Tunable Néel–Bloch Magnetic Twists in Fe₃GeTe₂ with van der Waals Structure

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1. Introduction

2D van der Waals (vdW) magnets provide an extraordinary platform for exploring magnetism and topological textures, such as skyrmions due to their unique layered structure and intrinsic long-range ferromagnetic order.[1,2] Skyrmions, being particle-like topological spin textures, have recently been reported in various vdW magnets.[3–10] As theoretically predicted, typical Bloch-type bubbles appear in centrosymmetric ferromagnets (FM) with magnetic dipolar interaction while Néel-type skyrmions are favored in thin heterostructure heavy metal/FM with interfacial DMI.[8,9,11–13] Several experimental observations support such theoretical predictions: Bloch-type bubbles, stabilized by the magnetic dipolar interaction in centrosymmetric FM, were claimed in the (001) thin plates (thinned from a bulk magnet) of vdW magnets Cr₂Ge₂Te₆[4] and FGT,[5,8] while Néel-type skyrmions were reported in a heterostructure WTe₂/FGT with an estimated large interfacial DMI strength of 1.0 mJ m⁻² at the interface, much larger than their calculated critical DMI value of ≈0.1 mJ m⁻² necessary for stabilizing Néel-type twists including skyrmions and domain walls.[6] The stability of Néel-type skyrmions is also predicted in multilayered films, such as Pt/oxidized-FGT/FGT/oxidized-FGT,[9] and FGT/Co/Pd.[7] However, the direct experimental identification of the Néel- or Bloch-type magnetic twists in vdW magnets remains elusive.

To identify the Néel- or Bloch-type magnetic twists, researchers[3–9,14] attempt to use Lorentz transmission electron microscopy (TEM) observations while simultaneously tilting the thin sample: the Lorentz TEM images present magnetic contrast for Bloch-type twists, but monotonic intensity (no contrast) for Néel-type twists when the tilt angle is zero. Whereas magnetic contrast arises for the Néel-type twists upon nonzero sample tilt. The conclusion has been drawn that one may therefore use Lorentz TEM jointly with sample tilt as the evidence for identification of Néel-type twists. However, such Lorentz TEM observations are not sufficient to unambiguously identify the magnetic twist type because the majority of the contrast measured at a tilt arises from the sample-plane projection of the out-of-plane magnetization, and the Néel- or Bloch-type spin-rotating signature is not directly detected. One
example is a demonstration of Bloch-type bubbles in a uniaxial FM Fe$_3$Ru$_2$(Mn$_{1-x}$Ru$_x$)$_3$O$_7$ with centrosymmetric crystal structure, where the DMI is absent.\cite{35} In this case, although there is monotonic intensity at zero tilt, bubble domains can be discerned at non-zero tilt angle, indicating that the Bloch-type domain walls are too narrow to be detected by defocused Lorentz TEM observations at zero-tilt angle.

On the other hand, micromagnetic simulations\cite{16,17} predict that Bloch-type skyrmions elongate along the direction perpendicular to the in-plane field while Néel-type skyrmions elongate along the field direction since the magnetic moments parallel to the in-plane bias field grow while the antiparallel one shrink. Previous Lorentz TEM observations have shown that in-plane bias fields align the $q$-vector of Néel-type helices perpendicular to the field direction in multilayered thin films,\cite{18} while changing that of the Bloch-type helices parallel to the field direction in the chiral magnet FeGe.\cite{19–21} The results suggest that the external in-plane field can induce a 90°-rotation difference of $q$ vectors for Néel and Bloch-type twists in DMI-stabilized systems. In addition, in-plane fields align skyrmion strings along the field direction in a thick FeGe thin plate relative to its periodicity.\cite{20–22} Note that the alignment of Bloch-type twists in magnetic dipolar systems may show differences since there are non-negligible magnetostatic charges.

Here we aim to tune the magnetic twists and hence to identify their twisting nature in the target vdW magnet FGT with/without interfacial DMI. On the basis of the abovementioned theoretical predictions\cite{16,17} for the in-plane magnetic field-aligned Bloch- and Néel-type skyrmions, we observe the changes in the magnetic twists in FGT when applying in-plane fields. We first measure a series of defocused Lorentz TEM images at non-zero sample tilt in a heterostructure Pt/oxidized-FGT/FGT/oxidized-FGT (named heterostructure FGT below)/a (001) thin plate fabricated from bulk FGT by focused ion beam (named fabricated FGT below). Skyrmions in the heterostructure FGT show monotonic intensity at zero tilt and the reversed contrast at opposite tilting angles, respectively, while skyrmionic bubbles in the fabricated FGT show clear contrast at zero tilt. Then we use an in-plane magnetic field, obtained by changing the electric current of an additional solenoid placed near the specimen plane in Lorentz TEM, to align the magnetic twists in two samples, and compare the field-induced $q$-vector reorientation with micromagnetic simulations. The real-space observations demonstrate that a weak in-plane magnetic field aligns the $q$-vector of magnetic twists perpendicular to the field direction in the FGT heterostructure, coinciding with that of Néel-type dominated twists in micromagnetic simulations. In contrast, the $q$-vector in the fabricated FGT rotates away from the in-plane field direction and the aligned twists hold fan-like modulations. On the basis of our experimental observations together with simulations, we discuss the possible mechanism of Néel-type twists forming in the FGT heterostructure, and alignments of twisted structures in two samples under in-plane magnetic fields.

### 2. Results and Discussion

In Figure 1a–g, we show the Lorentz TEM observation of magnetic skyrmions while applying a normal magnetic field in the heterostructure FGT. The FGT shows a layered crystalline structure with vdW gap (Figure 1a) and is sandwiched by asymmetric layers of naturally oxidized-FGT (O-FGT) and capped by a Pt layer (Figure 1b). The heterostructure FGT is then exfoliated and transferred on a Si$_3$N$_4$ membrane (Figure 1b) as described in Ref. [9]. We generate a metastable skyrmion lattice (Figure 1c) with 40 mT-field cooling (FC) from room temperature (RT) to 160 K (the external magnetic field is normally applied on the (001) FGT plane). Figure 1d–f shows a single skyrmion at various sample tilts. At zero-degree tilt, the skyrmion does not produce any contrast in the defocused Lorentz TEM image (defocused distance $\Delta f = 1$ mm). The skyrmions' magnetic contrast increases with increasing tilt angle while holding $\Delta f$ constant. At tilt angles of $\pm 20^\circ$ (the dashed line indicates the rotation axis), the skyrmion produces elliptical-shaped contrast with half-dark and half-bright, and the contrast reverses with reversing the tilt direction. Note that this contrast is only discerned in Lorentz TEM images obtained at a non-zero tilt angle, no contrast is observed at zero tilt. The Lorentz TEM image contrast, therefore, corresponds to the in-plane projections of out-of-plane magnetizations with tilting: upward in the center and downward at the periphery of the skyrmions (oriented in the $\pm z$-direction at 0° sample tilt) as drawn in the schematic of Figure 1u,v. The magnetic induction field map of the tilted skyrmion, analyzed using the transport-of-intensity equation,\cite{23} is shown in Figure 1g. The closed-loop in-plane magnetic induction field is attributable to the tilted normal magnetizations projected in-plane, which induces a non-zero electron phase change such that the Lorentz TEM image shows visible contrast.

Next, we measured the magnetic textures in the fabricated FGT thin plate (Figure 1h), which has no capping layers, to compare the difference with that in the above heterostructure FGT. The Lorentz TEM images (Figure 1i) and corresponding magnetic induction maps (Figure 1j) show typical magnetic bubbles generated after 250 mT-FC from RT to 94 K. Two types of bubbles are presented: one, which we call skyrmionic bubbles, have a winding number $N \approx 1$ (as shown by the magnetic induction field map in Figure 1n) and two helicities (i.e., clockwise and counter-clockwise) (as marked in Figure 1j, and Figure S1, Supporting Information). The other exhibits $N = 0$ (see details in Figure S1, Supporting Information). The magnetic bubbles are also observed in a thinner plate, suggesting the difference between the observed spin textures are not thickness related (see details in Figure S2, Supporting Information). Note that the skyrmionic bubbles are observed at zero tilt and at a relatively smaller $\Delta f = 200$ um than that for observing skyrmions in heterostructure FGT. We conclude here that the observed skyrmionic bubbles are Bloch-type, characteristic of those stabilized by the magnetic dipolar interaction in centrosymmetric FM. We also performed a series of Lorentz TEM observations at various tilt angles, as shown in Figure 1k–m. The magnetic contrast of the bubble is nearly symmetric at zero tilt (Figure 1l), and show unbalanced dark/bright contrasts at $\pm 20^\circ$ tilt that are reversed at opposite tilt angles (Figure 1k,m and Figure S3, Supporting Information). We also attribute this asymmetric contrast to the projected in-plane components of the core and peripheral out-of-plane magnetizations. The experimentally observed contrast is consistent with the
Lorentz TEM simulations of a Bloch-type bubble (Figure 1s,t) in terms of a multi-slice Fourier approach [24] (see Methods in Supporting Information), where the domain wall twist continuously rotates perpendicular to the radial direction (as illustrated in the schematic in Figure 1r).

From the defocused Lorentz TEM observations in the above two samples, the magnetic contrast at zero tilt is quite different: no contrast for skyrmions in heterostructure FGT (Figure 1e) versus clear contrast for bubbles in fabricated FGT (Figure 1l). While there is no doubt that the bubbles in the fabricated FGT are Bloch-type, the type of magnetic twist (skyrmion) in the heterostructure FGT is still unclear, even though the Lorentz TEM observations in Figure 1d–f seem to be consistent with the simulated Néel-type skyrmion (Figure 1p,q). Néel-type skyrmions, whose magnetic moments within the domain walls are oriented radially (Figure 1o,p), do not induce contrast due to the cancelling-out of azimuthal electron deflections when the sample is not tilted. On the other hand, the invisible contrast in defocused Lorentz TEM images observed at zero tilt angle could also be attributed to the Bloch-type twists where the domain wall is too narrow to be detected by defocused Lorentz TEM measurements, as reported in the aforementioned centrosymmetric FM. [15]

To identify the domain wall twist of skyrmions, we apply an in-plane field on the Au-capped heterostructure FGT and use Lorentz TEM to probe the field-aligned magnetic twists. The Lorentz TEM observations were performed at 20° sample tilt (as indicated in Figure 2a): after zero-field cooling from RT to 143 K, maze domains appear (Figure 2d); after in-plane field
(15 mT) cooling from RT to 143 K, a single-\emph{q} state appears with a \emph{q} vector perpendicular to the field direction (Figure 2e). The corresponding magnetic induction field maps (Figure 2f,g and Figure S5, Supporting Information) analyzed from defocused Lorentz TEM images indicate the in-plane projections of the out-of-plane moments at tilt, and not the twisted domain walls (as described by the schematics of Figure 2b,c). The Lorentz TEM images at 0° with or without in-plane fields show no contrast (Figure S4, Supporting Information).

We have compared the experimental observations of in-plane field-aligned magnetic twists with micromagnetic simulations of Néel-type dominated twists (Figure 2h–k, see Methods in Supporting Information). The simulated Néel-type maze domains (Figure 2h,j) are relaxed from the paramagnetic state at zero field. While holding an in-plane field of 275 mT to relax the magnetic twists, they are aligned well with a \emph{q} vector perpendicular to the in-plane field direction. Their projected magnetic induction field maps (Figure 2j,k) at 20° tilt are calculated.
from those at 0° tilt. The simulated Lorentz TEM image contrasts of the maze domains (Figure 2h) and the single-q state (Figure 2i) coincide with the experimental observations in the heterostructure FGT at 20° (Figure 2d,e). The consistency of the experimental observations (Figure 2d–g) and the simulations (Figure 2h–k) suggests that the observed magnetic twists in the heterostructure FGT (Figures 1c–f and 2d–g) are dominantly Néel-type.

In comparison with the Néel-type twists in the heterostructure FGT, we have also applied in-plane fields on the Bloch-type twists in the fabricated FGT (Figure 3). Lorentz TEM observations are performed at zero tilt since Bloch-type twists impart a measurable phase change on the passing electron wave, resulting in a build-up of intensity. Figure 3a,c show the Lorentz TEM images and magnetic induction field maps of the maze domains obtained after zero-field-cooling to 100 K. The magnetic twists align to a single-q state after FC at $\mu_0 H_x = 15$ mT (Figure 3b,d), where the q-vector rotates and aligns at an angle of $\sim 53°$ with respect to the in-plane field direction. Note that the aligned twists in Figure 3d are “fan-like twists” since the domain wall twists are oriented along a single direction, that is, they tend to be polarized with the in-plane field. The Lorentz TEM image of Figure 3b shows that the domain-wall contrasts hold alternating half-white and half-dark contrasts (as described by the inserted schematics).

**Figure 3.** Bloch-type twists with a rotated q-vector via an external in-plane magnetic field ($\mu_0 H_x$) applied on a fabricated FGT thin plate. a,b) Lorentz TEM images and c,d) corresponding magnetic inductions at a,c) $\mu_0 H_x = 0$ mT and b,d) $\mu_0 H_x = 15$ mT, reveal an alignment of the modulation q-vector at an angle of $53°$ with respect to the field direction. The inserted schematics in (b) describe the black/white contrasts of domain walls in Lorentz TEM images. e,f) Multi-slice Lorentz TEM simulations applied on micromagnetically simulated spin textures and g,h) corresponding magnetic in-plane induction maps of Bloch-type twists with e,g) $\mu_0 H_x = 0$ mT and f,h) $\mu_0 H_x = 275$ mT, which qualitatively agrees with (a–d). Bloch lines present in the domain walls are circled in yellow in (a,e,g).
We have further performed micromagnetic simulations for Bloch-type twists. The simulated maze domains (Figure 3e–g) are relaxed from a randomly distributed spin configuration at 0 mT. A single-$q$ state with fan-like twists forms upon FC (Figure 3f,h). The simulated Lorentz TEM image contrast, magnetic induction field maps of maze domains and the rotated-$q$ twists (Figure 3e–h) coincide with the experimental observations in fabricated FGT (Figure 3a–d). Fan-like twists with a rotated-$q$ vector in the present fabricated FGT is different from the field-aligned twists in the chiral magnets exhibiting DMI, where the $q$ aligns parallel to the in-plane field direction. It is possibly because there are many magnetostatic charge-induced Bloch-lines in the present FGT with the magnetic dipolar interaction, as marked by yellow circles in Figure 3a,e,g and Figure S1, Supporting Information. The Bloch lines are easily tuned to orient to the in-plane field direction, and hence it may facilitate the emergence of fan-like twists.

Our combined experimental and simulation efforts show that the magnetic twists in the heterostructure FGT hold a Néel-type dominated nature identified by the $q \perp \mu_0H_x$ alignment under the in-plane field together with the contrast changes in a series of tilted Lorentz TEM images. When we apply an external magnetic field while cooling the samples from a paramagnetic state, the magnetic field influences the magnetic ordering of the stripe domain as the order forms, inducing a preferred $q$ vector alignment. The applied magnetic field of 15 mT may slightly tilt the magnetizations toward the field direction, but the contrast of the domain walls of the stripes and skyrmions after cooling with or without a field applied doesn’t noticeably change, indicating that the external magnetic field does not change the wall type of stripes and skyrmions that is determined by the intrinsic parameters of the thin FGT samples. The identification of Néel-type twists suggests a possible inherent interfacial DMI existing in the heterostructure FGT, intrinsically different from the trivial bubbles stabilized by the magnetic dipolar interaction in the fabricated FGT thin plate. The DMI energy stabilizing the Néel-type twists arising from the asymmetric interface between the Pt or Au capping layers, O-FGT, and FGT itself is predicted in band calculations. The local symmetry breaking of the FGT crystal structure, the magnetic coupling between FGT and capping layer, and/or the asymmetric strain effects (arising from the difference between the free top surface of FGT and its confined bottom surface on the Si$_3$N$_4$ membrane) may also contribute to the asymmetric interaction. In addition to different thicknesses and interfaces between the heterostructure FGT and fabricated FGT, the surface state of each sample is different: the fabricated FGT exhibits a Ga-beam-damaged surface layer while the heterostructure FGT with capping layers should be damage-free. However, the damaged layer should be sufficiently thin to treat as a negligible effect on the magnetic order in the fabricated FGT. The sample thickness is also an important factor to affect the Néel or Bloch-type twists. A detailed theoretical explanation requires further studies. There is, however, clear experimental evidence coupled with micromagnetic simulations that the Néel-type nature of magnetic twists is dominant in the heterostructure FGT. On the other hand, we have shown that Bloch-type twists in the fabricated FGT thin plate have many intrinsic Bloch-lines (see Figure 3a,e,g and Figure S1, Supporting Information) at zero field and orient with a rotated $q$ vector with fan-like modulations under in-plane fields. These Bloch lines may facilitate the transition from maze domains into fan-like modulations. These characteristics show a remarkable difference from those in the heterostructure FGT (Figures 1 and 2) and chiral magnets.\[20,21]\]

3. Conclusion
In summary, we have demonstrated the alignment of magnetic twist $q$ vector perpendicular to the in-plane field direction in heterostructure FGT and a rotated $q$ vector in the fabricated FGT, which coincides with micromagnetic simulations. Comparing Lorentz TEM observations of magnetic twists in the heterostructure FGT and the fabricated FGT, we conclude here that we may tune Néel–Bloch twists by engineering capping layers in vdW-structure systems. Distinguishing and tuning Bloch- versus Néel-type magnetic twists is a significant aid in researchers’ quest to understand the physical nature of magnets and engineer 2D magnetic spintronic devices.

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 work are available from the corresponding authors upon reasonable request.

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