

Emergent Skyrmions in Cr_{0.85}Te nanoflakes at Room Temperature

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Chiral noncollinear magnetic nanostructures, such as skyrmions, are intriguing spin configurations with significant potential for magnetic memory technologies. However, the limited availability of 2D magnetic materials that host skyrmions with Curie temperatures above room temperature presents a major challenge for practical implementation. Chromium tellurides exhibit diverse spin configurations and remarkable stability under ambient conditions, making them a promising platform for fundamental spin physics research and the development of innovative 2D spintronic devices. Here, domain structures of Cr_{0.85}Te nanoflakes synthesized via chemical vapor deposition are investigated, using magnetic force microscopy at room temperature. The results reveal that the domain width of the as-grown nanoflakes scales with the square root of their thicknesses. Notably, the emergence and annihilation of skyrmions are observed, which can be reversibly controlled by external magnetic fields and thermal excitation in ambient air. Micromagnetic simulations suggest that the emergence of skyrmions in Cr_{0.85}Te nanoflakes arises from inversion symmetry breaking due to compositional gradients across the sample thickness, rather than the interfacial Dzyaloshinskii–Moriya interaction. These findings provide new insights into the mechanisms underlying skyrmion formation in 2D ferromagnets and open exciting possibilities for manipulating domain structures at room temperature, offering practical pathways for developing next-generation spintronic devices.

1. Introduction

Understanding the behavior of magnetic domains under external stimuli such as magnetic fields,^[1] electric currents,^[2] temperature,^[3] and mechanical stress^[4] is essential for gaining insights into macroscopic magnetic properties in both fundamental spin physics research and potential spintronics applications. Magnetic domain structure, domain wall motion, and topological nanostructures such as skyrmions hold significant promise for applications in memory devices.^[5] Skyrmions are particularly notable as topologically protected magnetic structures with a characteristic whirlpool-like configuration, as reported in numerous studies.^[6] Skyrmions were first observed in chiral magnet MnSi,^[7] which were stabilized by the Dzyaloshinskii–Moriya interaction (DMI)—an asymmetric exchange interaction between neighboring spins. DMI arises from the combination of broken inversion symmetry and strong spin–orbit coupling (SOC), and it can be generated in various systems, including

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chiral magnets,^[8] polar magnets,^[9] thin films with asymmetric interfaces^[10] or component gradients.^[11] In contrast to the DMI-induced skyrmions, magnetic bubbles can form in centrosymmetric materials, such as $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ ^[12] and MnNiGa ,^[13] where inversion symmetry breaking is not necessary. In one type of bubble (type-I), the cylindrical domain walls consist of Bloch walls, with magnetic moments exhibiting either clockwise or anticlockwise helicity. These bubbles share an integer topological winding number with skyrmions and are, therefore, referred to as skyrmion bubbles. In contrast, another type of bubble (type-II) features internal domain walls with reversed circulation, rendering them topologically trivial.^[14] The formation of magnetic bubbles typically involves the interplay of magnetic anisotropy, exchange interactions, and external magnetic fields.^[15]

In recent years, two-dimensional (2D) magnetic materials, $\text{Cr}_2\text{Ge}_2\text{Te}_6$,^[16] Fe_nGeTe_2 ($n = 3, 5$),^[17] $(\text{Fe}_{0.5}\text{Co}_{0.5})_5\text{GeTe}_2$,^[18] Fe_3GaTe_2 ,^[19] and $\text{Cr}_{1+\delta}\text{Te}_2$ ^[20] have gained significant attentions as promising host for skyrmions. Investigating skyrmions in 2D magnets offers unique opportunities to explore nanoscale control of magnetic domains, paving the way for advancements in spintronic devices. However, the long-range magnetic order in most 2D magnets is typically stabilized below room temperature.^[21] A limited number of 2D magnets exhibit both skyrmions and intrinsic Curie temperature (T_c) above room temperature, including $(\text{Fe}_{0.5}\text{Co}_{0.5})_5\text{GeTe}_2$,^[18] Fe_3GaTe_2 ,^[19a,b] and $\text{Cr}_{0.87}\text{Te}_2$.^[20a] Among these, the binary compound chromium telluride ($\text{Cr}_{1+\delta}\text{Te}_2$) stands out due to its chemical stability under ambient conditions, continuously adjustable composition, and tunable T_c achieved by varying the Cr content. This family of materials has CrTe_2 as the layered host, allowing for different Cr compositions through the insertion of Cr ions into the van der Waals (vdW) gaps.^[22] The T_c of $\text{Cr}_{1+\delta}\text{Te}_2$ can be tuned from 160 to 350 K as δ varies between 0.33 and 0.82.^[20c,23] Additionally, modifications in the Cr and Te composition can adjust both the magnetic anisotropy and the T_c . The magnetic anisotropy of $\text{Cr}_{1+\delta}\text{Te}_2$ also varies with thickness and fabrication method. For instance, bulk $\text{Cr}_{0.92}\text{Te}_2$ single crystal shows an in-plane magnetic anisotropy.^[24] Similarly, the exfoliated CrTe_2 nanoflakes^[25] also exhibit an in-plane magnetic anisotropy resulting from both shape and magnetocrystalline anisotropies. However, epitaxial CrTe_2 films^[22b] demonstrate strong perpendicular magnetic anisotropy (PMA) likely caused by surface effects and strain. These observations highlight the tunable magnetic properties of the $\text{Cr}_{1+\delta}\text{Te}_2$ family, including the possibility of smooth transitions between out-of-plane (OP) and in-plane (IP) magnetic configurations.^[23]

Notably, skyrmions have been observed in bulk $\text{Cr}_{0.87}\text{Te}_2$ at room temperature (298 K), resulting from a competition between magnetic dipole interactions and uniaxial easy-axis anisotropy.^[20a] However, the enhanced interlayer bonding be-

cause of additional Cr ions in $\text{Cr}_{1+\delta}\text{Te}_2$ bulk crystals makes it difficult to exfoliate into thin layers or nanoflakes.^[26] At the nanoscale, bi-skyrmions were captured in Cr_7Te_4 nanoflakes synthesized via the chemical vapor deposition (CVD) method, but these structures only survive at low temperatures (92 K).^[20d] The topological hall effect (THE), which enables electrical detection of chiral spin textures such as skyrmions, has been observed in epitaxial Cr_5Te_6 thin films up to 270 K. This phenomenon is attributed to the noncoplanar spin textures from the interaction of the in-plane ferromagnet and antiferromagnet infrastructures under OP magnetic field.^[20e] It thus raises the critical question of whether the nanoscale thickness and the T_c above room temperature are compatible in $\text{Cr}_{1+\delta}\text{Te}_2$. Although the creation of skyrmions in $\text{Cr}_{1+\delta}\text{Te}_2$ nanoflakes at room temperature remains challenging, progress has been made in observing skyrmions in more complex ternary tellurides, such as $(\text{Fe}_{0.5}\text{Co}_{0.5})_5\text{GeTe}_2$,^[18] Fe_3GaTe_2 ,^[19] and Fe_5GeTe_2 .^[27]

Here, we successfully grew 2D $\text{Cr}_{0.85}\text{Te}_2$ nanoflakes using the CVD method, showing clear magnetic domain patterns above room temperature. The as-grown nanoflakes demonstrated a thickness-dependent stripe magnetic structure at ambient conditions, consistent with the Kittel model, where the domain width scales with the square root of the nanoflake thickness. More importantly, we achieved a reversible transformation of magnetic domains from stripe patterns to localized skyrmionic structures, modulated by an external magnetic field. We propose that these room-temperature skyrmions are likely induced by bulk DMI, arising from the compositional gradient across the thickness of $\text{Cr}_{0.85}\text{Te}_2$ nanoflakes, as supported by our micromagnetic simulation results. Furthermore, we examined the effects of thermal excitation on the magnetic structure, revealing a T_c of ≈ 333 K for the $\text{Cr}_{0.85}\text{Te}_2$ nanoflakes. Skyrmion generation was also observed, facilitated by thermal perturbation and stray fields from the magnetic probe. These findings provide valuable insights into the practical applications of 2D ferromagnetic nanoflakes as platforms for next-generation spintronic devices, demonstrating their promise for practical applications in emergent spintronic technologies.

2. Results and Discussion

2.1. Compositional Characterization of $\text{Cr}_{0.85}\text{Te}_2$ Nanoflakes

$\text{Cr}_{0.85}\text{Te}_2$ nanoflakes were synthesized on SiO_2/Si substrates using the atmospheric pressure chemical vapor deposition (APCVD) method (detailed in Section 4). The nanoflakes were subsequently transferred to new substrates using a sugar transfer method for further characterization.^[28] A schematic diagram of the sugar transfer method is shown in Figure 1a. In this process, the caramelized sugar was boiled and dispersed onto the surface of the substrate. After cooling, the sugar was peeled off, carrying the sample adhered to its surface. The sugar with the sample was then placed on a new substrate, heated to ensure full contact, and allowed to cool before being washed with deionized water, leaving the sample on the new substrate. Using this method, $\text{Cr}_{0.85}\text{Te}_2$ nanoflakes were successfully transferred onto new SiO_2/Si and Au substrate (Figure S1, Supporting Information). A cross-sectional TEM sample was prepared via the focused ion beam (FIB) technique, cut parallel to the crystal edges

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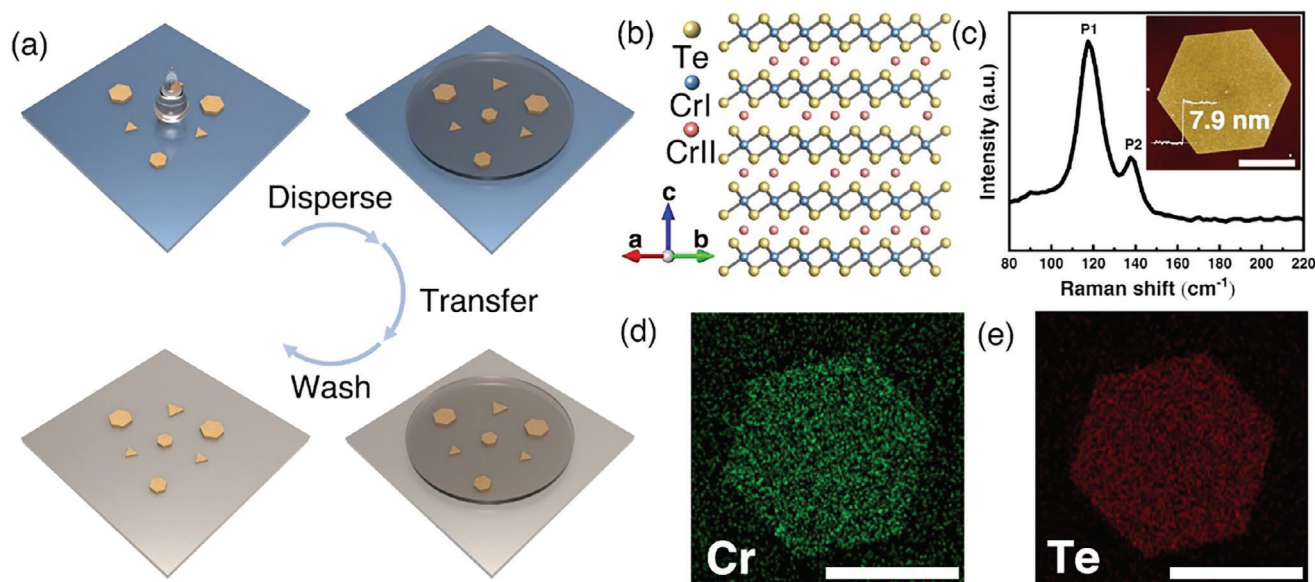


Figure 1. Transfer process and basic characterizations of $\text{Cr}_{0.85}\text{Te}$ nanoflakes. a) Schematic diagram of the sugar transfer method. b) Cross-sectional schematic of the $\text{Cr}_{0.85}\text{Te}$ crystal structure. c) Raman spectrum of $\text{Cr}_{0.85}\text{Te}$ nanoflakes synthesized on SiO_2/Si substrate. The inset displays an AFM image along with the corresponding height profile of a $\text{Cr}_{0.85}\text{Te}$ nanoflake. Scale bar: 5 μm . d,e) Elemental mapping of Cr element (d) and Te element. (e) Scale bar: 10 μm .

of the hexagonal nanoflake. Selected area electron diffraction (SAED) was conducted on three regions, near the surface of the nanoflake, in the middle, and near the substrate (Figure S2, Supporting Information). The diffraction spot from these regions demonstrated that the entire sample retains a hexagonal structure with no evidence of secondary phases. The reported CrTe crystal^[29] belongs to the hexagonal $P6_3/mmc$ space group and the SAED zone axis was determined to be $[\bar{1}10]$, based on the measured crystal plane spacing and indexed diffraction spots. As shown in Figure 1b, $\text{Cr}_{0.85}\text{Te}$ exhibits a self-intercalated crystal structure, where Cr atoms intercalate into the vdW gap between the CrTe_2 layers.^[26b] The Raman spectrum of the $\text{Cr}_{0.85}\text{Te}$ nanoflake (Figure 1c) shows two prominent characteristic peaks at 117 and 137 cm^{-1} , corresponding to A_{1g} and E_g vibration modes, respectively, consistent with the features previously reported for the $\text{Cr}_{1+\delta}\text{Te}_2$ system.^[29,30] The inset of Figure 1c presents an atomic force microscopy (AFM) morphology image of a hexagonal nanoflake with a thickness of 7.9 nm. Elemental analysis of the nanoflakes revealed average atomic percentages of 45.9% Cr and 54.1% Te (Table S1, Supporting Information), indicating an approximate Cr:Te atomic ratio of 0.85:1. Elemental mappings of Cr and Te (Figure 1d,e) display a uniform color distribution, confirming compositional homogeneity within the hexagonal nanoflake.

2.2. Thickness-Dependent Magnetic Domain Width of $\text{Cr}_{0.85}\text{Te}$ Nanoflakes

To investigate the magnetic properties of the $\text{Cr}_{0.85}\text{Te}$ nanoflakes, magnetic force microscopy (MFM) was employed to characterize their magnetic domain structures at room temperature under ambient conditions. Figure 2a–d shows the MFM images of

$\text{Cr}_{0.85}\text{Te}$ nanoflakes with different thicknesses. During the MFM measurements, the probe captured OP magnetic signals. Magnetic contrast was determined by the frequency shift relative to a reference value. A negative frequency shift indicates attraction between the probe tip and the sample, while a positive frequency shift indicates repulsion. As a result, two distinct contrasts were observed: red (blue) regions represent the magnetization direction of the sample parallel (antiparallel) to that of the magnetic probe. Since the magnetic orientation of the probe remained constant throughout the measurements, the red and blue contrasts correspond to magnetic domain structures with two opposite magnetization directions. In Figure 2a–d, the alternating red and blue stripe patterns indicate the presence of stripe domain structures. Along the stripe direction, magnetic moments are aligned, with periodic alternation between the upward and downward arrangement perpendicular to the stripe direction. The stripe magnetic domain pattern is formed due to the interplay of three types of interactions, including short-range ferromagnetic exchange coupling, long-range dipolar interaction, and uniaxial anisotropy.^[31] The dipolar interactions favor antiparallel spin arrangement and formation of stripe domains,^[32] while the domain width reflects the strength of perpendicular magnetic anisotropy (PMA).^[33] Figure 2e shows the MFM data along the white dotted lines in Figure 2a–d, revealing different periodicities of domain width, which increases with the nanoflake thickness monotonically. Additional domain pattern measurements on $\text{Cr}_{0.85}\text{Te}$ nanoflakes of various thicknesses are presented in Figure S3 (Supporting Information), and the relationship between domain width and nanoflake thickness is summarized in Figure 2f. The overall effective domain width was calculated as the average of all red and blue stripe domains. As the thickness of nanoflakes increased from 47.6 to 469.5 nm, the stripe domain width expanded from 74.3 to 243.4 nm. Following the Kittel

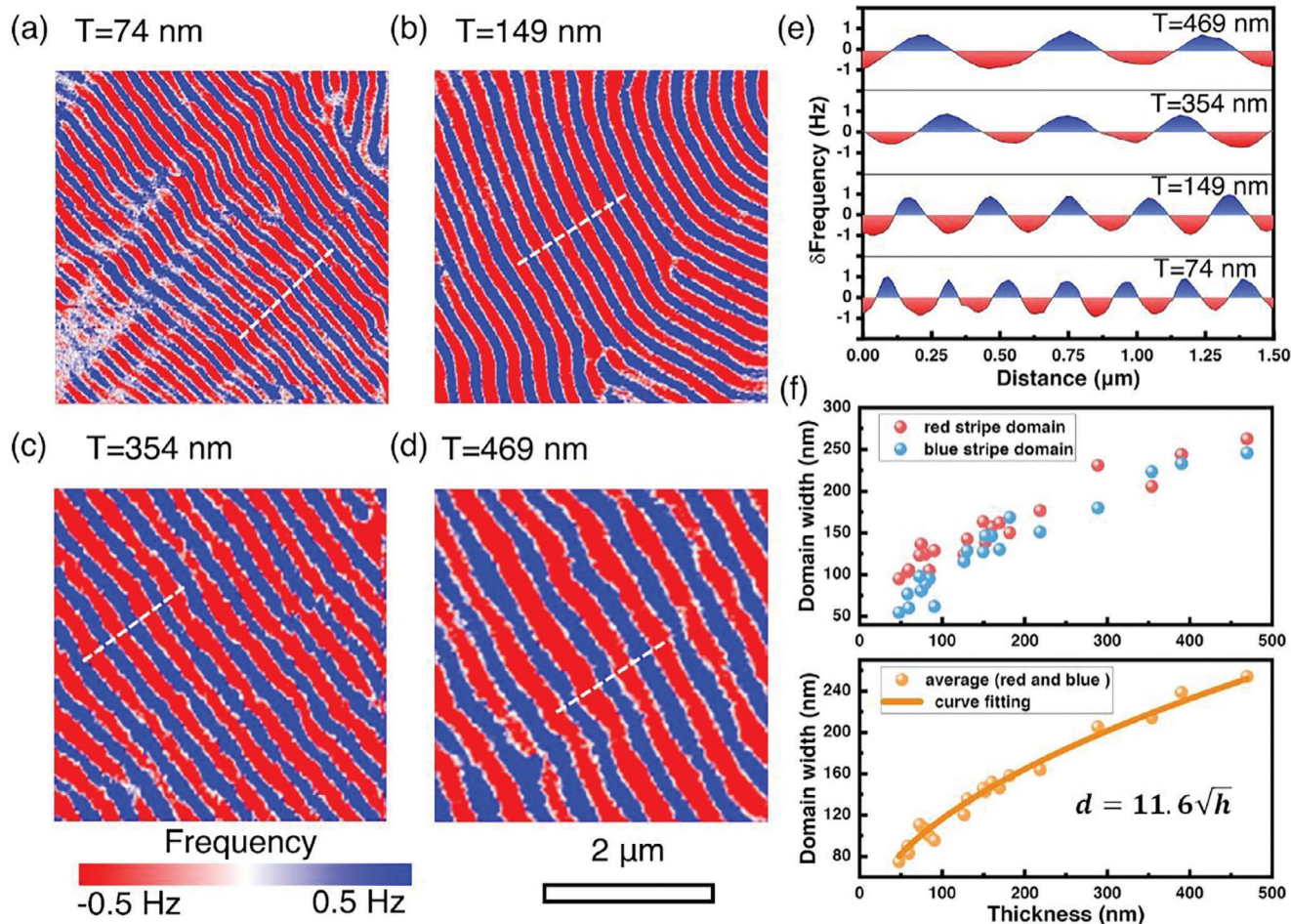


Figure 2. Stripe magnetic domain patterns of $\text{Cr}_{0.85}\text{Te}$ nanoflakes of varying thickness. a–d) MFM images of $\text{Cr}_{0.85}\text{Te}$ nanoflakes of different thicknesses (T) at room temperature. e) Profiles of stripe domains along the white lines in (a–d), revealing the domain width of $\text{Cr}_{0.85}\text{Te}$ nanoflakes. f) Domain width of the stripe magnetic domains as a function of the thickness of $\text{Cr}_{0.85}\text{Te}$ nanoflakes. The red (blue) spheres represent the width of the red (blue) stripe domains, while the yellow spheres denote the average stripe domain width for specific nanoflake thicknesses.

model,^[34] the domain width (d) can be fitted using the following formula:

$$d = 3.84\sqrt{Qh} \quad (1)$$

where Q is the quality factor, and h denotes the thickness of the nanoflake. The experimental data closely align with the fitted curve described by a power function, expressed as:

$$d = 11.6\sqrt{h} \quad (2)$$

From the fit, the Q value was determined to be 9.13. The lower thickness limit for nanoflakes exhibiting stripe domains is ≈ 47.6 nm, as shown in Figure S3 (Supporting Information). When the thickness is further reduced, samples with stripe domains become increasingly difficult to observe. Additional experiments were conducted to analyze the MFM response of samples with thickness down to about 20 nm to directly probe the possible crossover thickness. For example, in a nanoflake with a thickness of ≈ 19.5 nm, the topography and corresponding MFM image are presented in Figure S9 (Supporting Information). The absence

of a clear stripe domain structure in the MFM image suggests that the crossover to in-plane magnetization may occur between 47.6 and 19.5 nm. Generally, magnetocrystalline anisotropy is an intrinsic property of a material and is not significantly affected by thickness. However, in our samples, surface and interface effects appear to influence the overall magnetic anisotropy. The thickness-dependent magnetocrystalline anisotropy is closely associated with the changes in the magnetic easy axis as the thickness decreases.

2.3. Magnetic Domain Evolution Under External Magnetic Field

Stripe domains can be regarded as precursors to skyrmions as they can transform into skyrmionic structures under an external magnetic field, as reported in previous studies.^[35] The evolution of the stripe domains under an OP magnetic field was investigated, and the corresponding MFM images of the $\text{Cr}_{0.85}\text{Te}$ nanoflake under various OP magnetic fields are shown in Figures 3a–i and S4 (Supporting Information). The symbols “+” and “−” indicate the direction of the magnetic field, with

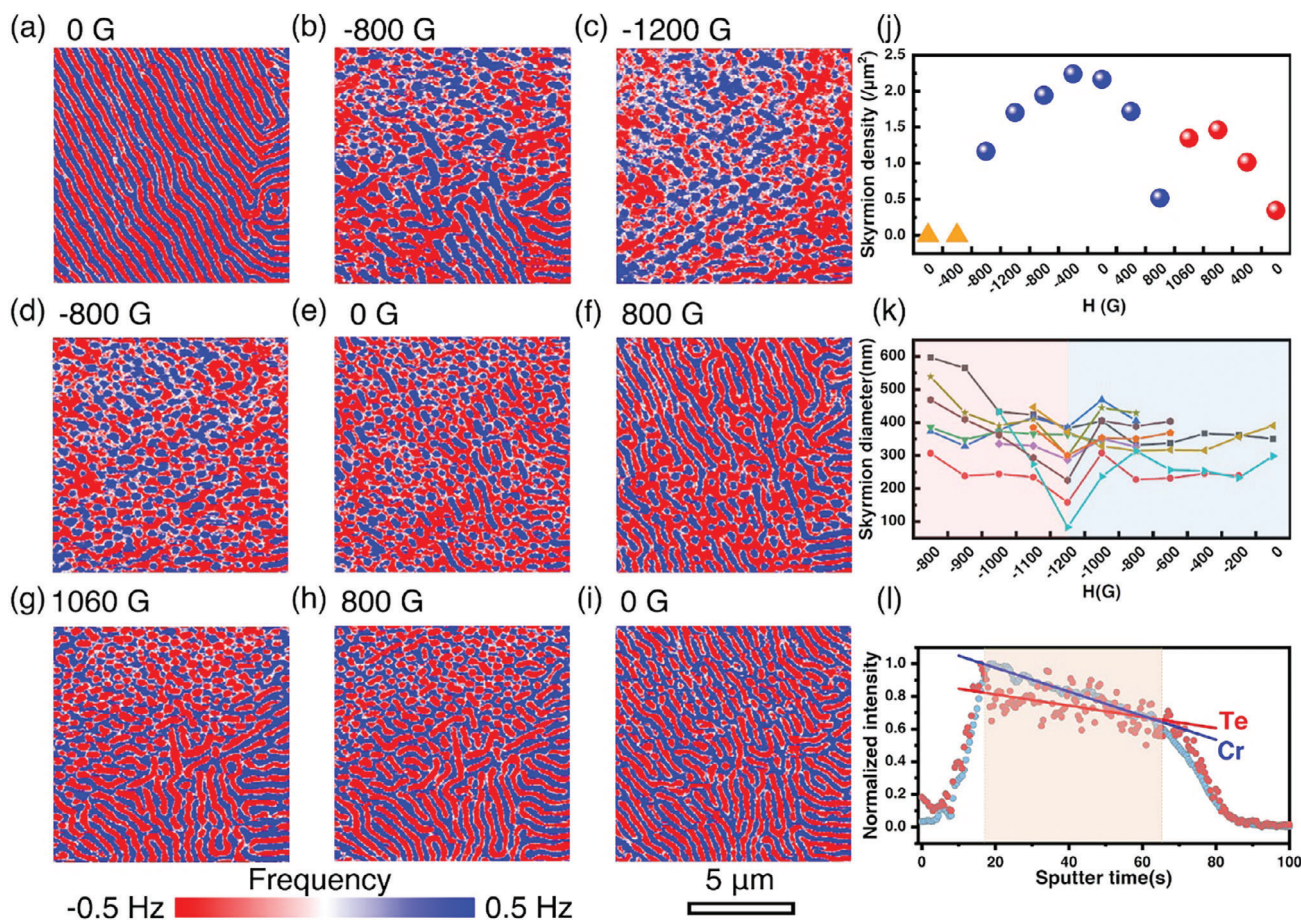


Figure 3. Magnetic domain evolution of a 469.5-nm-thick $\text{Cr}_{0.85}\text{Te}$ nanoflake under a continuously varying magnetic field at room temperature and ambient conditions. a–i) MFM images captured under different magnetic fields. j,k) Dependence of skyrmionic structure density (j) and diameter (k) on the magnetic field. l) ToF-SIMS depth profile (normalized to maximum) of an 81-nm-thick $\text{Cr}_{0.85}\text{Te}$ nanoflake.

“+” (“−”) representing a field pointing upwards (downwards) (perpendicular to the nanoflake). In the initial state, stripe domains were observed in the nanoflakes. When a negative external magnetic field (“−”) was applied, the red domain regions expanded while the blue stripe domains shrunk. Here, the “−” symbol corresponds to the application of a magnetic field parallel to the sample magnetization, aligning with the red regions, while the “+” symbol indicates a field antiparallel to the sample magnetization, aligning with the blue regions. As the external magnetic field increased to −800 G, some stripe domains began to transform into localized skyrmionic structures, as observed through in situ MFM imaging. The “−” magnetic field further expanded the red domains, causing the blue stripe domains to contract and break into smaller sections before eventually condensing into skyrmions.^[36] At −1200 G, nearly all the stripe domains had transformed into skyrmionic structures. When the magnetic field was reduced back to 0 G, many skyrmions remained stable, while others reverted to stripe domains. Reversing the magnetic field direction to +800 G caused most of the remaining skyrmions to transition back to the stripe domains. As the field strength increased to +1060 G, part of the scanned region (Figure 3g) exhibited the transformation of stripe domains into red skyrmions, while other areas showed blue skyrmions re-

verting to stripe domains. Reducing the magnetic field to +800 G stabilized the red skyrmionic structures, and further decreasing the field to 0 G led to most skyrmionic structures reverting to stripe domains. Throughout the measurement process, the applied magnetic field should not reverse the magnetization direction of the probe tip due to its coercive field being greater than 5000 Oe. Consequently, the red and blue contrasts consistently represent opposite magnetization directions of the magnetic domains. These results demonstrate that as the external magnetic field increases, stripe domains can disperse and evolve into skyrmionic structures while decreasing the field can lead to the coalescence of skyrmions back into stripe domains. This transition is reversible as the magnetic field cycles. Ideally, with a continuous increase in external magnetic field, the magnetic phase order should progress from stripe domains to skyrmionic structures and finally to a uniform single-domain phase, involving two phase transitions.^[37] However, the single domain phase was not observed in this study due to the limitations in the magnetic field range provided by the variable field module integrated with the MFM for in situ measurement.

The density of skyrmionic structures at different magnetic fields was quantified as shown in Figure 3j, to examine their variation with field strength. The red and blue dots in the

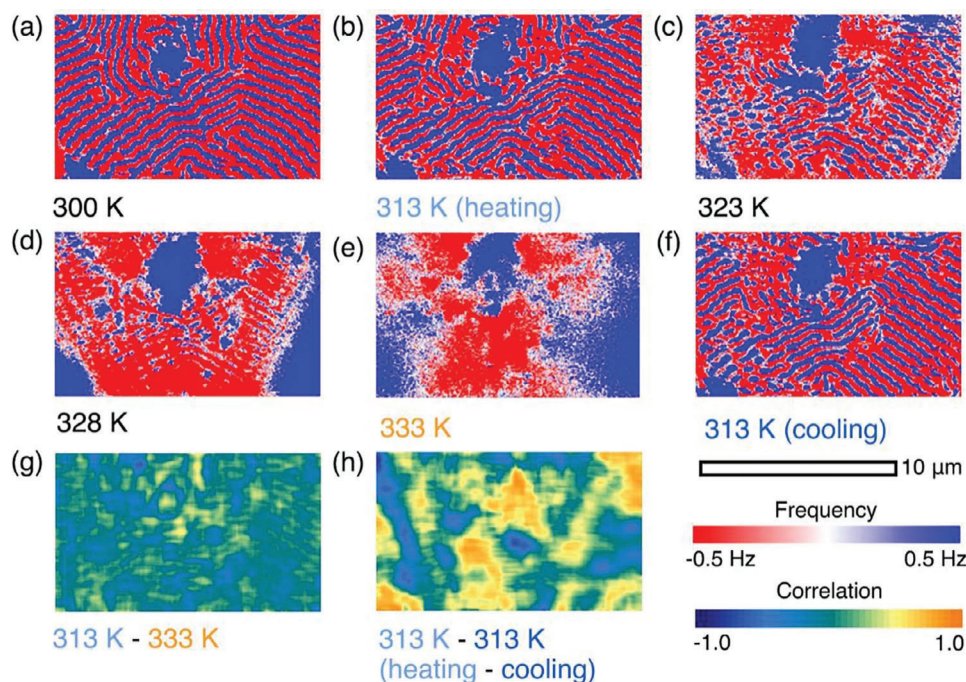


Figure 4. a–f) MFM images of a 257-nm-thick $\text{Cr}_{0.85}\text{Te}$ nanoflake at different temperatures. g) Correlation analysis image for comparison between (b) and (e). h) Correlation analysis image for comparison between (b) and (f).

figure represent skyrmionic structures with opposite spin configurations. Additionally, the field-dependent diameter of 10 representative skyrmionic structures with circular domains (that do not overlap during the field change) was analyzed, as illustrated in Figure 3k. As the magnetic field strength increased to 1200 G, the number of skyrmionic structures increased, accompanied by a decrease in their size. When the field strength was subsequently decreased from 1200 to 400 G (corresponding MFM images are provided in Figure S4, Supporting Information), the density of skyrmionic structures continued to increase, albeit their size also showed a trend of gradual growth. These behaviors are consistent with observations of skyrmionics structures in other studies.^[20b,38] As the magnetic field increased to +800 G in the opposite direction, the area of the blue magnetic domains aligned with the field direction expanded, causing some blue skyrmionic structures to connect and form stripe domains. This transition resulted in a reduced density of skyrmionic structures. Further increasing the magnetic field to +1060 G led to the emergence of red skyrmionic structures with spin configurations opposite to those of the blue skyrmionic structures.

According to Figure 3k, the average size of the skyrmionic structures under varying magnetic fields ranged from 200 to 400 nm, consistent with the size of skyrmions observed in the $\text{Cr}_{1+\delta}\text{Te}_2$ system.^[20] These skyrmionic structures arrange themselves in quasi-hexagonal lattices (Figure 3d,h), further supporting the identification of the observed skyrmionic structures in $\text{Cr}_{0.85}\text{Te}$ nanoflakes as skyrmions. To investigate the origin of these skyrmionic structures, a depth profile analysis of the nanoflake was conducted using a time-of-flight secondary ion mass spectrometer (ToF-SIMS), as shown in Figure 3l. The intensities profiles of Cr and Te revealed different rates of decline with

increasing sputtering time, indicating a variation in the composition along the depth direction of the nanoflake. DMI is intrinsically linked to the breaking of spatial inversion symmetry.^[39] The observed compositional gradient in the depth profile can induce broken inversion symmetry, leading to the emergence of bulk-DMI.^[11] Therefore, the skyrmionic structures formed under magnetic field modulation are expected to be skyrmions rather than magnetic bubbles.

2.4. Magnetic Domain Evolution Under Thermal Excitation

The magnetic domain structure of the $\text{Cr}_{0.85}\text{Te}$ nanoflakes at elevated temperatures was investigated using in situ MFM (Figure 4). The stripe domains remained stable as the temperature increased from 300 to 313 K. When the temperature was further increased to 323 K, some stripe domains transformed into short-domain segments or skyrmions, rather than exhibiting changes in stripe width. This observation suggests that the PMA is not significantly affected by thermal agitation. In the absence of an external magnetic field, the skyrmion phase can be metastable. The long-range order of magnetic dipoles in the stripe domains is affected by thermal perturbation and stray fields from the MFM tip during heating, leading to the generation of skyrmions.^[18] Further heating the nanoflake to 323 K destabilized the skyrmions, and when the temperature reached 333 K, both stripe domains and skyrmions disappeared, indicating that T_c is near this threshold (Figure S10, Supporting Information). Upon cooling the nanoflake back to 313 K, the magnetic domain structure reverted to a stripe pattern, though it did not fully restore to its original configuration before the heating.

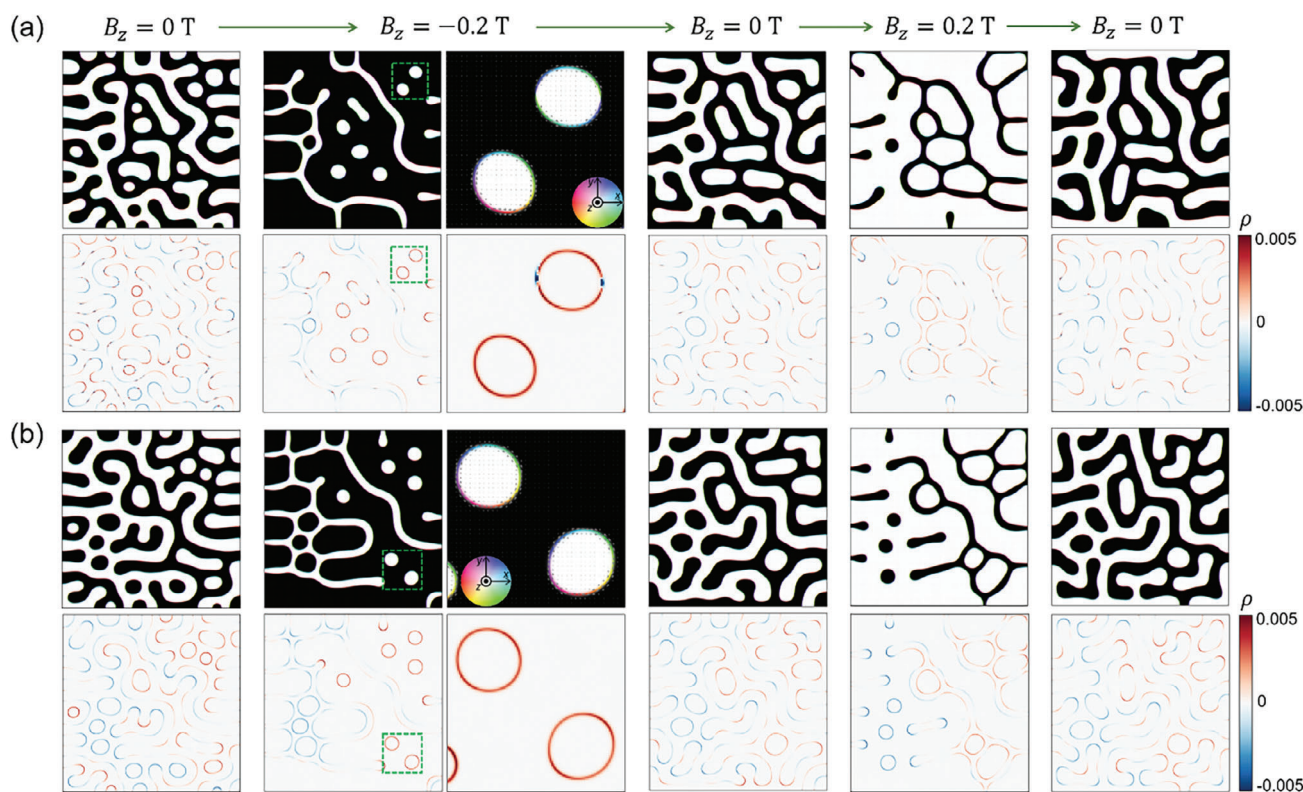


Figure 5. Simulated magnetic domain pattern (top) and the associated topological charge density (bottom) of a CrTe film with a) $D = 0$ and b) $D = 0.1 \text{ mJ m}^{-2}$. Here, we first relaxed the magnetic structure from a random initial state without a magnetic field and then observed the evolution of the domains with the continuously varying magnetic field, as denoted at the top of the figure.

Figure 4g,h presents a correlation analysis of the MFM images, highlighting the regions where the magnetic domain structure changed with temperature variation (closer to 1 indicates higher similarity). A low correlation value (Figure 4g) between the domain structures at 313 and 333 K reflects significant changes, suggesting the loss of magnetic properties at higher temperatures. Conversely, a high correlation value (Figure 4h) between the structures before and after cooling to 313 K indicates partial recovery of the magnetic domain structure, with the T_c estimated around 333 K. Additionally, the nanoflake sample demonstrated robust stripe domains and preserved morphology even after one year of exposure to ambient conditions (Figure S7, Supporting Information). Elemental analysis of the sample's cross-section revealed no signs of oxidation (Figure S8, Supporting Information), further supporting the stability of both the magnetic and crystalline structure in $\text{Cr}_{0.85}\text{Te}$ nanoflakes.

2.5. Micromagnetic Simulation of Skyrmion Formation

To further investigate the formation of the skyrmion-like regions (i.e., the skyrmionic structures), we performed the micromagnetic simulation on a CrTe flake. The exchange stiffness and magnetic anisotropy were first calculated using a nonequilibrium Green's function approach and then incorporated into a simulated flake system of dimensions $1 \mu\text{m} \times 1 \mu\text{m} \times 40 \text{ nm}$. The

system was initially relaxed from a random initial state without a magnetic field, followed by the continuous application of a z-directional magnetic field B_z to study the associated evolution of the magnetic domain pattern. Figure 5a shows the simulation without considering DMI. At $B_z = 0$, stripe domains emerged, emphasizing the crucial role of dipolar interaction in stripe domain formation. This is consistent with experimental observations (Figure 2f), where stripe domain wall widths follow the Kittel model.^[34] Applying a finite B_z caused the stripe domains to shrink, leading to the appearance of circular skyrmion-like regions. When B_z was removed, the stripe domain pattern recovered. We noted that the negative B_z is more favorable for the formation of the skyrmion-like regions. Upon closer examination of the relaxed magnetic structures under finite B_z , some circular regions were identified as Bloch-type skyrmions, while others were trivial bubbles comprising segments with opposite chiralities. We also calculated the topological charge densities of the simulated magnetic structures. It revealed singularities at most domain walls, where the charge density abruptly changes its sign, corresponding to the points of the chirality reversal on the domain walls and the bubbles. These chirality reversals were also observed in prior micromagnetic simulations of $\text{Cr}_{0.87}\text{Te}$,^[20a] where only dipolar interaction was considered.

Previous reports^[11] suggest that concentration gradient can break the inversion symmetry and introduce finite DMI, typically in the range of $0.1\text{--}0.2 \text{ mJ m}^{-2}$, which is much smaller

than the DMI parameter in systems with intrinsic structural asymmetry.^[39] However, our simulations show that even a modest DMI is sufficient to suppress trivial bubbles and stabilize skyrmions. Figure 5b illustrates that introducing the DMI parameter of $D = 0.1 \text{ mJ m}^{-2}$ prevents singularities on stripe domain walls, and all the circular regions that emerged under finite Bz are Bloch-type skyrmions rather than trivial bubbles. Both positive and negative Bz are favorable for the formation of skyrmions. Further tests with different D values revealed that even a minimal $D = 0.04 \text{ mJ m}^{-2}$ was sufficient to eliminate singularities on domain walls (Figure S11, Supporting Information). These findings suggest that the observed magnetic domain is governed by the interplay of intrinsic dipolar interactions and concentration gradient-induced DMI. Dipolar interactions primarily drive stripe domain formation, while even the relatively small DMI can stabilize the domain wall chirality, leading to skyrmion formation.

Additionally, we noted that the skyrmions remain Bloch-type even when DMI is considered, which aligns with their origin as shrunken stripe domains. Dipolar interaction dictates stripe domain formation, which is evident from the experimental observation that the domain wall width is well described by the Kittel model. Therefore, the Bloch-type domain walls should emerge in this case to minimize the system energy, leading to skyrmions with Bloch-type character. The DMI introduced by the concentration gradient can help stabilize the chirality of the skyrmions but is not sufficient to change the morphology of these skyrmions.

3. Conclusion

In this work, we successfully synthesized high-quality $\text{Cr}_{0.85}\text{Te}$ nanoflakes and investigated their magnetic behavior across a range of temperatures and magnetic fields primarily using MFM. Our experimental data revealed the emergence of stable skyrmions under sweeping magnetic fields and thermal stimulation. Through detailed analysis and support by micromagnetic simulations, we demonstrated that the stabilization of the strip domain and skyrmions in the $\text{Cr}_{0.85}\text{Te}$ nanoflakes is governed by the interplay between dipolar interactions and a compositional gradient-induced DMI. Even a weak DMI was identified as a crucial factor for realizing room-temperature skyrmions in the CVD-grown $\text{Cr}_{0.85}\text{Te}$ system. A promising direction for future research is to explore the detailed spin configuration of the skyrmions using Lorentz TEM, which would offer deeper insights into the magnetic properties of the nanoflake. The successful synthesis and manipulation of magnetic domains in $\text{Cr}_{0.85}\text{Te}$ nanoflakes under ambient conditions significantly advance the field of 2D magnetic materials, particularly in understanding and realizing room-temperature skyrmion states. These findings underscore the vast potential of the Cr–Te material system for next-generation spintronic applications, enabling controlled manipulation of skyrmions and other exotic spin textures.

4. Experimental Section

Synthesis of $\text{Cr}_{0.85}\text{Te}$ Nanoflakes: The $\text{Cr}_{0.85}\text{Te}$ nanoflakes were synthesized on SiO_2/Si substrates using CVD systems with dual temperature intervals. Te powders (20 mg, Macklin) were placed in the upstream zone

and sublimed at 550°C . CrCl_3 (2–3 mg, Aladdin) powders were placed in the middle of the second heating zone with the SiO_2/Si substrates covered above CrCl_3 powders, as shown in Figure S1a,b (Supporting Information). The polished downward-facing substrate was supported from the bottom by two polished upward-facing substrates, which constructed a confined space for growth. Before heating, the furnace was purged by 500 sccm high-purity Ar gas for 5 min. The two temperature zones were heated to 550 and 850°C within 40 min, respectively. The temperature was held for 20 min. The gas precursors were carried by mixed gases of 12 sccm H_2 and 48 sccm Ar in the growth process. After synthesis, the system was cooled down to room temperature naturally.

SEM/EDS Analysis: The morphology and composition of $\text{Cr}_{0.85}\text{Te}$ samples were characterized using a scanning electron microscope with energy-dispersive X-ray spectroscopy attachment (Zeiss Sigma300).

TEM Characterizations: The cross-sectional TEM specimen was prepared using the focused ion beam (FIB, Helios G3) by standard lift-out procedure, followed by 2 kV polishing. TEM characterization techniques, including bright-field (BF) imaging and selected area electron diffraction (SAED), were carried out on Talos F200 operated at 200 kV.

AFM and MFM Characterizations: Scanning probe microscopy (MFP-3D-Infinity, Oxford instrument, USA) was used to assess the thickness, surface morphology, and microscale magnetic domains of $\text{Cr}_{0.85}\text{Te}$ nanoflakes. The AFM measurements employed a probe (AC160TS-R3: spring constant (k) $\approx 26 \text{ N m}^{-1}$, resonance frequency (f) $\approx 300 \text{ kHz}$) operating in tapping mode. In this mode, the resonating probe was employed to scan the surface of the nanoflakes. Surface undulations induced variations in the contact force, consequently altering the probe's amplitude, thereby obtaining topography information. For MFM measurements, a magnetic probe (ASYMFMHC: 30 nm CoPt/FePt coating, $H_c > 5000 \text{ Oe}$, $k \approx 2.8 \text{ N m}^{-1}$, $f \approx 75 \text{ kHz}$) was employed. Initially, a line scan was performed to acquire surface topography data, followed by lifting the probe to a specified height (50 nm) to detect long-range magnetic force signals. The magnetic field between the probe and the nanoflakes generated a magnetic force gradient, leading to a deviation in the probe's resonance frequency, thereby elucidating the magnetic domain structure of the nanoflakes. Negative (positive) frequency shifts indicated attractive (repulsive) interactions between the tip and the nanoflakes. Additionally, in situ external magnetic fields and heating were applied using the Variable Field Module 4 (VFM 4) module and the high-temperature module, respectively, enabling simultaneous investigation of the magnetic domain evolution.

ToF-SIMS: The ToF-SIMS analysis was performed using a Time-of-Flight Secondary Ion Mass Spectrometer (IONTOF M6, Germany). Before analysis, the samples were sputtered with a low-energy argon ion beam to remove any surface contamination and to ensure a clean surface for analysis. The ToF-SIMS analysis was conducted under a vacuum atmosphere. The primary ion beam used for analysis was the O_2 plasma beam. The primary ion beam was rastered over the sample surface with a defined scan area and dwell time to ensure comprehensive analysis. The secondary ions emitted from the sample surface were collected and analyzed using a time-of-flight mass spectrometer. The mass spectra were acquired over a specific mass range and with a defined mass resolution to obtain detailed information about the surface composition and molecular structure. The ToF-SIMS images were acquired by scanning the primary ion beam across the sample surface and collecting the secondary ion signals at each pixel. The acquired ToF-SIMS data were processed and analyzed using SurfaceLab 7 software.

Micromagnetic Simulations: First-principles calculations are performed based on density functional theory (DFT) as implemented in the Vienna ab initio simulation package (VASP). The pseudopotentials are described using the projector augmented wave (PAW) method, and the exchange–correlation function is treated within the generalized gradient approximation (GGA) developed by Perdew–Burke–Ernzerhof (PBE). The cutoff energy for the plane-wave expansion is set to 500 eV, and the k -point grid is set to $12 \times 12 \times 10$ to sample the irreducible Brillouin zone.

The exchange parameters and the magnetic anisotropy are carried out using the nonequilibrium Green's function formalism (DFT+NEGF approach),^[40] as implemented in QuantumATK (version V-2022.12-SP1, <https://www.synopsys.com-1silicon/quantumatk.html>). A cutoff energy of

145 Hartree, and k -point meshes of $13 \times 13 \times 7$ are used for the self-consistent calculations. The relativistic SG15 pseudopotentials are included in the calculations.

Micromagnetic simulations were performed by the software Mumax3 based on the LLG function. The nanoflake size as $1000 \times 1000 \times 40$ nm with a rectangle mesh of size $2 \times 2 \times 2$ nm was set. The saturation magnetization $M_s = 6.98 \times 10^5$ A m⁻¹, the exchange stiffness constant $A_{ex} = 3.75 \times 10^{-12}$ J m⁻¹, the uniaxial magnetic anisotropy constant $K_u = 5.67 \times 10^5$ J m⁻³ calculated by the DFT calculation, and the gyromagnetic ratio $\alpha = 0.5$ were used in the simulation.

Supporting Information

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

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

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

Keywords

Cr_{0.85}Te nanoflakes, magnetic domain evolution, magnetic force microscopy, room temperature 2D ferromagnetic, skyrmions

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