devices, but also offer valuable insights into the fundamental physics underlying the voltage manipulation of skyrmion clusters.

1. Introduction

Magnetic skyrmion is a vortex-like swirling spin configuration with a quantized topological charge.[1–27] When electrons pass through a skyrmion, its twisted spins endow these electrons with an emergent electromagnetic field, yielding a variety of unconventional magnetoelectronic phenomena, such as topological Hall effect[12] and ultralow current density for skyrmion motion.[23–26] These properties together with the nanoscale size and topological stability, make magnetic skyrmion promising potential for future high-density, low-power-consuming magnetic memory devices.[1–3] Like many quasiparticles in condensed matter physics, magnetic skyrmion also has a particle-like characteristic.[1–3] Owing to this feature, multiple skyrmions can aggregate and dissipate in a defined geometry, which holds promise for applications beyond conventional binary memories, such as multi-level memories,[1,28,29] neuromorphic computing,[28–30] probabilistic computing,[31] and nano-oscillators.[32,33] In previous work, the experiment demonstrated an accumulation and dissipation of isolated skyrmions in micrometer-sized geometries using the spin-polarized pulse current.[30] However, since the skyrmion–skyrmion interaction between different

DOI: 10.1002/adma.202107908
isolated skyrmions is negligible, their nucleation and annihilation are stochastic. Consequently, the number of skyrmions cannot be precisely controlled. Moreover, the current scheme leads to a high energy consumption, and the unavoidable Joule heating produced by the current reduces the stability of the information bits. These drawbacks thus underscore the challenging question of how to precisely and controllably manipulate the number of skyrmions in a defined geometry, particularly via a low-dissipation and low-Joule-heating scheme.

It has been widely demonstrated that the isolated skyrmions can be gathered to condense into skyrmion clusters by increasing the area of the geometrical confinement due to a delicate balance between the skyrmion–skyrmion interaction and geometrical confinement. In the cluster state, a certain number of skyrmions are closely coupled with each other, obeying a short-ranged ordering with specific geometrical symmetries. More importantly, the energy density of the cluster states with different numbers of skyrmions is cascading, which allows a controlled accumulation and dissipation of skyrmions by finely tuning the energy density of the magnetic system through external stimuli. Hitherto, several experimental studies have been conducted to understand their behaviors. For instance, Zhao et al. conducted a magnetic-field-driven cascading transition of skyrmion clusters and they observed intermittent jumps between different skyrmion clusters recently, a thermally driven Brown diffusion of skyrmions is also observed. These experiments thus underscore the challenging question of how to precisely and controllably manipulate the number of skyrmions in a defined geometry, particularly via a low-dissipation and low-Joule-heating scheme.

2. Result and Discussion

2.1. Magnetization Process of the Skyrmion Clusters Confined in the FM/FE Multiferroic Heterostructure

Figure 1a shows the structural details of the FM/FE multiferroic heterostructure used in our experiments, which is composed of an array of [Pt(2.5 nm)/Co(2.2 nm)/Ta(1.9 nm)]12 FM nanodots and a (001)-oriented 0.7PbMg1/3Nb2/3O3–0.3PbTiO3 (PMN-PT) FE substrate with Ta (top) and Au (bottom) electrodes. To accommodate the formation of skyrmion clusters in the nanodots, their diameter (d) was fabricated to be approximately 900 nm (see Figure 1b) with a thickness (t) of 60 nm (see Figure 1c). Detailed fabrication procedures are described in Experimental Section.

We first studied the magnetic-field-dependent domain evolution process in the nanodots. An external magnetic field (B) was applied along their out-of-plane direction (marked as a white circled dot in Figure 1d) and the magnetic domain structures at different B were imaged using in situ magnetic force microscopy (MFM) (see Experimental Section). Figure 1d shows the MFM images of a typical nanodot taken at different B (MFM images for an array of nanodots are shown in Figure S1, Supporting Information). For each image, repeated MFM scans were performed to ensure its reproducibility. At the ground state (B = 0 mT), a series of stripe domains were observed in the nanodot. Although the geometrical confinement forces the stripe domains to be distorted in morphology, they maintain a periodical arrangement. By increasing B, the skyrmions are gradually isolated from the stripe domains, creating a mixed state of stripe domains and skyrmions at B = 70 mT. When B increases to 86 mT, the stripe domains disappear completely and the mixed domain state converts into eleven skyrmion clusters, which are densely packed into short-ranged hexagonal lattices. The appearance of these lattices indicates that the skyrmions are strongly coupled with each other through the skyrmion–skyrmion interaction and confirms the formation of skyrmion cluster in the d = 900 nm nanodot. With further increasing of B, the number of skyrmions (N) in the skyrmion cluster state shows a cascading reduction, accompanied by a symmetry variation, due to the delicate balance between the external magnetic field, skyrmion–skyrmion interaction, and geometrical confinement. For instance, six skyrmions condense into a solid-pentagon cluster state at B = 202 mT; as B increases, the N = 6 cluster continuously transforms into N = 5, 4, 3, 2, and 1 states with a hollow-pentagon, square, triangle, double-bell, and single-point geometry, respectively. When B is increased to 225 mT, the N = 1 skyrmion cluster is annihilated and eventually transforms into an out-of-plane FM state (N = 0). Notably, for the magnetization process, the position of
certain skyrmions in the cluster state (e.g., \( N = 1 \) and 4 clusters) deviates from where they are theoretically expected. This observation is attributed to the pinning effect originating from the defects in the nanodot.\(^{[35,38]}\) We should also note here that, when \( B \) increases above 192 mT, the MFM images of skyrmions become vague (see Figure S2, Supporting Information). This is for two reasons: i) the skyrmion size comes near the spatial resolution limit of the MFM (approximately 50 nm); ii) the stabilization of skyrmions decreases, making both their morphology and magnetization behaviors easy to be affected by the subtle stray field from the MFM tip. To address these issues, during MFM measurement, we slightly decreased the magnetic field by several oersted with the number of skyrmions keeping unchanged. This operation not only increases the skyrmion size but also enhances the stability of the skyrmions, allowing us to obtain clearer MFM images of skyrmions within the high magnetic field range, as shown in Figure 1d and Figure S2, Supporting Information.

**2.2. Electric-Field-Induced Multi-State Cascading Transition of Skyrmion Clusters**

After establishing the magnetization process of the skyrmion clusters, the electric-field (\( E \)) effect on their transitions was studied. An external \( E \) was applied on the PMN-PT substrate via the top Ta and bottom Au electrodes (see Figure 1a), and an in-plane strain was generated in the substrate. Such a strain can be transferred to the nanodot via a mechanical coupling between the FE and FM layers. However, due to the mini-size of the nanodot, it is difficult to measure the transferred strain (\( \varepsilon_t \)) directly. Instead, its distribution was simulated via a finite element analysis (see Figure 2a and Note S1, Supporting Information). The simulations reveal a nearly uniform \( \varepsilon_t \) in the nanodot except for the edges. To simplify the strain distribution, \( \varepsilon_t \) is assumed to be uniform and represented by \( \varepsilon \) on the PMN-PT substrate. Figure 2b presents a typical \( E-\varepsilon \) curve established in this work. As \( E \) increases from 0 to +10 kV cm\(^{-1}\), \( \varepsilon \) first exhibits a tensile strain (\( \varepsilon > 0 \)) when \( E \) ranges from 0 to +4 kV cm\(^{-1}\) and then converts into a compressive one (\( \varepsilon < 0 \)). The maximum tensile and compressive strains are 0.15% at +3 kV cm\(^{-1}\) and −0.45% at +10 kV cm\(^{-1}\), respectively. As \( E \) sweeps towards the negative range, \( \varepsilon \) shows a non-volatility and is asymmetric with the positive \( E \) range. The \( E \)-dependent variation of \( \varepsilon \) originates from the switching of the FE polarization in the PMN-PT substrate, and their detailed correlations are discussed in Note S2, Supporting Information.

Figure 2c shows a series of MFM images that record the \( E \)-dependent domain evolution processes at five typical magnetic fields. These images reveal that the external assisting magnetic field plays a crucial role in the \( E \)-driven transition of skyrmion clusters. At \( B = 0 \) mT, only stripe domains are observed in a cycle of \( E \) from +10 to −10 kV cm\(^{-1}\) (see Figure S3, Supporting Information), suggesting that \( \varepsilon \) is insufficient to drive the stripe-skyrmion conversion without applying \( B \). However, when \( B \) is increased to 55 mT, the stripe domains gradually transform...
into skyrmions and eventually convert into an $N = 11$ skyrmion cluster state under the tensile strain. Compared with the $N = 11$ cluster stabilized by a pure magnetic field ($B = 86$ mT), the assisting magnetic field decreases significantly, suggesting that the tensile strain serves as an efficient field that shifts the $N = 11$ skyrmion cluster to a lower energy state. As $E$ increases further, the strain transforms into the compressive type. In contrast to the assisting role of the tensile strain, the compressive strain plays a similar role to the decrease of $B$, which forces the skyrmion clusters to switch back to the stripe domain. As increasing $B$ to 195 mT, the initial state ($ε = 0\%$) transforms from the stripe domain to the $N = 7$ skyrmion cluster. Since the tensile strain has a similar effect as increasing $B$, it is reasonable to observe a decrease of $N$ from 7 to 6 within the tensile strain range. However, although the maximum compressive strain is three times larger than that of tensile strain, only a binary switching between $N = 6$ and 7 skyrmion clusters is observed. By further increasing $B$ to 210 mT and above, the variation of $N$ becomes sensitive to the compressive strain and the expected multi-bit switching of skyrmion clusters occurs. In particular, when $B$ increases up to 217 mT, we have obtained a continuous four-bit switching of $N$ from zero to three in the $E$ range from 0 to +10 kV cm$^{-1}$. As $E$ sweeps towards the negative range, the variation of skyrmion clusters shows an asymmetric feature, due to a lower strength of strain. Importantly, after the $E$ cycle, the skyrmion cluster can return to the initial state ($N = 1$ skyrmion cluster), suggesting that the switching exhibits a reversal feature that is well suited to build magnetic memories. We also find that multi-bit switching occurs at a narrow $B$ range. When $B$ is increased slightly to 210 mT, only a binary switching between the $N = 1$ skyrmion cluster and FM state is observed. Here, it should be noted that the $E$-driven multi-bit switching of skyrmion clusters is not limited to the single nanodot shown in Figure 2c, and similar results are obtained in an array of nanodots (see Figure S4, Supporting Information) and different samples (see Figure S5, Supporting Information). These results strongly support the reliability of the observed $E$-driven multi-state switching of skyrmion clusters.

We further carried out the four-bit switching via the electric-field pulse (pulse width is 1 ms) under a fixed $B$ of 217 mT (see Figure 3 and Figure S6, Supporting Information). This feature reveals the non-volatility of the strain-induced switching of skyrmion clusters. Moreover, the average energy dissipation for the creation and annihilation of a single skyrmion is estimated to be 713 and 63 pJ, respectively (see Note S3, Supporting Information). These values are more than three orders of magnitude lower than that required for the current-induced manipulation and motion of skyrmions.$^{[10,11,30]}$ We note that the thickness of the PMN-PT substrate used in our experiments is 200 µm. If the thickness was decreased to 1 µm, the energy dissipation could potentially be lower than 5 fJ for the creation or annihilation of a single skyrmion. From a practical point of view, the non-volatile multi-state switching in the geometrically confined nanodots provides a compelling route to design the low-energy-consumption, multi-bit skyrmionic devices.

2.3. Discussion

We experimentally achieved a reliable strain-mediated $E$-driven switching of skyrmion clusters based on the nanostructured FM/FE multiferroic heterostructure. In the following, its physical mechanism will be examined. First, we have calculated the magnetic-field-dependent free energy of the $N = 0$, 1, 2,...11 skyrmion clusters ($G_N$) and the energy barrier between two adjacent skyrmion clusters ($G_N^{N\rightarrow N+1}$) at the strain-free state ($ε = 0\%$) and $T = 300$ K (see Experimental Section and Notes S4 and S5, Supporting Information). Figure 4a shows $G_N$ ($0\%$) and

---

Figure 2. Electric-field-induced variation of $N$. a) Simulated strain distribution on a $d = 900$ nm, $t = 60$ nm nanodot subjected to a constant biaxial compressive strain of 0.4% from the PMN-PT substrate. The color bar represents the absolute value of the strain. b) Transferred strain $ε$ of the nanodot in a cycle of $E$ ranging from $+10$ to $−10$ kV cm$^{-1}$. Positive $E$ represents the direction of applied $E$ is opposite to that applied to polarize PMN-PT to saturation at the initial state and negative $E$ represents the direction of applied $E$ is the same as the direction of the initial $E$ for polarization. Positive $ε$ (red dots) represents the tensile strain while negative $ε$ (blue dots) represents the compressive strain. c) MFM images of the $d = 900$ nm [Pt/Co/Ta]$_2$ nanodot in a cycle of $E$ ranging from $+10$ to $−10$ kV cm$^{-1}$ under different $B$. MFM images were taken during the application of electric field. The MFM images enclosed by the black dashed box demonstrate the variation process of a four-state switching of the number of skyrmions. The MFM contrast represents the MFM tip resonant frequency shift ($Δf$). The scale bar in (c) represents 500 nm.
Generally, if a strain-driven cascading switching from the $N_1$ to $N_2$ skyrmion cluster state occurs ($N_1$ and $N_2$ represent the number of skyrmions in the cluster state), the following two conditions must be satisfied: i) the free energy of the $N_1$ skyrmion cluster at the strained state $G_{\text{strain}}(N_1)$ should be lower than that of the $N_1$ skyrmion cluster $G_{N_1}(\varepsilon)$, namely $G_{\text{strain}}(N_1) < G_{N_1}(\varepsilon)$, which stimulates $N_1$ state to evolve toward the $N_2$ state; ii) the absolute value of the strain-driven free energy differences of the $N_1$ state $|\Delta G_{\text{strain}}(\varepsilon)|$ is larger than the sum of the absolute value of energy barriers between the $N_1$ and $N_2$ clusters at the strained state $G_{\text{strain}}^{N_1 \rightarrow N_2}(\varepsilon)$

$$G_{\text{strain}}^{N_1 \rightarrow N_2}(\varepsilon) = \sum_{i=N_1}^{N_2} |\Delta G_{i}(\varepsilon)|,$$

namely $|\Delta G_{N_1}(\varepsilon)| > G_{\text{strain}}^{N_1 \rightarrow N_2}(\varepsilon)$, so that the energy of the system gained from the strain will be sufficient to make the $N_1$ state overcome a series of energy barriers between $N_1$ and $N_2$ and transform into the $N_2$ state. Details about the model are described in Note S6, Supporting Information. Figure 4b,c shows the calculated $\varepsilon$-dependent $G_{N_1}$, $G_{\text{strain}}^{1 \rightarrow N_2}$, and $|\Delta G_{1}|$. When the strain is applied, $G_{N_1}$, $G_{\text{strain}}^{1 \rightarrow N_2}$, and $|\Delta G_{1}|$ vary accordingly. Once the two conditions ($G_{N_1}(\varepsilon) < G_{1}(\varepsilon)$ and $|\Delta G_{1}(\varepsilon)| > G_{\text{strain}}^{1 \rightarrow N_2}(\varepsilon)$) are satisfied, the switching from the 1 to $N$ cluster state is expected to occur. Based on the theoretical model, the strain-driven switching is simulated via Monte-Carlo simulations at the experimentally established strain range and $T = 300$ K. We use a large variety of configurations as the starting state and find the final state with minimal free energy within the switching restrictions. As shown in Figure 4d, the strain-driven four-bit switching between the $N = 0$, 1, 2, and 3 skyrmion clusters are reproduced well, which strongly confirms the reliability of our model. We further find that the strain-induced switching of skyrmion clusters is always a metastable transition. For example, when the transition from the $N = 1$ to 2 cluster state occurs at $\varepsilon = -0.1\%$, the minimum free energy corresponds to the $G_{7}$ ($N = 7$ skyrmion cluster) rather than $G_{2}$, as shown in Figure 4b. It is because that $|\Delta G_{1}|$ at $\varepsilon = -0.1\%$ is limited and insufficient to overcome the sum of the energy barriers between $N = 1$ and 7 states (see Figure 4c). Despite the metastability, the strain-induced clusters show strong robustness due to the high $G_{2}^{N_1 \rightarrow N_2}(\varepsilon)$ (see Figures 2c and 3a). This feature is crucial for constructing multi-bit memories based on the strain-induced cluster states.

Next, the respective roles of DMI and uniaxial anisotropy in the switching were analyzed. Based on the free energy equation shown in Experimental Section, the strain-induced free energy difference can be expressed as $\Delta G_{\text{DMI}}(\varepsilon) = \Delta G_{\text{DMI}}^{\text{Uni}}(\varepsilon) = \Delta G_{\text{DMI}}^{\text{Uni}}(\varepsilon) + \Delta G_{\text{DMI}}^{\text{Uni}}(\varepsilon)$, where $\Delta G_{\text{DMI}}^{\text{Uni}}(\varepsilon)$ and $\Delta G_{\text{DMI}}^{\text{Uni}}(\varepsilon)$ denote the strain-induced energy difference contributed by the uniaxial anisotropy and DMI, respectively. Figure 4e presents the strain-dependent $\Delta G_{\text{Uni}}^{\text{Uni}}$ and $\Delta G_{\text{DMI}}^{\text{Uni}}$ for the $N = 1$ cluster state. Notably, $\Delta G_{\text{DMI}}^{\text{Uni}}(\varepsilon)$ was calculated based on the strain-dependent $K_{\text{eff}}(D)$, with $D$ ($K_{\text{eff}}$) fixed to be a constant at the strain-free state. The results indicate that $\Delta G_{\text{Uni}}^{\text{Uni}}$ is positive while $\Delta G_{\text{DMI}}^{\text{Uni}}$ is negative within the compressive strain range, revealing that the strain-induced variations of DMI and uniaxial anisotropy play opposite roles in the switching: that is, the variation of uniaxial anisotropy pushes the system to overcome the energy barriers while the DMI variation suppresses the switching of skyrmion clusters. Moreover, the absolute value of $\Delta G_{\text{Uni}}^{\text{Uni}}$ ($|\Delta G_{\text{DMI}}^{\text{Uni}}|$) is demonstrated to be always larger than $|\Delta G_{\text{DMI}}^{\text{Uni}}|$, suggesting that the variation of uniaxial anisotropy dominates the strain-induced

**Figure 3.** Non-volatile switching of $N = 0, 1, 2, 3$ skyrmion clusters induced by using pulse electric field with a pulse width of 1 ms under a fixed magnetic field of 217 mT. The insets show the MFM images taken after the pulse electric field. MFM contrast represents the MFM tip resonant frequency shift ($\Delta f$). Inset number represents the number of skyrmions ($N$) in the skyrmion cluster state. The scale bar is 1 $\mu$m.
switching of skyrmion clusters in the Pt/Co/Ta multilayered film.

Furthermore, we simulated the strain-induced switching of skyrmion clusters at a relatively low magnetic field (see Figure S7, Supporting Information). The simulations reveal that the variation of the number of skyrmions is more sensitive at a high magnetic field than that at a low magnetic field, which is well consistent with the experimental observations (see Figure 2c, Figures S4 and S5, Supporting Information). To understand this point, $\Delta G_{\text{Uni}}$ and $\Delta G_{\text{Ele}}$ at different $B$ were compared (see Figure S8, Supporting Information). We find that the enhancement of $B$ increases $|\Delta G_{\text{Uni}}|$ but decreases $|\Delta G_{\text{Ele}}|$ leading to a larger $\Delta G_{\text{Uni}}(\varepsilon)$ at a higher magnetic field. This is because the increasing $B$ enlarges the out-of-plane component of magnetization ($M_{ij}$) and suppresses the modulation part of magnetization within a skyrmion.\(^{44}\) Since the $\Delta G_{\text{Uni}}$ determines whether the system could gain sufficient energy to overcome the energy barriers between two skyrmion clusters, this explains the observed magnetic-field-dependent tunability of skyrmion clusters.

3. Conclusions

We have conducted an electric-field-induced cascading transition of skyrmion clusters in the nanostructured FM/FE multiferroic heterostructure, which allows a continuous multilevel transition of the number of skyrmions, one by one. The energy for the voltage operation can be more than three orders of magnitude lower than that of the current scheme,\(^{10,11,16}\) leading to the development of low-power, multi-bit skyrmionic devices. Moreover, the transition is non-volatile and reversible, which is well suited for the construction of skyrmionic devices. Theoretical simulations reveal that the variation of uniaxial anisotropy promotes the transition while it is suppressed by the DMI variation.

4. Experimental Section

Fabrication of the FM/FE Multiferroic Heterostructure: i) [Pt(2.5 nm)/Co(2.2 nm)/Ta(1.9 nm)]\(_{12}\) multilayered films were deposited on PMN-PT substrate by using magnetron sputtering. ii) Monolayer polystyrene
sphere particle arrays were paved on the surface of the magnetic film. iii) Poly styrene sphere particles were etched to a diameter of 900 nm by using oxygen plasma. iv) An Au-ion beam was used to etch the magnetic film with an appropriate etching time and the film blocked by poly styrene sphere particles was protected while the rest was etched. v) Poly styrene sphere particles were removed by chloroform solution and the periodically ordered nanodots were obtained.

**MFM Measurements** MFM measurements were performed by using scanning probe microscopy (MFP-3D, Asylum Research). To exclude the magnetic field from the MFM tip on the magnetic domain structures, a low-moment magnetic tip (PPP-LM-MFMR, Nanosensors) was used, and the distance between the tip and sample was maintained at a constant distance of 30 nm. VFM3 component (Asylum Research) was integrated into the MFP-3D to vary the perpendicular magnetic field.

**Theoretical Calculations** For the magnetic system subjected to an equiaxial strain, the free energy (G) can be described as

$$G(M) = \int \left\{ A M^2 + \frac{\partial M}{\partial X_1} \frac{\partial M}{\partial X_1} + D(T+\Gamma)(M_{11}+L_{222}) \right\} dV$$

where $A$ denotes the magnetic exchange constant, $M$ denotes the magnetization vector, $\Gamma$ denotes the slope of strain-dependent $D$ curve, $L_{ij} = \frac{\partial M_j}{\partial X_i} - M_i \frac{\partial M_j}{\partial X_i}$ denotes the Lifshitz invariants, $B = (0, 0, B)$ denotes the applied magnetic field, $\Lambda$ denotes the slope of the $\epsilon$-dependent $K_{\alpha}$ curve, $\alpha$ denotes the second-order Landau expansion coefficient, $\beta$ denotes the fourth-order Landau expansion coefficient, and $T$ denotes the temperature. Detailed derivation procedures of Equation (1) are described in Note S4, Supporting Information. It should be noted here that, the values and units of the parameters $A$, $D$, and $K_{\text{eff}}$ are different from those used in ref. [41]. This was because that the two Landau expansion terms were included in the model while the model used in ref. [41] does not contain the two terms. The magnitude of magnetization in the model was assumed to be 1 at $T = 300K$ and their relationship are listed below:

$$M_s(T = 300K) = M_s(T = 300K) = 697kA/m$$

$$A = \frac{A'}{M_s(T = 300K)} = 1.85 \times 10^{-2} J/A^2$$

$$D = \frac{D'}{M_s(T = 300K)} = 2.20 \times 10^{-1} J/A^2$$

$$K_{\text{eff}} = \frac{K_{\text{eff}}'}{M_s(T = 300K)} = -1.92 \times 10^{-2} J/m^2$$

where $A'$, $D'$, $K_{\text{eff}}'$, and $M_s(T = 300K)$ represents the magnetic exchange constant, DM constant, uniaxial anisotropy constant, and saturation magnetization (at 300 K) in the model without considering the two Landau expansion terms, and their values were 9 J/m$^{-1}$,[5] 1.07 $\times 10^{-3}$ J/m$^{-1}$, 0.93 $\times 10^{-3}$ J/m$^{-1}$, and 697 kA/m$^{-1}$,[6] respectively. Notably, both the values of $A'$ and $K_{\text{eff}}'$ were slightly larger than those (1.01 m$^{-1}$) for $D'$ and $-0.97 \times 10^{-3}$ J/m$^{-1}$ for $K_{\text{eff}}'$ reported in ref. [41]. Moreover, $\Gamma$ denotes the slope of strain-dependent $D$ curve and was equal to be $-39^{[6]} \Lambda$, $\alpha$ denotes the slope of the $\epsilon$-dependent $K_{\text{eff}}'$ curve and was equal to be $-50^{[6]} \alpha$ and $\beta$ were fitted from the equations: $[46]$ and $[47]$.

$$M_s(T = 300K) = \sqrt{\frac{\alpha(T_0 - 300)}{2 \beta}} = 697kA/m$$

$$\frac{D^2}{4 \alpha} + T_0 = T_0 = 670kA/m$$

$$t = \frac{4 \alpha (300 - T_0)}{D^2}$$

The values of $\alpha$ and $\beta$ are determined to be $5.3 \times 10^{-10}$ J/m$^{-1}$ and $3.3 \times 10^{-10}$ J/m$^{-1}$, respectively.

If a strain-induced phase transition between an $N$ skyrmion cluster state and an $N + 1$ skyrmion cluster state was to occur, a necessary condition was that the absolute value of the free energy differences $|\Delta G_{\text{uni}}(\epsilon)|$ (where $\Delta G_{\text{uni}}(\epsilon)$) must be larger than the absolute value of the energy barrier $|G_{\text{uni}}^{\text{N+1}}(\epsilon) - G_{\text{uni}}^{\text{N}}(\epsilon)|$) between the two states. In this case, by calculating the total free energy change $|\Delta G_{\text{uni}}(\epsilon)| = |\Delta G_{\text{uni}}(\epsilon) - G_{\text{uni}}^{\text{N+1}}(\epsilon)|$ of the magnetic system that can gain energy was obtained through the change of strain from $\epsilon = 0$ to $\epsilon = \epsilon_0$. The magnitude of the strain on the free energy was composed of two parts, for which $\Delta G_{\text{uni}}(\epsilon) = -K_{\text{uni}}(\epsilon) + G_{\text{uni}}^{\text{N+1}}(\epsilon)$ can be written as, where

$$\Delta G_{\text{uni}}(\epsilon) = -K_{\text{eff}} M^2$$

$$\Delta G_{\text{uni}}^{\text{N+1}}(\epsilon) = D T_0 (L_{11} + L_{222})$$

Notably, Equations (7) and (8) are only approximately correct, because, during a change of $\epsilon$, the magnetization distribution was changed even in the same state (i.e., the magnetization field of the $N$ skyrmion cluster state was not the same at the two conditions, i.e., $\epsilon = 0$ and $\epsilon = \epsilon_0$).

**Supporting Information**

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

**Acknowledgements**

The authors thank for the financial supports from the National Key Research and Development Program of China (No. 2020YFA0309300), the National Natural Science Foundation of China (Nos. 11904018, 12172090, 11772360, 11974298, 92163210, and 6196136006), and the Science and Technology Program of Guangzhou (No. 202002030052). X.Z. was an International Research Fellow of the Japan Society for the Promotion of Science (JSPS). X.Z. was supported by JSPS KAKENHI (Grant No. JP20F0363). X.L. acknowledges the support by the Grants-in-Aid for Scientific Research from JSPS KAKENHI (Grant No. JP20F0363, JP21H01364, and JP21K18872). Y.Z. acknowledges the support by the Guangdong Special Support Project (Grant No. 2019B030203030), Shenzhen Peacock Group Plan (Grant No. KQTD2018041318702403), and Pearl River Recruitment Program of Talents (Grant No. 2017GC010293).

**Conflict of Interest**

The authors declare no conflict of interest.

**Author Contributions**

Z.P.H. conceived and designed the experiments. C.F., F.M., and Y.D.W. synthesized the heterostructures. Y.D.W. performed the MFM measurements. Y.F.H., X.M.L., X.J.W., and Z.P.H. performed the theoretical analysis and micromagnetic simulations. The manuscript was drafted by Z.P.H. with contributions from Y.F.H., X.C.Z., G.F.Z., Y.Z., X.X.L., G.H.Y., X.S.G., and J.M.L. All authors discussed the results and contributed to the manuscript.
Data Availability Statement

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

Keywords

electric fields, low-power-consuming devices, multi-state switching, skyrmion clusters

Received: October 3, 2021
Revised: December 18, 2021
Published online: