

Enhanced Curie temperature and room-temperature 50-nm skyrmions achieved in hexagonal ferromagnet $\text{Mn}_5\text{Ge}_{3.2}$ synthesized via a high-pressure method

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Received September 11, 2025; accepted November 18, 2025; published online February 9, 2026

The development of new high-temperature ultrasmall-size skyrmion materials holds immense significance for the promising applications of topological spintronic devices. In this study, we demonstrate that a high-pressure synthesis technique can significantly elevate the Curie temperature of $\text{Mn}_5\text{Ge}_{3.2}$ crystals, from 294 to 350 K. It is possible that this enhancement arises from the combined effects of lattice contraction and increased Ge content, a conclusion supported by our Density Functional Theory calculations. Additionally, our real-space magnetic imaging reveals the stability of dipolar skyrmions with diameters of approximately 50 nm at room temperature and zero magnetic field. Our micromagnetic simulations closely replicate the diverse experimental topological magnetic textures observed. Furthermore, magnetotransport measurements indicate the potential for the electrical distinction between various topological magnetic textures in skyrmion-based devices. We also report deterministic manipulations on single dipolar skyrmions in confined nanostructures by using in-plane currents. The observation, electrical manipulation, and electrical detection of room-temperature ultrasmall topological magnetic textures underscore the potential of $\text{Mn}_5\text{Ge}_{3.2}$ as a promising platform for spintronic device applications.

$\text{Mn}_5\text{Ge}_{3.2}$, Curie temperature, dipolar skyrmions, asymmetric magnetoresistance

Citation: Y. Zhang, W. Liu, M. Shi, S. Zhang, S. Qiu, Y. Wu, J. Jiang, H. Zhang, H. Han, K. Wang, D. Shao, Z. Zi, C. Ma, H. Du, M. Tian, S. Wang, and J. Tang, Enhanced Curie temperature and room-temperature 50-nm skyrmions achieved in hexagonal ferromagnet $\text{Mn}_5\text{Ge}_{3.2}$ synthesized via a high-pressure method, *Sci. China-Phys. Mech. Astron.* **69**, 247511 (2026), <https://doi.org/10.1007/s11433-025-2860-4>

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1 Introduction

Magnetic skyrmions, as topologically protected spin textures, have garnered significant interest for their potential applications in future spintronic devices [1-4]. These nanoscale spin configurations, resembling vortex-like structures, hold the promise of ultra-low power consumption and high-density data storage due to their resilience against external perturbations and their facile manipulation with electric currents [5-9]. The stability of skyrmions at room temperature is crucial for their practical implementation in technological devices [10,11]. A reduction in the size of room-temperature skyrmions means that more data can be stored within the same area, which is particularly important for high-density storage media and the miniaturization of devices [1,12-15].

Magnetic skyrmions are commonly observed in materials like FeGe [13] and MnSi, which feature a non-centrosymmetric B20-type cubic crystal structure. This distinctive structure promotes the Dzyaloshinskii-Moriya interaction (DMI), thus facilitating skyrmion stabilization. Magnetic skyrmions have been found to have the smallest size down to sub-10 nm in non-centrosymmetric magnets such as GaV₄S [16], centrosymmetric magnets such as Gd₂PdSi [17], and magnetic multilayers such as Fe/Ir(111) [18], but they stabilize only at very low temperatures. Recent studies have explored room-temperature skyrmion materials, including non-centrosymmetric magnets like Co_xZn_yMn_{20-x-y} [19], Mn_{1.4}Pt_{1-x}Pd_xSn [20], and (Fe_xNi_yPd_{1-x-y})₃P [21], magnetic multilayers [22], and centrosymmetric uniaxial magnets [23-26]. However, the size of room-temperature chiral skyrmions is generally above 100 nm, with only a few reports of sizes below 50 nm in Pt/Co/Ir multilayer films [22]. Therefore, the exploration and identification of new room-temperature 50-nm skyrmion materials remain a critical research objective.

In this study, we report a new room-temperature 50-nm-size skyrmion-hosting hexagonal ferromagnet, Mn₅Ge_{3.2}. Combining Density Functional Theory (DFT) calculations, we propose that lattice contraction and increased Ge content induced by the high-pressure (HP) growth method could elevate the Curie temperature of Mn₅Ge_{3.2} from near-room temperature to above room temperature (350 K). Through Lorentz transmission electron microscopy (Lorentz-TEM), we observed room-temperature nanoscale dipolar skyrmions. Additionally, through magnetotransport measurements, we identify the potential for magnetoresistance detection of magnetic textures. Finally, we demonstrate the deterministic manipulation of individual dipolar skyrmions in confined nanostructures using in-plane currents. These findings elucidate the formation mechanisms of ultrasmall room-temperature topological magnetic structures within ferromagnetic Mn₅Ge_{3.2}, paving the way for the development of novel spintronic devices.

2 Experimental methods

2.1 Sample preparation

Mn₅Ge₃ and Mn₅Ge_{3.2} crystals were prepared through two different growth methods. The first method involved the Ge self-flux method. Specifically, Mn granules (99.9% purity) and Ge granules (99.999% purity) were placed in an alumina crucible and then sealed into a vacuum quartz tube. The quartz tube was placed in a furnace and maintained at 1150°C for 1 d to ensure complete dissolution of Mn in the Ge solution. The furnace temperature was then slowly reduced to 850°C and held at this temperature for 10 h. Finally, the quartz tube was inverted and inserted into a centrifuge to separate the single crystals from the excess Ge flux.

The second method involved HP synthesis. In this process, slightly more than stoichiometric amounts of high-purity Mn (99.9%) and Ge (99.999%) powders were used. The components were melted in a vacuum arc furnace. The resulting ingot was then annealed under high-pressure and high-temperature conditions (8 GPa, 1000°C; model: Max Voggenreiter LPR-1000) for 2.5 h.

Notably, there exists a degree of flexibility around the stoichiometric ratio of Mn₅Ge₃. The binary phase diagram of the Mn-Ge alloy is provided in Figure S1. Even when deviating from the stoichiometric ratio, excess Ge atoms can be accommodated within the single-phase matrix through solid solubility, without precipitating secondary phases. This indicates that compositions moderately enriched in Ge can still maintain the desired single-phase structure, as the lattice is capable of accommodating compositional fluctuations within the solubility limits of the solid solution.

2.2 Preparation of Mn₅Ge_{3.2} lamellas and structure characterization

The 150 and 50-nm-thick Mn₅Ge_{3.2} nanostructured lamellas were fabricated from a bulk single crystal using a standard lift-out method with an SEM-FIB dual-beam system (Helios Nanolab 600i, FEI). The HAADF-STEM images and electron diffraction patterns were acquired using a transmission electron microscope (Thermo Scientific Themis Z 3.2) equipped with a spherical aberration corrector and operated at 300 kV. The stoichiometry is a typical value obtained from EDS analysis across multiple regions, and “ $x \approx 0.2$ ” represents the characteristic enrichment level of Ge within the solid solution achieved by the high-pressure synthesis method.

2.3 Device fabrication and magnetic transport measurement

First, a slice approximately 12 μm in length, 6 μm in width, and 1 μm in thickness was fabricated on the sample using FIB technology. This slice was then transferred onto an intrinsic silicon substrate with template electrodes using a custom-designed transfer platform. Finally, the targeted size and shape of the four-terminal electrode sample were precisely etched using FIB technology. Magnetic transport measurements were conducted using a Physical Property Measurement System (Quantum Design, PPMS). The magneto-transport properties were measured using a standard four-terminal method.

2.4 Lorentz-TEM measurements

In-situ Fresnel imaging was performed in a Lorentz-TEM (Talos F200X, FEI) with an acceleration voltage of 200 kV to investigate magnetic domains in the $\text{Mn}_5\text{Ge}_{3.2}$ lamellas. The TEM holder (model 636.6, Gatan) supported variable temperature measurements.

2.5 First-principles calculations

The exchange stiffness describes the energy of a long-wavelength spin wave excitation. In particular, if the energy as a function of wave number q is written as $E(q) = E_0 + A_{\text{ex}} q^2 + \dots$, the exchange stiffness constant, A_{ex} is seen to be given by the curvature of the $E(q)$ vs. q curve. A_{ex} can be calculated from the Heisenberg exchange parameters as [27]:

$$A_{\text{ex}} = \frac{2\mu_B}{3M} \sum_{\mathbf{R}} \sum_{ij} J_{i0,j\mathbf{R}} r_{i0,j\mathbf{R}}^2,$$

where $r_{i0,j\mathbf{R}}$ is the distance between atom i in cell $\mathbf{0}$ and atom j in cell $\mathbf{R}$.

The magnetic exchange parameters $J_{i0,j\mathbf{R}}$ is calculated based on a real-space Green's function [28,29] implemented in QuantumATK [30], including $\mathbf{k}$ -point density of 12 Angstrom along x , y and z directions. A default cut-off energy, and $\mathbf{k}$ -point density of 7 Angstrom along x , y , and z directions, Perdew-Burke-Ernzerhof (PBE) functional [31], and the PseudoDojo pseudopotentials [32] are used for the self-consistent calculations. The GGA+U functional is included in the calculations [33].

2.6 Micromagnetic simulation

The zero-temperature micromagnetic simulations for magnetic domains in 150-nm-thick lamellas were performed using MuMax3. The numerical simulation parameters were specifically tailored to match the material properties of $\text{Mn}_5\text{Ge}_{3.2}$, including exchange stiffness $A_{\text{ex}} = 2.30 \times 10^{-11} \text{ J m}^{-1}$, saturation magnetization $M_s = 7.58 \times 10^5 \text{ A m}^{-1}$,

and uniaxial anisotropy constant $K_u = 1.14 \times 10^5 \text{ J m}^{-3}$. The cell size adopted for the simulations was 4 nm $\times$ 4 nm $\times$ 4 nm.

3 Results and discussion

3.1 Enhancement of Curie temperature in high-pressure synthesized $\text{Mn}_5\text{Ge}_{3.2}$

MnGe has recently been confirmed as a material capable of hosting skyrmions at low temperatures, existing in a non-centrosymmetric phase wherein one of the smallest known skyrmions is formed [34-36]. Additionally, Mn-Ge phases encompass some centrosymmetric phases that exhibit magnetic ordering temperatures close to room temperature. For example, Mn_5Ge_3 [37,38], a material with a centrosymmetric crystal structure, features gapped Dirac magnons and is renowned for its unique properties as a topological magnon insulator [39]. Mn_5Ge_3 also displays uniaxial magnetic anisotropy [40,41], and the interplay between uniaxial magnetic anisotropy and magnetic dipole-dipole interactions is expected to generate a variety of topological spin configurations [23-25]; however, there are currently few reports in this area. In the manganese germanium series, whether in the non-centrosymmetric phase or the centrosymmetric phases, the Curie temperature is below room temperature. Therefore, there is an urgent need to develop materials capable of hosting small-sized room-temperature skyrmions.

Mn_5Ge_3 is a ferromagnetic material that typically exhibits a hexagonal D_{8h} -type crystal structure with a space group of $\text{P6}/\text{mcm}$ (No.193) [42,43], and the easy axis is aligned along the $[001]$ axis. As shown in Figure 1(a), the magnetic atoms Mn occupy two distinct crystallographic positions, 4d and 6g, with the magnetic moments of Mn1 (0.250, 0, 0.250) and Mn2 (0.333, 0.667, 0.250) being 1.96 μ_B and 3.23 μ_B [44,45], respectively, aligned along the c -axis. The Ge atoms also occupy the 6g positions but with different coordinates (0.610, 0, 0).

The crystal structure of $\text{Mn}_5\text{Ge}_{3.2}$ was analyzed using high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM). Both $[001]$ - and $[010]$ -oriented lamellae (with a thickness of $\approx 45 \text{ nm}$) were prepared by the lift-out method using a focused ion beam (FIB) dual-beam system. Figure 1(d) presents an atomic-resolution HAADF-STEM image of $\text{Mn}_5\text{Ge}_{3.2}$ (HP) oriented along the $[001]$ direction. A series of regularly arranged bright and lighter spots represents the positions of atoms. Variations in brightness reflect their respective average atomic numbers, with brighter spots corresponding to heavier elements (Ge) and lighter contrast regions representing lighter elements (Mn) [46]. This allows us to determine the specific occupancies of Mn and Ge atoms, which is further confirmed by X-ray energy-dispersive spectroscopy elemental mapping (EDS-Mapping) analysis (Figure S2). Selected-area electron

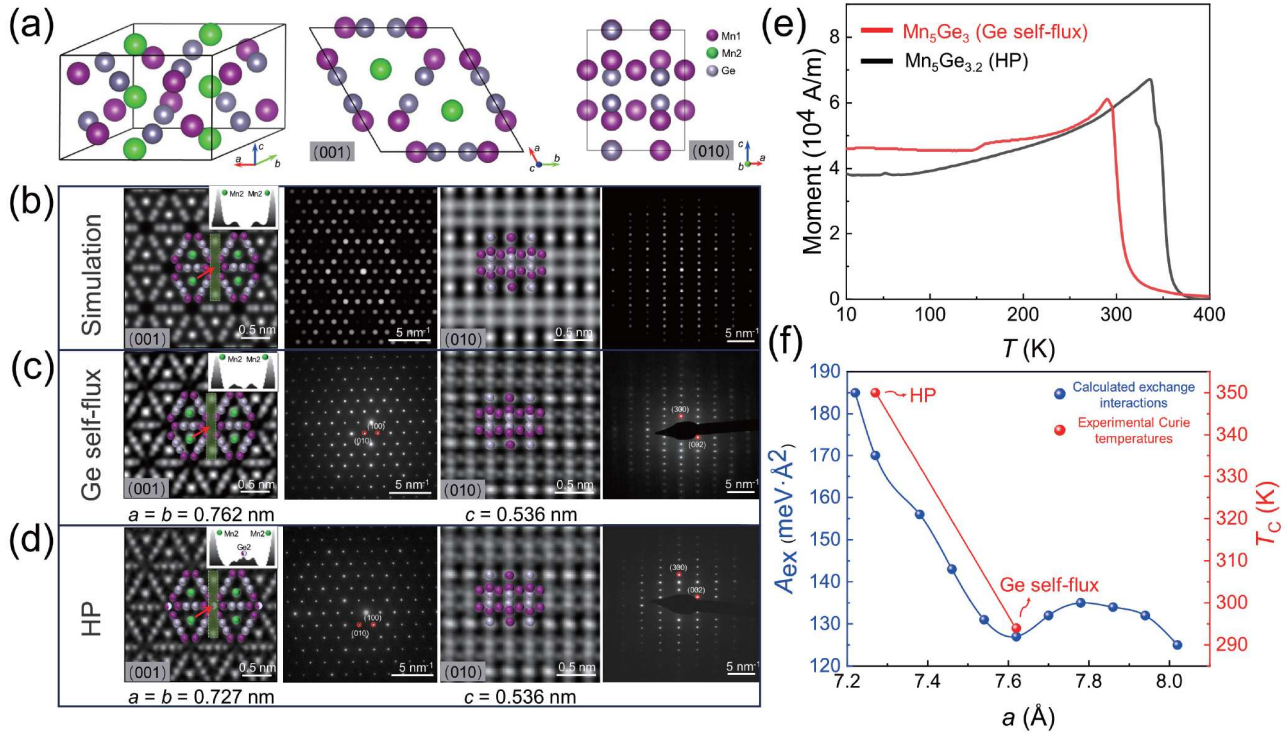


Figure 1 (Color online) (a) The crystal structure of Mn_5Ge_3 , with top-view and cross-sectional views along the c -axis and b -axis. (b)–(d) The high-resolution STEM images of the (001) and (010) planes for simulated Mn_5Ge_3 , Mn_5Ge_3 grown by the Ge self-flux method, and $\text{Mn}_5\text{Ge}_{3.2}$ synthesized by the HP method, along with the corresponding SAED patterns. (e) M - T curves of Mn_5Ge_3 (growth by the Ge self-flux method) and $\text{Mn}_5\text{Ge}_{3.2}$ (synthesis by the HP method) measured during subsequent heating after cooling under a constant 20 mT magnetic field applied along the [001] direction. (f) Calculated exchange interactions of Mn_5Ge_3 as a function of the lattice parameter using DFT calculations, with c fixed at 5.36 Å, and experimental Curie temperatures for Mn_5Ge_3 and $\text{Mn}_5\text{Ge}_{3.2}$ crystals grown by the Ge self-flux and the HP methods.

diffraction (SAED) confirms that this crystal plane is oriented along the [001] direction. When comparing the atomic arrangement of HP synthesized $\text{Mn}_5\text{Ge}_{3.2}$ (001) with the schematic representations of simulated Mn_5Ge_3 (001) and single-crystal Mn_5Ge_3 (001) grown by the Ge self-flux method, we find that the (000) position of the hexagonal network in the $\text{Mn}_5\text{Ge}_{3.2}$ lattice (marked by the red arrow) is occupied by excess Ge atoms, a fact also supported by the profile of the green rectangle area. EDS analysis also confirmed that the crystal is enriched with Ge (Figures S2 and S3). As shown in Figure 1(b), the HAADF-STEM image showing atoms aligned along the [010] orientation displays a basic layer-like arrangement of Mn-Ge atomic columns, which matches perfectly with the Mn_5Ge_3 (010) and $\text{Mn}_5\text{Ge}_{3.2}$ (010) crystal structure in Figure 1(c) and (d), strongly corroborating the high quality of the crystal. Notably, the $\text{Mn}_5\text{Ge}_{3.2}$ crystals synthesized using the HP method can accommodate a certain degree of compositional fluctuations within the solubility range of the solid solution, thereby exhibiting the presence of Ge-rich phases. However, the fundamental crystallographic framework of these crystals remains consistent with that of typical Mn_5Ge_3 . To accurately describe the crystal structure characteristics obtained under HP conditions, we introduce the notation $\text{Mn}_5\text{Ge}_{3.2}$ to

represent this structure.

Figure 1(e) shows the typical magnetization-temperature (M - T) curves of Mn_5Ge_3 (Ge self-flux) and $\text{Mn}_5\text{Ge}_{3.2}$ (HP) measured during subsequent heating after cooling under a constant magnetic field of 20 mT applied along the [001] direction (field-cooling (FC)). The figure indicates that the T_C of Mn_5Ge_3 (Ge self-flux) is approximately 294 K, near room temperature; whereas the T_C of $\text{Mn}_5\text{Ge}_{3.2}$ (HP) is approximately 350 K, significantly exceeding room temperature. We speculate that this can be directly attributed to the changes in lattice parameters during the HP synthesis of $\text{Mn}_5\text{Ge}_{3.2}$ [47], resulting in a lattice constant of ($a = b = 0.727$ nm, $c = 0.536$ nm) for $\text{Mn}_5\text{Ge}_{3.2}$. Compared to Mn_5Ge_3 grown by the Ge self-flux method ($a = b = 0.762$ nm, $c = 0.536$ nm, see Figure S4 for lattice constant determination), the lattice constants (a and b) are reduced by approximately 4.6%. This alteration impacts the electronic structure of the material, thereby affecting the magnetic interactions between Mn ions [48,49]. The modification of these interactions allows the material to maintain its ferromagnetic state at higher temperatures. Additionally, differences in the arrangement or chemical environment of Ge atoms also influence the T_C . Our EDS experiments reveal that the Ge content slightly increased by 6.6% (to $\text{Mn}_5\text{Ge}_{3.2}$) following the HP method.

DFT calculations indicate that this slight increase in Ge content results in an enhancement of the exchange stiffness constant. Consequently, it is possible that the increase in the T_C observed in the $\text{Mn}_5\text{Ge}_{3.2}$ crystal is due to the combined effects of lattice contraction and the increased Ge content (Figure 1(f) and Figure S5), and further systematic investigations are required to establish a definitive correlation between Ge content and magnetic properties. When the lattice parameter of the crystal decreases compared to $a = 7.62 \text{ \AA}$, the exchange interactions increase. Specifically, when a contract from $7.62 \text{ \AA}$ for Mn_5Ge_3 (Ge self-flux) to $7.27 \text{ \AA}$ for $\text{Mn}_5\text{Ge}_{3.2}$ (HP), the dominant exchange parameter increases from 127 to $170 \text{ meV \AA}^2$. The magnetization curves at temperatures of 2 and 300 K are provided in Figure S6, and we determined the uniaxial anisotropy constant K_u of $\text{Mn}_5\text{Ge}_{3.2}$ (HP) to be approximately $1.14 \times 10^5 \text{ J m}^{-3}$, and the saturation magnetization M_s to be $7.58 \times 10^5 \text{ A m}^{-1}$ at 300 K . Since the ferromagnetic-paramagnetic transition temperature is below room temperature, no topological spin structures were observed in the Mn_5Ge_3 lamella grown by the Ge self-

flux method at room temperature using Lorentz-TEM (Figure S7). All references to $\text{Mn}_5\text{Ge}_{3.2}$ hereafter pertain to samples synthesized using HP synthesis.

3.2 Observation of room-temperature ultrasmall topological magnetic textures in $\text{Mn}_5\text{Ge}_{3.2}$ lamella

We performed magnetic domain observations on $\text{Mn}_5\text{Ge}_{3.2}$ lamella cut along the $[001]$ direction using Lorentz-TEM, which enables visualization of in-plane magnetization (M_{xy}). As shown in Figure 2(a), typical stripe domains appear in a 150-nm -thick $\text{Mn}_5\text{Ge}_{3.2}$ lamella prepared at zero magnetic field and room temperature, with an average width of $\approx 150 \text{ nm}$, as shown in Figure 2(b). These domains are stabilized by the combined effect of perpendicular magnetic anisotropy and dipole-dipole interactions. Upon application of an out-of-plane magnetic field, the stripe domains gradually shrink (Figure 2(a), 230 mT) and transform into dipolar skyrmions at high fields (Figure 2(a), 368 mT). Due to the centrosymmetry of the hexagonal ferromagnet, there is

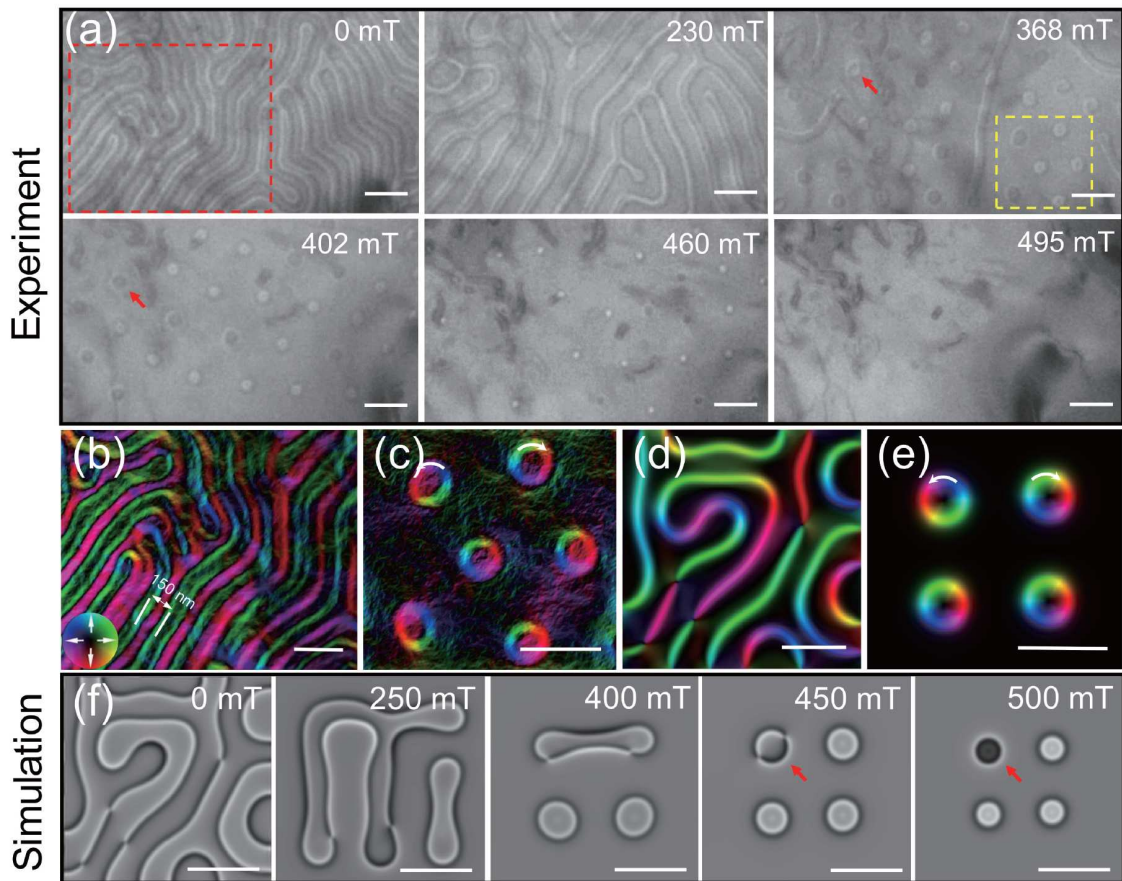


Figure 2 (Color online) Observation of dipolar skyrmions above room temperature. (a) Magnetic evolutions from zero-field stripe domains driven by an out-of-plane magnetic field at room temperature. (b) and (c) Representative in-plane magnetization mappings of stripe domains and dipolar skyrmions retrieved from TIE analysis, correspond to the red and yellow dashed rectangles in (a), respectively. (d) and (e) The in-plane magnetization mappings of stripe domains and dipolar skyrmions, obtained from TIE analysis, as shown in (f). (f) Corresponding simulated Fresnel images in the field-increasing process. The color wheel represents the in-plane magnetizations. Defocus, $-800 \mu\text{m}$. Scale bar, 300 nm .

no chiral DMI, leading to the observation of coexisting skyrmions with opposite winding directions [50] (counter-clockwise skyrmions appear as black underfocused Fresnel contrast, while clockwise skyrmions appear as white underfocused Fresnel contrast), as shown in Figure 2(a) and (c). It is evident that in this state, stripe domains, fragmented magnetic domains, and dipolar skyrmions coexist. With a further increasing magnetic field, the residual stripe domains first gradually evolve into dumbbell-shaped domains and then completely shrink into dipolar skyrmions (Figure 2(a), 402 mT). As the magnetic field increases further, the size of the dipolar skyrmions decreases, and they ultimately annihilate in the ferromagnetic state. These observations clearly indicate that dipolar skyrmions can be obtained by applying a magnetic field perpendicular to the lamella. Energy calculations in Figure S8 reveal that the formation of skyrmions is a direct consequence of the competition between the exchange, Zeeman, perpendicular magnetocrystalline anisotropy, and demagnetization energies, with the demagnetization energy playing a critical role in the stabilization process. The process of entire field-driven evolution is well reproduced in our simulations, as shown in Figure 2(f). Additionally, the corresponding TIE analysis fully corroborates the experimental observations (Figure 2(d) and (e)). Notably, the field-increase process generates a small number of type-II magnetic bubbles (Figure 2(a)), which undergo a topological transition to type-I magnetic bubbles, also known as dipolar skyrmions (as indicated by red arrows). The topological transition process is also highly reproduced in our simulations (Figure 2(f)). However, the variation in Ge content has a multifaceted impact on the material's physical properties. It not only modulates the exchange interaction but may also alter parameters such as the magnetocrystalline anisotropy. Thus, whether excess Ge critically affects the size or stability of skyrmions remains an open question, requiring further systematic experimentation with controlled Ge content to reach a definitive conclusion.

At room temperature, by first increasing the magnetic field to generate dipolar skyrmions and then reducing the field to zero, the dipolar skyrmions remain stable in the zero magnetic field, as shown in Figure S9. The size of the dipolar skyrmions decreases gradually with increasing magnetic field, ranging from approximately 60 to 150 nm, comparable to those observed in chiral CoZnMn [2] (≈ 115 – 117 nm), Fe_{0.5}Co_{0.5}Si [51] (≈ 90 nm), and in magnetic multilayers [52]. When the applied magnetic field is below 414 mT, the rate of decrease in the size of dipolar skyrmions is relatively small; when it exceeds 414 mT, the rate of decrease markedly increases. As the magnetic field increases to a certain value, the external field gradually dominates, altering the internal energy balance of the skyrmions. To minimize the total system energy, skyrmions adjust their size and shape to adapt to the changes in the external field, which leads to an

accelerated reduction in skyrmion size [53]. The dipolar skyrmions ultimately annihilate at 495 mT, as shown in Figure 3(a). When the lamella thickness is reduced to 50 nm, the skyrmion diameter exhibits a slight decreasing trend with increasing magnetic field, declining from 58 nm at 0 mT to 52 nm at 368 mT (Figure 3(c)). This behavior can be attributed to a reorganization of the competing energy terms at the limiting thickness. As the magnetic field increases, the Zeeman energy and demagnetization energy converge and nearly cancel each other out, while the anisotropy energy exhibits only a marginal decrease (Figure S10(c)). Energy analysis confirms that the sensitivity of the skyrmion size to external magnetic fields is significantly reduced in such an ultrathin sample—a conclusion well supported by experimental data (Figure S10(a) and (b)). The long-term stability of the 50-nm dipole skyrmions was confirmed via a 24-h test at room temperature and zero field. Lorentz-TEM comparison and statistical analysis show a skyrmion retention rate of $\sim 100\%$ and negligible size variation (Figure S11), demonstrating exceptional robustness for practical applications. During zero-field warming from 100 to 350 K, the ground-state striped domains maintain strong structural stability, with their period remaining essentially constant at about 150 nm ($t \approx 150$ nm, Figure 3(a)) and 50 nm ($t \approx 50$ nm, Figure 3(c)). This indicates that, while thermal fluctuations increase with rising temperature, the contribution of perpendicular magnetic anisotropy is sufficiently large to counteract the thermal fluctuations, thus maintaining the periodicity of the striped domains.

Kittel's Law [54,55] describes the relationship between the period d of magnetic or polar domains and the film thickness t , commonly expressed as $d \propto \sqrt{t}$. The essence of this law lies in how ferromagnetic systems adjust their domain wall configurations to minimize the sum of exchange energy and anisotropy energy, thereby achieving an energetically optimal domain structure. As the sample thickness decreases, the relative contribution of demagnetizing energy to the total energy significantly increases, compelling the system to reduce the stripe domain period to lower the overall energy. Our experimental results are consistent with the trend predicted by Kittel's Law (Figure S13). The dependence of the stripe domain and skyrmion size on the lamella thickness under zero field is summarized in Figure S13, showing an increasing trend with increasing thickness, confirming the dipolar origin as the appearance of skyrmions in Mn₅Ge_{3.2} [56–58]. Figure 3(e) illustrates the generation of approximately 50-nm-sized skyrmions and the field-driven magnetic evolution in a 50-nm-thick lamella. Figure 3(b) and (d) show the magnetic phase diagrams of dipolar skyrmions as a function of temperature and magnetic field during the field-increasing process in 150- and 50-nm thick lamella, respectively. Dipolar skyrmions in Mn₅Ge_{3.2} magnet can sur-

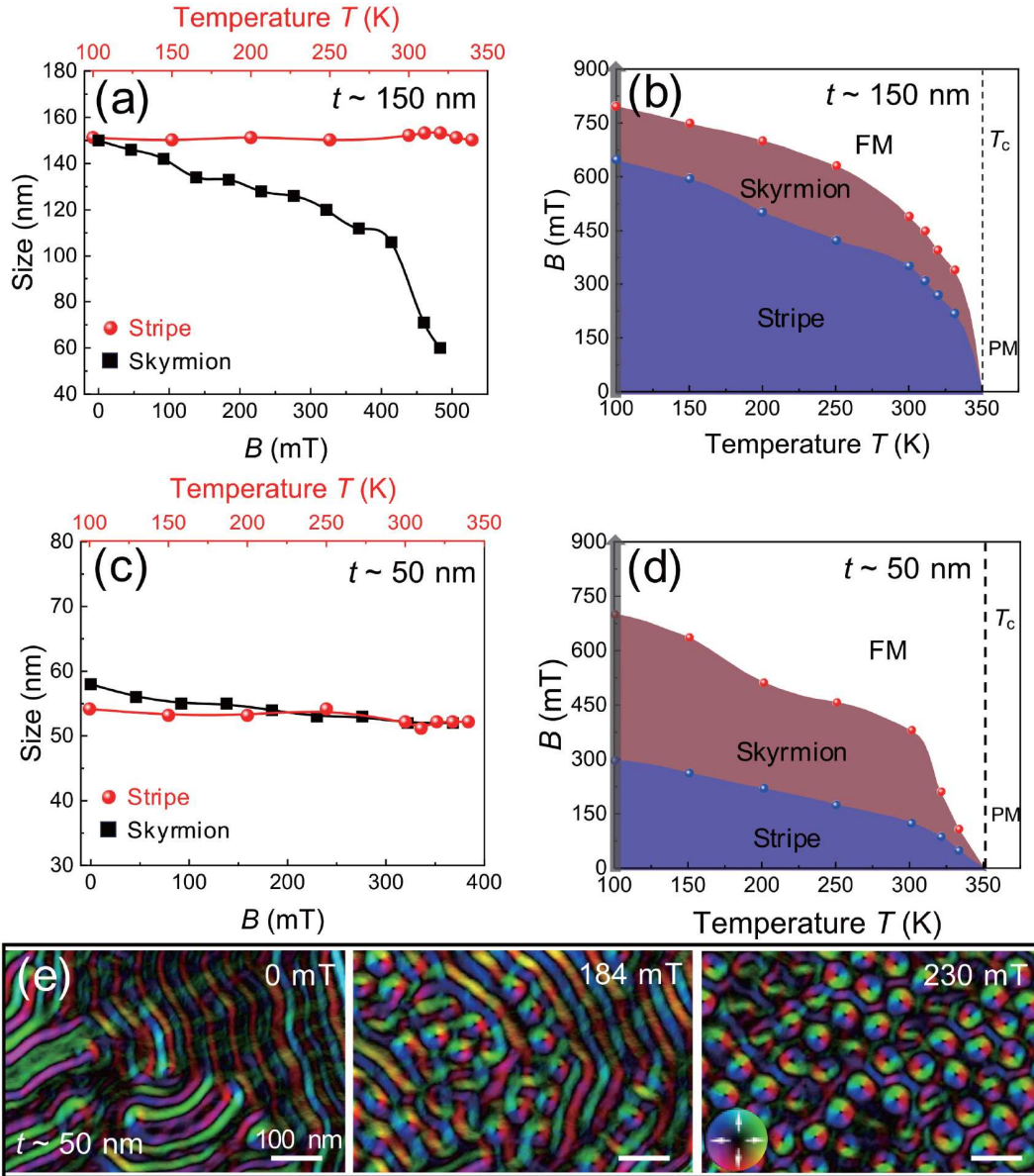


Figure 3 (Color online) Diameter of the dipolar skyrmion as a function of the field at room temperature (black line) and stripe period as a function of temperature T during zero-field warming (red line) in 150-nm (a) and 50-nm (c) thick $\text{Mn}_5\text{Ge}_{3.2}$ lamella. (b) and (d) Magnetic phase diagrams of 150- and 50-nm thick $\text{Mn}_5\text{Ge}_{3.2}$ plate as a function of temperature and magnetic field. (e) Field-driven magnetic evolutions in 50-nm-thick $\text{Mn}_5\text{Ge}_{3.2}$ lamella. The color wheel represents the in-plane magnetizations.

vive in a broad field temperature region under field-increasing.

3.3 Electrical detection of topological magnetic textures in $\text{Mn}_5\text{Ge}_{3.2}$

Generally, when the three-dimensional spatial dimensions of a crystal are reduced to a characteristic length scale within the crystal, the interaction between these dimensions and the crystal's characteristic length can lead to the emergence of new quantum phenomena [59]. We further explored the transport properties of $\text{Mn}_5\text{Ge}_{3.2}$ crystals at the mesoscopic

scale. As shown in Figure 4(a), using a FIB in conjunction with a custom-designed transfer technique, we fabricated microstructured nanostripe devices on intrinsic silicon substrates equipped with template electrodes (Figure S14). At low temperatures, the $\text{Mn}_5\text{Ge}_{3.2}$ magnet exhibits metallic conductivity properties, and anomalies were detected in the R - T curve (Figure 4(a)) around 350 K, where the temperature of resistance transition aligns well with the T_C point, confirming this point as the magnetic transition from the ferromagnetic to the paramagnetic state.

To gain deeper insights into the evolution of magnetic structures at the nanoscale, we conducted magnetoresistance

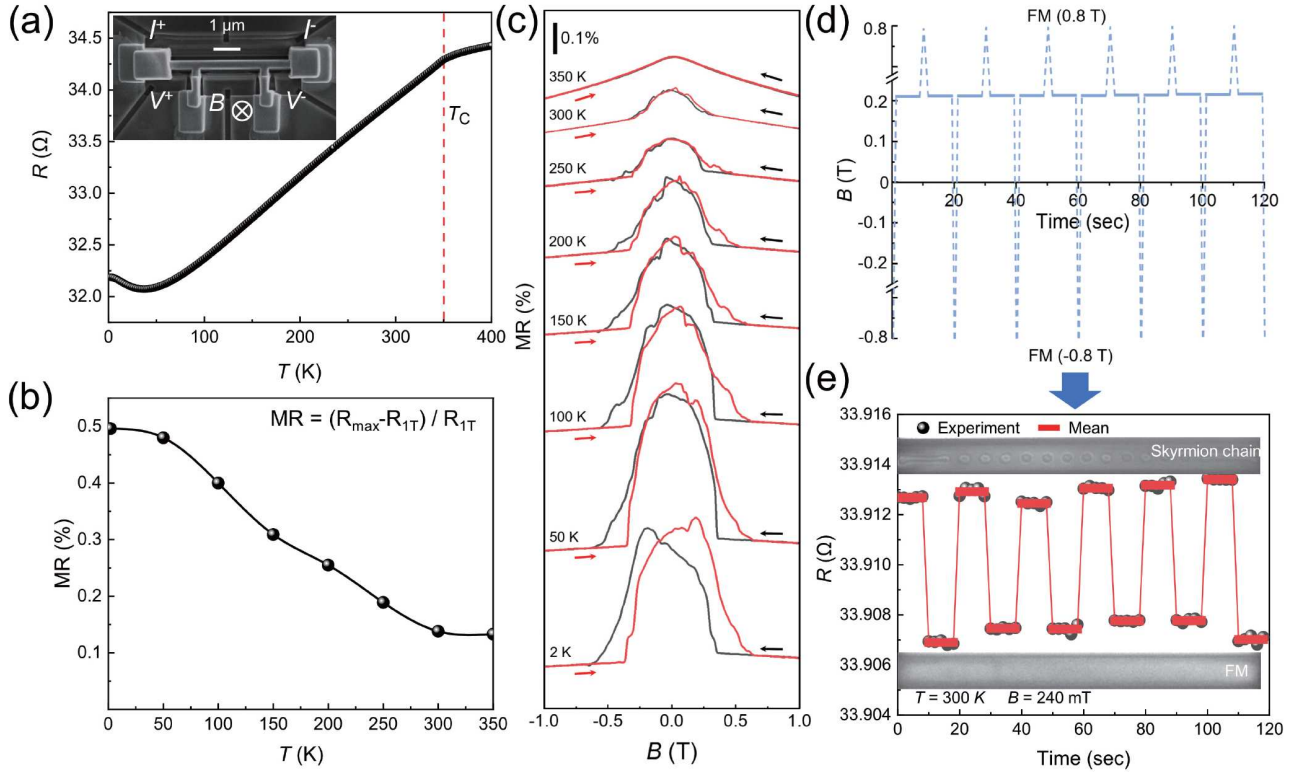


Figure 4 (Color online) Magnetotransport properties related to the magnetic texture evolution. (a) R - T curve. The inset is an experimental diagram of a typical four-terminal device. (b) The temperature dependence of the maximum magnetoresistance ($MR_{\max}$) at fixed temperatures. (c) MR as a function of the magnetic field at different temperatures. The magnetic field range sweeps from 1 to -1 T (black line) and back to 1 T (red line). The arrow denotes the direction of the applied magnetic field. (d) The protocol for applying the perpendicular magnetic field as a function of time. (e) The longitudinal resistance-time dependence is acquired according to the magnetic field application method described in (d). The inset in (e) shows the Lorentz Fresnel images of the corresponding magnetic states.

(MR) measurements at various temperatures with the magnetic field oriented such that $B//c$ (Figure 4(c)). The MR of ferromagnetic metals typically exhibits a symmetrical relationship with the applied magnetic field, i.e., $R_{xx}(H) = R_{xx}(-H)$. As shown in Figure 4(c), in the temperature range of 100–300 K, the raw MR displays asymmetric peaks. To further investigate this peculiar phenomenon, we applied a vertical magnetic field to the microdevice at 300 K following the method outlined in Figure 4(d). Specifically, we initially applied a fixed magnetic field of 0.24 T for approximately 8 s, followed by a rapid switch to -0.8 T, inducing a ferromagnetic state. We then quickly returned the field to 0.24 T for another 8 s before rapidly increasing it to 0.8 T and subsequently setting it back to 0.24 T, repeating this cycle. The resulting changes in resistance are shown in Figure 4(e), where the resistance variations exhibit a largely periodic relationship with the cycling magnetic field, and the corresponding magnetic configurations during this process were characterized using Lorentz-TEM, as shown in Figure 4(e). A more detailed correspondence between magnetotransport signals and magnetic structural states is presented in Figure S15.

The magnetoresistance changes can be attributed to distinct magnetic states [60]—specifically, a single skyrmion

chain and a uniform magnetization state—obtained under different field histories, as illustrated in Figure 4(e). A direct correlation between the measured transport properties and magnetic domain configurations is further demonstrated in Figure 5. Starting from the saturated magnetization state, reversing the magnetic field reveals stable uniform magnetization for $B > 305$ mT (Figure 5(c)). Horizontal stripe domains then emerge, which subsequently transform into zigzag-like stripe domains at zero fields. Upon further field reversal, a single skyrmion chain stabilizes across the magnetic field interval spanning from -365 to -485 mT, progressively transitioning to a ferromagnetic (FM) state at $B < -530$ mT. This magnetic evolution is reproducibly observed during the field-swapping process from -1 to 1 T. The magnetoresistance corresponding to these domain configurations (Figure 5(c)) is highlighted in Figure 5(a). To illustrate the electrical detection of topological domains, two representative states (labeled 2 and 17 in Figure 5(c)) are analyzed at a fixed field of 380 mT. State 2, representing the FM phase, is achieved by decreasing the field from 1000 to 380 mT. Conversely, State 17, corresponding to the single-skyrmion-chain configuration, is obtained by increasing the field from -1000 to 380 mT. The single-skyrmion-chain state exhibits a resistance 0.03 Ω higher than that of the FM

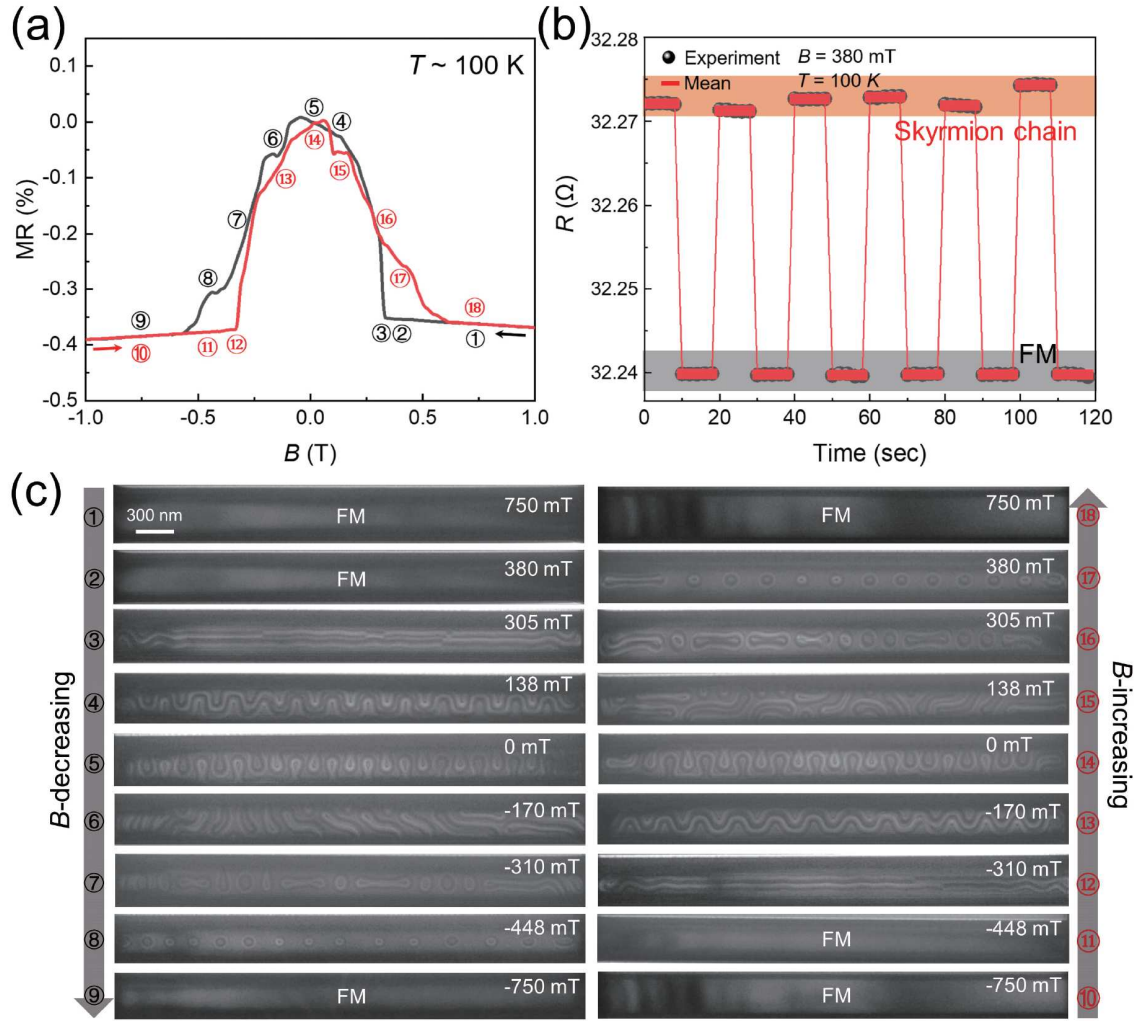


Figure 5 (Color online) (a) MR as a function of the magnetic field at 100 K. The magnetic field range sweeps from 1 to -1 T (black line) and back to 1 T (red line). The arrow denotes the direction of the applied magnetic field. (b) The longitudinal resistance-time dependence acquired according to the magnetic field application method described in Figure 4(d). (c) Lorentz Fresnel images depicting the domain structure states under the same conditions as those in panel (a), with the numbers indicating the correspondence.

state, confirming its distinct transport signature. Therefore, the changes in magnetic structure induced by the processes of increasing and decreasing the magnetic field are responsible for the non-reciprocal symmetry of the MR curve. Within the applied magnetic field range (± 1 T), the $MR_{\max}$ generally exhibits a decreasing trend with increasing temperature, as shown in Figure 4(b). The hysteresis of magnetoresistance also becomes more prominent at low temperatures (Figure 4(c)), suggesting a larger detection signal of topological domains at low temperatures. These provide a feasible approach for the subsequent electrical probing of topological magnetic structures in $Mn_5Ge_{3.2}$.

3.4 Electrical manipulation of a single dipolar skyrmion in $Mn_5Ge_{3.2}$ nanostructured cells

For skyrmion-based devices, achieving deterministic control

over skyrmions is a critical factor for their practical application [61–63]. A major challenge in the field of topological spintronics is the precise electrical manipulation of individual topological magnetic structures within strongly confined functional units at room temperature. In the previous section's magnetoresistance-field curves, we clearly observed hysteresis behavior, providing the prerequisite for creating and deleting skyrmions at a fixed field using current.

The fabrication of 50-nm-thick nanostructured devices suitable for electrical measurements (requiring intact electrodes) presents significant process challenges. Therefore, for this work, we fabricated a nanostructured microdevice with dimensions of ~ 250 nm in width and 150 nm in thickness, which is presented in Figure 6(a). Initially selecting the FM state as the starting point, we explored the evolution of magnetic states under pulsed currents with a pulse width of 5 ns at $B = 350$ mT (Figure 6(b)). At low current densities,

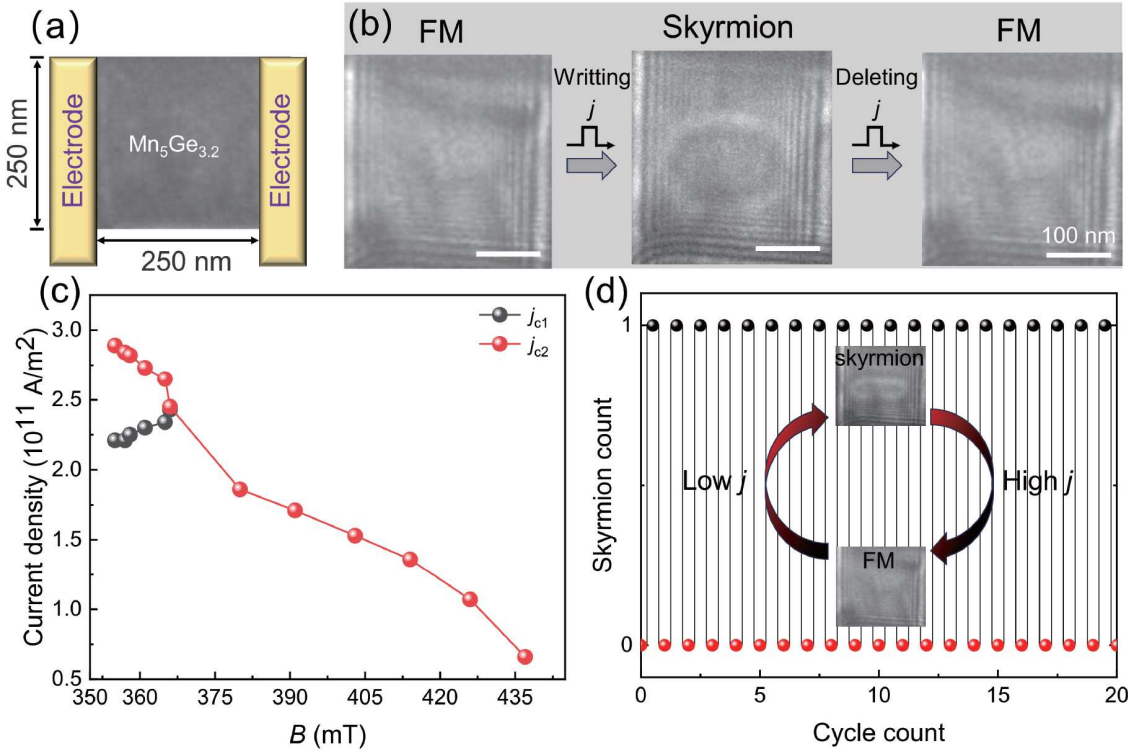


Figure 6 (Color online) Current-induced creation and deletion of a single skyrmion at room temperature. (a) Schematic diagram of the Mn₅Ge_{3.2} nanostructured cell sample with a (001) plane. (b) Defocused images of different magnetic states in the nanostructured cell after applying pulsed currents. The defocused distance is $-300\ \mu\text{m}$. (c) Dependence of critical current densities j_{c1} and j_{c2} on magnetic field B at a fixed pulse width, $w = 5\ \text{ns}$. (d) Cycle count dependence of skyrmion creation and deletion at $B = 350\ \text{mT}$. Each cycle count includes a 5-ns low current pulse with a density of $\approx 2.23 \times 10^{11}\ \text{A m}^{-2}$ and a subsequent 5-ns high current pulse with a density of $\approx 2.82 \times 10^{11}\ \text{A m}^{-2}$.

the sample remains in the FM state until reaching $2.23 \times 10^{11}\ \text{A m}^{-2}$, after which a skyrmion appears due to the spin-transfer torque (STT) effect induced by the current. With further increases in current density, the skyrmion persists until transitioning back to the FM state at $2.82 \times 10^{11}\ \text{A m}^{-2}$. This distinct behavior during the transformation of magnetic states under various pulsed currents allows for the unambiguous identification of two critical current densities, j_{c1} and j_{c2} . Here, j_{c1} marks the lower critical current density for the transition from the FM state to the skyrmion state, while j_{c2} indicates the higher critical current density for the transition from the skyrmion state back to the FM state. Therefore, the creation and deletion of a single skyrmion can be controlled by applying pulsed currents to the nanostructured cells, encompassing two significant transition processes—from the FM state to the skyrmion state and *vice versa*.

The deterministic electrical control of single skyrmions is attributed to magnetic domain hysteresis (Figure S16) and combined current-induced effects. In the low-field regions of magnetic domain hysteresis, the skyrmion state represents a more stable phase compared to the uniform magnetization state (Figure S17(b)). The current-induced spin-transfer torque effect serves as a stimulus to trigger the transition from the metastable FM state to the skyrmion state (Figure S17(c)). However, when current density further increases,

the Joule thermal heating effect becomes dominant. Our results show the thermal cycle from 300 to 320 K and then back to 300 K leads to the skyrmion-to-FM transformation (Figure S17(d) and (e)).

The critical current densities j_{c1} (for writing) and j_{c2} (for deleting) are highly sensitive to the magnetic field B , as shown in Figure 6(c). When the magnetic field exceeds 365 mT, j_{c1} may surpass j_{c2} , leading to sufficient Joule heating that reduces the saturation magnetization and stabilizes the FM state, making it impossible to transform the FM state into dipolar skyrmions using pulsed currents. On the other hand, with the increasing magnetic field, the stability of the FM state is enhanced for j_{c2} , requiring less reduction in saturation magnetization, which results in a decrease in j_{c2} , as shown in Figure 6(c). Based on the transition process between the FM and skyrmion states, we applied a pulse with a width w of 5 ns current to the microdevice and selected two discrete current densities to establish a write-delete cycle for the nanostructured unit. Under a pulse width of 5 ns, a current density of $2.23 \times 10^{11}\ \text{A m}^{-2}$ was chosen for writing a single skyrmion from the FM state, while a current density of $2.82 \times 10^{11}\ \text{A m}^{-2}$ was selected for reverse conversion. By alternately applying these two pulsed currents to the sample, we achieved deterministic cycles of writing and deleting (Figure 6(d); Supplementary Video 1). The successful electrical

manipulation of skyrmions at room temperature highlights the potential of $\text{Mn}_5\text{Ge}_{3.2}$ materials for applications in spintronic devices.

4 Conclusion

In summary, we successfully elevated the T_C of $\text{Mn}_5\text{Ge}_{3.2}$ using a HP synthesis method, achieving a ferromagnetic transition above room temperature (≈ 350 K). We obtain stable dipolar skyrmion at room temperature in $\text{Mn}_5\text{Ge}_{3.2}$ lamella, achieving zero-field stabilized sizes as small as ≈ 50 nm. Furthermore, we successfully show the deterministic electrical creation and deletion of individual skyrmions in confined $\text{Mn}_5\text{Ge}_{3.2}$ unit (size ≈ 250 nm $\times$ 250 nm) using single nanosecond-scale pulsed currents. Finally, we propose that single skyrmion chains in confined nanostripes can be detected via magnetoresistance measurements. These results not only establish $\text{Mn}_5\text{Ge}_{3.2}$ as a promising material for topological magnetic studies but also open up new avenues for developing spintronic devices with enhanced functionalities operating at ambient conditions. Future work will focus on fabrication of ultrathin $\text{Mn}_5\text{Ge}_{3.2}$ films and the construction of corresponding nano-devices via advanced lithography techniques, aiming to acquire more direct experimental evidence.

This work was supported by the National Key Research and Development Program of China (Grant No. 2024YFA1611303), the Scientific Research Innovation Capability Support Project for Young Faculty (Grant No. SRICSPYF-BS2025073), the National Natural Science Foundation of China (Grant Nos. 521300103, 52325105, 12422403, U24A6001, 52501305, 12174396, 12104123, 12204006, and 12241406), the Anhui Provincial Natural Science Foundation (Grant Nos. 2308085Y32, 2408085QA022, and 2508085MA018), the Natural Science Foundation of Jiangsu Province of China (Grant No. BK20220303), the Natural Science Project of Colleges and Universities in Anhui Province (Grant Nos. 2022AH030011 and 2024AH030046), the Anhui Province Excellent Young Teacher Training Project (Grant No. YQZD2023067), the 2024 Project of GDRCCY No. 217, the China Postdoctoral Science Foundation (Grant Nos. 2023M743543, and 2024M760006), the Postdoctoral Fellowship Program of CPSF (Grant No. GZB20250792), the China Postdoctoral Science Foundation—Anhui Joint Support Program (Grant No. 2025T003AH), and the Anhui Postdoctoral Scientific Research Program Foundation (Grant No. 2025B1080).

Conflict of interest The authors declare that they have no conflict of interest.

Supporting Information

The supporting information is available online at <http://phys.scichina.com> and <https://link.springer.com>. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.

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