Néel-type skyrmion in WTe$_2$/Fe$_3$GeTe$_2$ van der Waals heterostructure

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Abstract

The promise of high-density and low-energy-consumption devices motivates the search for layered structures that stabilize chiral spin textures such as topologically protected skyrmions. Recently discovered long-range intrinsic magnetic orders in the two-dimensional van der Waals materials provide a new platform. Here we demonstrate the Dzyalosinskii-Moriya interaction and Néel-type skyrmions are induced at the WTe$_2$/Fe$_3$GeTe$_2$ interface. Fe$_3$GeTe$_2$ is a ferromagnetic material with strong perpendicular magnetic anisotropy. We demonstrate that the strong spin-orbit interaction in 1T’-WTe$_2$ does induce a large interfacial Dzyalosinskii-Moriya interaction at the interface with Fe$_3$GeTe$_2$ due to the inversion symmetry breaking to stabilize skyrmions. Transport measurements show the topological Hall effect in this heterostructure for temperatures below 100 K. Furthermore, Lorentz transmission electron microscopy is used to directly image Néel-type skyrmions along with aligned and stripe-like domain structure. This interfacial coupling induced Dzyalosinskii-Moriya interaction is estimated to have a large energy of 1.0 mJ/m$^2$, which can stabilize the Néel-type skyrmions in this heterostructure. This work paves a path towards the skyrmionic devices based on van der Waals heterostructures.

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Atomically thin, layered van der Waals (vdW) materials have been experimentally shown to host long-range magnetic orders recently\textsuperscript{1,2}, which could push the magnetic memory and information storage to the atomically thin limit and lead to ultra-compact next generation spintronics\textsuperscript{3,4}. Since the discovery of the ferromagnetism in Cr\textsubscript{2}Ge\textsubscript{2}Te\textsubscript{6}\textsuperscript{5,6}, CrI\textsubscript{3}\textsuperscript{7–10} and Fe\textsubscript{3}GeTe\textsubscript{2} (FGT)\textsuperscript{11–14}, such layered crystals have been at the frontier of material research. Besides the material itself, interfacial engineering in vdW heterostructure offers an effective methodology to spin-polarize or valley-polarize 2D materials. Proximity effect from the interface has been widely researched for spin or valley polarization in 2D materials, like graphene on transition metal dichalcogenides (TMDs)\textsuperscript{15–19} and WSe\textsubscript{2} on CrI\textsubscript{3}\textsuperscript{20–23}. Coupling 2D magnets to vdW materials not only lends the magnetic properties of these 2D magnets to the adjacent materials, but also modifies the magnetic properties of the 2D magnets themselves\textsuperscript{24}. Among all the possible interfacial coupling for 2D magnets, spin orbit coupling proximity can play an important role when the atoms of 2D magnets are in contact of heavy elements, given that the magnetic properties are intrinsically related to the spin orbit coupling.

Rashba spin-orbit coupling is reported to lead to a strong Dzyaloshinskii-Moriya interaction (DMI) at the interface\textsuperscript{24,25}, where the broken inversion symmetry at the interface can change the magnetic states. DMI has been recognized as a key ingredient in the creation, stabilization and manipulation of skyrmions and chiral domain walls. Whereas skyrmions from DMI becomes significant in the heavy metals/ferromagnet systems\textsuperscript{26–28}, there have been no direct observations of skyrmion in van der Waals heterostructures, even the topological Hall effect has been reported in Cr-doped topological insulator (TI)/TI\textsuperscript{29} and Mn-doped TI systems\textsuperscript{30}.

In this work, we have observed the topological Hall effect in WTe\textsubscript{2}/FGT vdW heterostructures from transport measurements. More importantly, a large DMI energy of 1.0 mJ/m\textsuperscript{2} has been determined in this system and the formation of Néel-type skyrmions has been captured with Lorentz transmission electron microscopy (L-TEM). The sizes of the directly observed skyrmions are \textasciitilde 150 nm at 94 K and \textasciitilde 80 nm at 198 K. This helps promote 2D materials for ultra-compact spintronic devices.

**Thickness characterization of WTe\textsubscript{2} and FGT**

Before we show the proximity effect, we first determine the properties of WTe\textsubscript{2} and FGT separately. The 1T’ crystal structure of WTe\textsubscript{2} has been confirmed in the previous work\textsuperscript{31}.
Atomic force microscopy was adopted to measure the thickness of this material as shown in Figs. 1 b-c. It shows that the 1L WTe$_2$ on the SiO$_2$/Si substrate has a height of about 1.1 nm. The transport data of 1L WTe$_2$ are given in the Supporting Information A. Bulk FGT consists of weakly bonded Fe$_3$Ge layers that alternate with two Te layers with a space group P6$_3$mmc as shown in Fig. 1a. For FGT, the 1L thickness is 0.8 nm$^{11}$. This thickness is used to determine the number of layers for FGT as shown in Figs. 1d-e. As shown in Figs. 1f-i, the Curie temperature decreases from $\sim$200 K to $\sim$100 K when the thickness of FGT goes down from 60L to 4L. The perpendicular magnetic anisotropy is well preserved for FGT with thickness down to seven layers. For thin FGT samples with seven layers and four layers, hexagonal boron nitride (h-BN) thin flakes are used for protection from the ambient.

**Topological Hall effect in WTe$_2$/FGT heterostructure**

WTe$_2$ possesses a largest spin-orbit coupling among the transition metal dichalcogenides$^{32}$. At the interface, the terminated atoms in WTe$_2$ are the same as those in FGT: two layer of Te atoms are easier to be coupled, where the strong spin-orbit interaction from WTe$_2$ could play a significant role in reorganizing the spin polarizations in FGT. Such effect can be captured by transport measurements in thin films, where transport data of h-BN/WTe$_2$/FGT heterostructures are shown in Fig. 2 with h-BN serves as the protection layer. For the h-BN/1L WTe$_2$/4L FGT sample as shown in Figs. 2a-b, the longitudinal resistance increases when temperature goes down from room temperature. The Curie temperature for this device is below 100 K, which is close to that of a 4L FGT on SiO$_2$/Si sample. Dips and peaks near the magnetic phase transition edge, as a signature for the topological Hall effect, show up below 100 K as shown in Figs. 2c-d. For example, at 50 K there's a dip or peak at a field strength $\sim$ ±975 Oe as indicated by the dashed lines. Different from this sample where the interfacial coupling has been reflected through the topological Hall effect, a 2L WTe$_2$/30L FGT heterostructure shows the perpendicular magnetic anisotropy is well preserved in FGT when the temperature is below 180 K in the Figs. 2e-g, indicated by the square loop of the Hall resistance. However, within an intermediate temperature range (180 K -200 K) as shown in Fig. 2g, the hysteresis loops differ in having transitions in steps. For example, at 190 K, the Hall resistance suddenly jumps from the low saturation value to an intermediate level and then changes linearly. Finally it saturates to the high saturation value at a relatively high positive field. The time-reversed process does the opposite as expected. A possible explanation for
this behavior is the formation of labyrinthine domain structures in FGT which results in multi domains\(^{12}\), and this will be confirmed in the following sections. Compared to the 1L WTe\(_2\)/4L FGT heterostructure as shown in Figs. 2a-d, this sample as shown in Figs. 2e-g with a much thicker FGT flakes measures the averaged transport signal which may lead to the smearing out of the topological Hall signal at the interface. In the following part, we utilized L-TEM to directly investigate the domain structure in 2L WTe\(_2\)/30L FGT heterostructure and confirm a large DMI at the interface.

**Experimental observation of Néel-type skyrmions with L-TEM**

Compared to other techniques\(^{33}\), L-TEM affords the advantage of a spatial resolution below 5 nm, which is one of the most direct methods to observe the magnetic domain structures, domain walls and skyrmions. The contrast formed in L-TEM is traditionally explained from the deflection of electrons due to the in-plane magnetic field. In our case, sample plane was tilted to have partially the in-plane magnetization for detecting the Néel-type skyrmion as schematically shown in Fig. 3a. The resulted image for a skyrmion should be of dark-bright contrast due to the contributions both from the outside and core of the skyrmion\(^{34}\). The 2L WTe\(_2\)/30L FGT heterostructure was successfully transferred onto a 15 nm-thick suspended SiN thin film for the L-TEM measurements as shown in Fig. 3b. Figure 3c shows the formation and disappearance of a Néel-type skyrmion when the magnetic field increases from 0 Oe to 900 Oe for this sample at 94 K. The Néel-type skyrmion is well developed at a field along the z direction with a strength of 540 Oe and 600 Oe, having dark at the top side and bright at the bottom. The size is \(\sim 150 \text{ nm}\). The out-of-plane magnetic fields are \(\sim 507 \text{ Oe}\) and \(\sim 564 \text{ Oe}\), correspondingly. At an elevated temperature of 198 K, skyrmions in this heterostructure show up with bright at the top side and dark at the bottom at a field of 390 Oe with a smaller size of \(\sim 80 \text{ nm}\) shown in Fig. 3d. When the temperature is raised to 203 K which is a little bit higher than the Curie temperature, there are no skyrmions formed as shown in Fig. 3e. For the 1L WTe\(_2\)/4L FGT heterostructure, a larger DMI from the interfacial coupling is expected since it shows a topological Hall effect in Fig. 2c-d. However, a high resolution domain structure using L-TEM for the 1L WTe\(_2\)/4L FGT heterostructure has not been obtained as a high energy electron beam tends to damage the samples with thinner FGT more easily.

**Experimental evidence of WTe\(_2\) induced DMI**

The strong perpendicular magnetic anisotropy favors out-of-plane magnetization, which
makes the spin polarization in the FGT form the domain in the up or down directions. Fig. 4a shows the magnetic domain images for a 30L FGT thin flake without WTe$_2$. The film exhibits labyrinth domains at 94 K and 0 T. When the in-plane field is tuned to the opposite direction, the contrast of domain edge completely switches the sign as shown in the cases of a tilting angle of $\alpha=-20^\circ$ and $\alpha=21^\circ$ as shown in Fig. 4a.

The formation and structure of domain walls are usually a result of the interplay between exchange interaction, magnetic anisotropy and dipolar interaction. Reducing the thickness of the FGT flakes decreases the dipolar interaction, thus, perpendicular magnetic anisotropy leads to a stabilized single domain$^{14}$. Meanwhile, the exchange interaction term here includes DMI, which is an antisymmetric exchange interaction that favors a chirally rotating magnetic structure of a specific rotational direction.

In the WTe$_2$/FGT heterostructure, there’s no contrast when $\alpha = 0^\circ$ as shown in Fig. 4b, which leads to the Néel-type domains. When $\alpha \neq 0$, we have observed this well aligned and stripe-like domains with a domain width $w=290$ nm at 0 T, which is sharply different from the domain structure of the FGT flake as shown in Fig. 4a, but with a much smaller domain width. Thus DMI is largely enhanced$^{28}$ for this heterostructure from the interfacial Te atoms coupling. A phenomenological model defines the dependence of the domain width $w$ on the domain wall energy $\delta_W$ by$^{35}$:

$$w = \beta \frac{4\pi\delta_W}{M_s^2},$$

(1)

here domain wall energy $\delta_W$ is related to exchange stiffness $A$, effective anisotropy constant $K_{eff}$, and DMI constant $|D|$ by $\delta_W = 4\sqrt{A K_{eff}} - \pi|D|$.$^{36}$ $\beta$ is a phenomenological fitting parameter and taken to be 0.31 for FGT$^{35}$. A domain width of 290 nm leads to a domain wall energy $\delta_w=0.77$ mJ/m$^2$ including the DMI term. For pristine FGT without DMI, the domain wall energy is simply expressed as $4\sqrt{A K_{eff}}$. Then by comparing these two domain wall energies with and without the DMI contribution, we obtain a DMI energy $|D|=1.0$ mJ/m$^2$ in our system, which is comparable to the previous value in heavy metal/ferromagnet thin film systems$^{27}$ (The detailed estimation of this $|D|$ can be found in the Supporting Information E). One can compare $|D|$ to the critical value $|D_c|$, which is required to stabilize chiral Néel domain wall$^{27,37}$,

$$|D_c| = \frac{4}{\pi} \sqrt{\frac{A}{K_{eff}}} K_d,$$

(2)

here the magnetostatic or stray field energy constant $K_d=2\pi M_s^2$ prefers a Bloch
configuration. Thus, $|D_c|$ is calculated to be $\sim 0.1 \text{ mJ/m}^2$, so that $|D| > |D_c|$ and chiral Néel textures are expected. Compared to the transport measurements which takes the averaged transport signal of the interfacial few nanometers FGT coupled to WTe$_2$ near the interface and other FGT layers away from the interface, L-TEM helps confirm the DMI and Néel-type skyrmion at the interface for the WTe$_2$ with FGT samples.

In summary, we reported the observed Néel-type skyrmions from L-TEM and the discovery of topological Hall effect in the WTe$_2$/FGT heterostructure. A large DMI energy of $\sim 1.0 \text{ mJ/m}^2$ from the interfacial coupling may be a result of the broken inversion symmetry from Rashba spin-orbit coupling. We showed Néel-type skyrmions of a small size ($\sim 150 \text{ nm at 94 K and } \sim 80 \text{ nm at 198 K}$) at the interface of the vdW heterostructure. Further researches can include electrically gating to control skyrmions in 2D vdW heterostructures, and this may open a new area in the field of ultra-compact next-generation spintronics.

**Methods**

**Sample assembly using a pick-up transfer technique:** In preparation, we coated the polydimethylsiloxane (PDMS) stamp on a glass slide with a polypropylene carbonate (PPC). During the assembly of heterostructure, we first exfoliated h-BN onto the 300-nm-thick SiO$_2$/Si substrate. Then the PDMS/PPC on the glass slide was used to pick up the h-BN from the SiO$_2$/Si substrate by heating up to around 40°C. After that, we aligned the PDMS/PPC/h-BN and the exfoliated WTe$_2$ on the 300-nm-thick SiO$_2$/Si substrate, and used the similar heating temperature to pick up WTe$_2$. Then we used PDMS/PPC/h-BN/WTe$_2$ to pick up FGT. By heating the samples up to 110°C, PPC was released from the PDMS and the PPC/h-BN/WTe$_2$/FGT was transferred onto the prepared bottom electrodes. Then acetone was used to remove the PPC thin film. The metal contacts used for the transport measurements of FGT thin flakes have two types, one is evaporation of Cr/Au electrodes after exfoliation of FGT onto the SiO$_2$/Si substrates and the other one is using the pick-up transfer technique to accurately align FGT with the prepared bottom Cr/Au electrodes. The length and width of contacts in a Hall bar geometry as shown in Fig.1f-i for 60L FGT, 30L FGT, h-BN/7L FGT, h-BN/4L FGT are $15 \mu\text{m} \times 9 \mu\text{m}$, $15 \mu\text{m} \times 9 \mu\text{m}$, $3.5 \mu\text{m} \times 0.5 \mu\text{m}$ and $3.5 \mu\text{m} \times 0.5 \mu\text{m}$, respectively.
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AUTHOR CONTRIBUTIONS

Y. W. and K. L. W. conceived and supervised the project. W. W. grew the FGT bulk crystals and Y. L. Z. and J. H. grew the WTe$_2$ bulk crystals. T. T and K. W provided the h-BN crystal. Y. W. fabricated devices and performed transport measurements. S. Z. and J. Z. performed the Lorentz transmission electron microscopy measurements. K. W. and Y. W. carried out the atomic force microscopy measurements. C. F., C. W. and X. H. prepared the bottom electrodes. Y. W. and K. L. W. wrote the manuscript with input from all authors.

DATA AVAILABILITY

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

COMPETING INTERESTS

The authors declare no competing interests.
Figure 1: Thickness characterization of WTe$_2$ and FGT flakes using atomic force microscopy (AFM) and layer-dependent transport properties of FGT flakes. a, Schematic graph for WTe$_2$ on FGT. b, Microscopic image of exfoliated WTe$_2$ flakes. c, Cross-sectional profile of the WTe$_2$ flakes along the blue line shown in b. d, Microscopic image of exfoliated FGT thin films. e, Cross-sectional profile of the FGT flakes along the black line shown in d. Temperature dependence of Hall resistance for f, 60L, g, 30L, h, 7L, and i, 4L FGT flakes shows that the Curie temperature decreases as the thickness of FGT decreases. Insets show the devices for the measurements separately. Scale bar: 10 $\mu$m.
Figure 2: Transport measurements of WTe$_2$/FGT heterostructures. a, Microscopic image of h-BN/1L WTe$_2$/4L FGT heterostructure. b, Longitudinal resistance dependence on the temperature shows the $R_{xx}$ increases when temperature drops. c, Magnetoresistance and d, Hall resistance of the heterostructure shown in a. Both longitudinal resistance and Hall resistance show a peak and dip near the transition edge before the magnetization saturates, which is a sign of the topological Hall effect. For each case, an offset is used for clarity. e, Microscopic image of h-BN/2L WTe$_2$/30L FGT heterostructure. f, Longitudinal resistance dependence on the temperature shows the metallic behavior when temperature drops. g Hall resistance of the heterostructure shown in e. An offset is used for clarity. Scale bar: 10 µm.
Figure 3: **Néel-type skyrmion observed under L-TEM.** a, Schematic diagram of a Néel-type skyrmion on a tilting sample for L-TEM imaging. The red and blue circles are for positive and negative magnetizations along z, respectively. Brown arrows indicate the in plane magnetization component while green arrows indicate the Lorentz force. b, Microscopic image of a 2L WTe$_2$ on a 30L FGT supported by a 15-nm-thick suspended SiN thin film with a width and length of 100 µm. Scale bar: 10 µm. c, L-TEM observation a Néel-type skyrmion at T=94 K, $\alpha=21.86^\circ$ and $H=540$ Oe, 600 Oe, where $\alpha$ is the angle between the sample plane and x-y plane. The yellow arrow points to a skyrmion. The skyrmion size is $\sim$150 nm. d, L-TEM observation of Néel-type skyrmions at 198 K with a field of 390 Oe and $\alpha=-29.19^\circ$. The yellow arrows point to the skyrmions. e, All skyrmions disappear at 203 K with a field of 390 Oe and $\alpha=-29.19^\circ$. 
Figure 4: Magnetic domains of FGT (region 1) and WTe$_2$/FGT heterostructures (region 2) with tilt angles $\alpha$. a, A typical labyrinth domain in 30L FGT thin flakes. b, From the aligned and stripe-like domain structure of the WTe$_2$/FGT, a DMI energy is estimated to be $\sim$1.0 mJ/m$^2$. 
Supporting Information for Néel-type Skyrmion in WTe$_2$/Fe$_3$GeTe$_2$ van der Waals heterostructure

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A. Hall resistance of monolayer WTe\(_2\)

To confirm the dip and peak near the magnetic transition edge in the transport signal as shown in Fig. 2 in the main text are from the topological Hall effect, we have shown the Hall resistance of h-BN/monolayer WTe\(_2\) in Figure S1. This helps exclude the possibility that the dip and peak are from the transport signal of WTe\(_2\) and thus leads to the conclusion that topological Hall effect exists at the interface.

Figure S1. Hall resistance of a monolayer WTe\(_2\) covered by h-BN on bottom electrodes.

B. Field-dependence of the stripe-like domains

Figure S2 show the evolution of the magnetic domains in the WTe\(_2\)/Fe\(_3\)GeTe\(_2\) heterostructure with increasing magnetic fields. When the field increases, the magnetic domains are transforming to a single domain.
Figure S2. Field dependence of the magnetic domains at $\alpha=-20^\circ$ and $T=94$ K in WTe$_2$/Fe$_3$GeTe$_2$ heterostructure from 0 Oe to 1200 Oe.

C. Skyrmion size

As shown in Figure S3, a line profile is used to analyze the contrast for a skyrmion. The distance between the lowest and highest data points is the skyrmions size.

D. Measurement of domain width

Bodenberger and Hubert[1] used a stereological method to define the surface magnetic domain width $w$ of complicated or arbitrary magnetic structure patterns. In their method, $w$ is defined as:

$$w = \frac{2 \times \text{total test line length}}{\pi \times \text{number of intersections}},$$

which appears to be the most universal and commonly applied method[2-4]. In this method, an effective domain width is defined as the ratio of total test line length to the number of intersections of domain walls. For the purpose of evaluating the total domain width, four test straight lines running in random directions is used; the method is illustrated in the
**Figure S3.** Line profile for the image of skyrmions. a, The skyrmion size is determined to be ∼ 150 nm at 94 K. b, The skrymion size is determined to be ∼ 80 nm at 198 K.

The image of Figure S4, where four test lines are drawn. The determined domain width is 290 ± 10 nm.
E. Estimation of DMI constant

Based on the Stoner-Wohlfarth model[5], the uniaxial anisotropy constant $K_u$ can be derived via:

$$\frac{2K_u}{M_s} = \mu_0 H_{\text{sat}}. \quad (2)$$

As shown in Figure S5 for 2L WTe$_2$/30L FGT heterstructure, $H_{\text{sat}}$ decreases when the temperature increases. Thus we can determine the ratio of the uniaxial anisotropy constant at 5 K $K_u$ at 5K and at 94 K $K_u$ at 94K. Meanwhile, Ref[6] lists the parameters for bulk FGT around 5 K: $M_s$ at 5K=376 emu/cm$^3$, $K_u$ at 5K=1.46 $\times$ $10^7$ erg/cm$^3$, $A = 10^{-7}$erg/cm. Thus $K_u$ at 94K $\sim$ 9.7$\times$10$^6$ erg/cm$^3$. Since $K_d \ll K_u$, the effective anisotropy constant $K_{\text{eff}}$ $\sim$ 9.7$\times$10$^6$ erg/cm$^3$. As a result, the domain wall energy for FGT without the DMI contribution is $\sim$3.9 mJ/m$^2$. A DMI constant of 1.0 mJ/m$^2$ is obtained in our system.

Figure S4. Representative image used to obtain the average domain size of WTe$_2$/FGT sample.
Figure S5. Hall resistance of 2L WTe$_2$/30L FGT heterostructure at 5 K and 94 K.

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