

Supplementary Information for

Above-room-temperature strong intrinsic ferromagnetism in 2D van der Waals Fe_3GaTe_2 with large perpendicular magnetic anisotropy

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Supplementary Note 1. XPS analysis of the Fe₃GaTe₂ crystal.

Before the XPS measurement, an exfoliated fresh Fe₃GaTe₂ crystal were quickly put into the vacuum XPS chamber to avoid oxidation. Except for the C element (C 1s, 285 eV) used to calibrate the peak position, only Fe, Ga, and Te were detected from the spectra of the Fe₃GaTe₂ crystal.

As shown in Supplementary Fig. 3a, the high-resolution Fe 2*p* spectra was deconvoluted into Fe 2*p*_{3/2} and Fe 2*p*_{1/2}. The two peaks at 706.9 and 720.0 eV originate from the metallic iron (Fe⁰) in Fe₃GaTe₂. The two other doublets with binding energies of 710.8 and 724.4 eV (with 713.7 and 727.3 eV satellites) can be ascribed to Fe³⁺ in Fe₃GaTe₂. Further, the area ratio of Fe³⁺ peaks and Fe⁰ peaks is calculated as 2.08:1, which very close to the theoretical ratio 2:1 and corresponded to the Fe-I (Fe³⁺) and Fe-II (Fe⁰) in Fe₃GaTe₂. Meanwhile, the analysis of the high-resolution Ga 2*p* spectra in Supplementary Fig. 3b shows Ga 2*p*_{3/2} and Ga 2*p*_{1/2} peaks with binding energies of 1117.6 and 1144.5 eV. Similarly, Te phase (Te²⁻) in the high-resolution Te 3*d* spectra was also deconvoluted into Te 3*d*_{5/2} and Te 3*d*_{3/2} with binding energies of 572.8 and 583.2 eV, respectively (Supplementary Fig. 3c).

Supplementary Note 2. Room-temperature magnetic anisotropy of the Fe₃GaTe₂ crystals.

The magnetic anisotropy of Fe₃GaTe₂ bulk crystals is analyzed by magnetic field dependent magnetization (M-H) tests at 300 K. As shown in Fig. 2b-d (main text) and high in-plane magnetic field test (Supplementary Fig. 5), Fe₃GaTe₂ bulk crystals show a clear out-of-plane magnetic anisotropy with in-plane saturation field $B_{sat}=4$ T and saturation magnetization $M_{sat}=32.7$ emu/g (0.3 T). The magnetic anisotropy energy density (K_u) is calculated as $\sim 4.79 \times 10^5$ J/m³ at 300 K, following the formula¹:

$$K_u = \frac{B_{sat} M_{sat}}{2\mu_0} \quad (1)$$

where M_{sat} is the saturation magnetization, B_{sat} is the saturation field, and μ_0 is the permeability in free space.

The magnetic anisotropy of Fe₃GaTe₂ few-layer nanosheet (9.5 nm) is analyzed by angle-dependent anomalous Hall effect (AHE) at 300 K. The angle (θ_B) is defined as the tilt angle between the sample plane and the magnetic field (For example, $\theta_B=90^\circ$ and $\theta_B=0^\circ$ correspond to the perpendicular and parallel magnetic field, respectively). The square hysteresis loops suggest an out-of-plane magnetic anisotropy. An in-plane magnetic field ($B>0.5$ T, $\theta_B=0^\circ$) drives the magnetization to flip in an in-plane direction, R_{xy} disappears because it is proportional only to the out-of-plane component of the magnetization. To eliminate the effect of domains, the R_{xy} value is extracted at high magnetic field ($B=4$ T), and the angle between magnetization and the sample plane, θ_M , is calculated by using the formula²:

$$\theta_M(\theta_B) = \arcsin \left[\frac{R_{xy}(\theta_B)}{R_{xy}(\theta_B=90^\circ)} \right] \quad (2)$$

The magnetic anisotropy energy density K_u is estimated by fitting the data of θ_B and θ_M with the Stoner-Wohlfarth model³. The total energy of the system is:

$$E = K_u \sin^2(\theta_M) - H_k M_{sat} \cos(\theta_B - \theta_M) \quad (3)$$

At first-order derivative:

$$\frac{\partial E}{\partial \theta_M} = 2K_u \sin(\theta_M) \cos(\theta_M) + H_k M_{sat} \sin(\theta_B - \theta_M) = 0 \quad (4)$$

The fitting curve in Fig. 2g (main text) is obtained from the above equation, and the K_u is calculated as $\sim 3.88 \times 10^5 \text{ J/m}^3$.

Supplementary Note 3. Additional magneto-transport analysis of the single-sheet Fe₃GaTe₂ nanosheets

The AHE measurements are performed under the perpendicular magnetic field at a series of temperature. In a ferromagnetic material with a uniaxial out-of-plane magnetic anisotropy, the R_{xy} can be expressed by the following formula:

$$R_{xy} = R_0 \mu_0 H + R_s M \quad (5)$$

where R_0 and R_s are normal and anomalous Hall coefficients, respectively, and M is the magnetization. For $\mu_0 H = 0$, the remanent R_{xy} , namely R_{xy}^r , is only directly proportional to the zero-field magnetization, namely spontaneous magnetization. So the onset of non-zero R_{xy}^r reflect the emergence of spontaneous magnetization, and further demonstrates the long-range ferromagnetic order (or intrinsic ferromagnetism) in Fe₃GaTe₂ nanosheet. Therefore, the T_C is determined at the onset of non-zero R_{xy}^r .²

In Supplementary Fig. 10, the normal Hall coefficient (R_0), anomalous Hall coefficient (R_s), carrier density (n), and mobility (μ) are calculated by using the formulas:

$$R_0 = \frac{\Delta R_{xy}}{\Delta B} * d \quad (6)$$

$$R_s = \frac{\rho_{xy}(0)}{M(0)} \quad (7)$$

$$n = \frac{I}{e R_0} \quad (8)$$

$$\mu = R_0 \times \sigma_{xx} \quad (9)$$

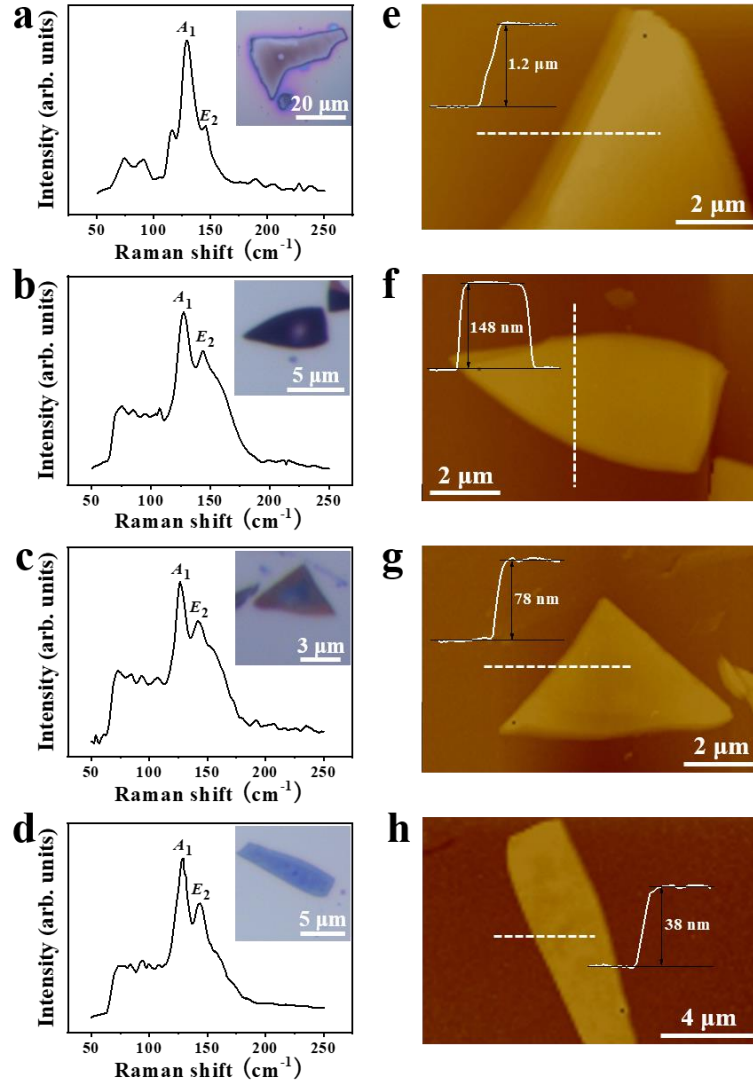
where $M(0)$ is the zero-field magnetization, σ_{xx} is the longitudinal conductivity, $e \approx 1.602 \times 10^{-19}$ C.

In Supplementary Fig. 11, the anomalous Hall conductivity (σ_{AH}) and anomalous Hall angle (θ_{AH}) are calculated by using the formulas:

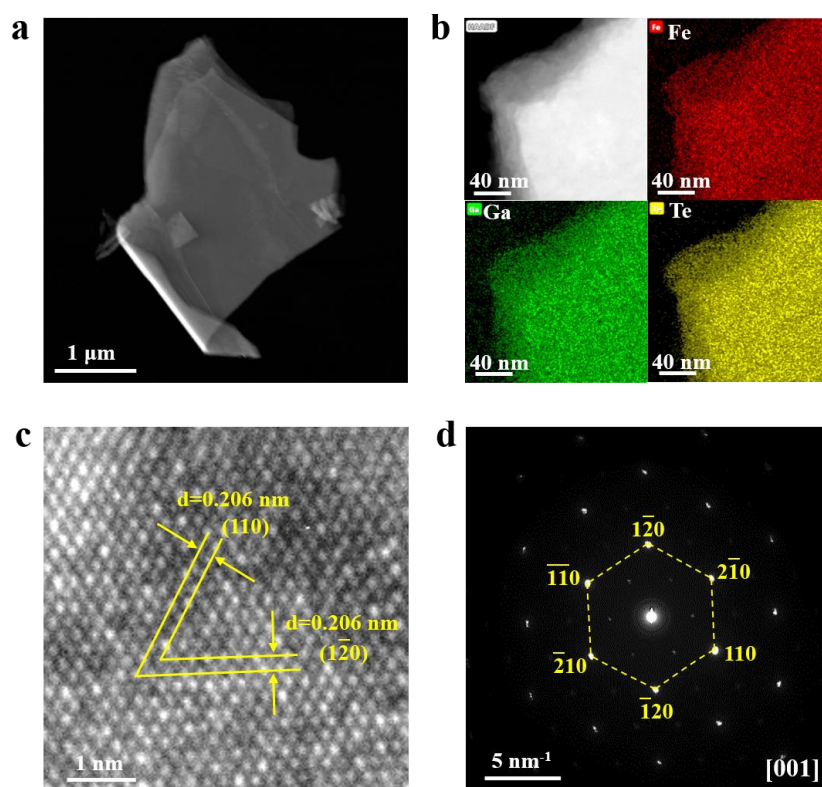
$$\sigma_{AH} = \frac{\rho_{AH}}{\rho_{AH}^2 + \rho_{xx}^2} \quad (10)$$

$$\theta_{AH} = \frac{\sigma_{AH}}{\sigma_{xx}} \times 100\% \quad (11)$$

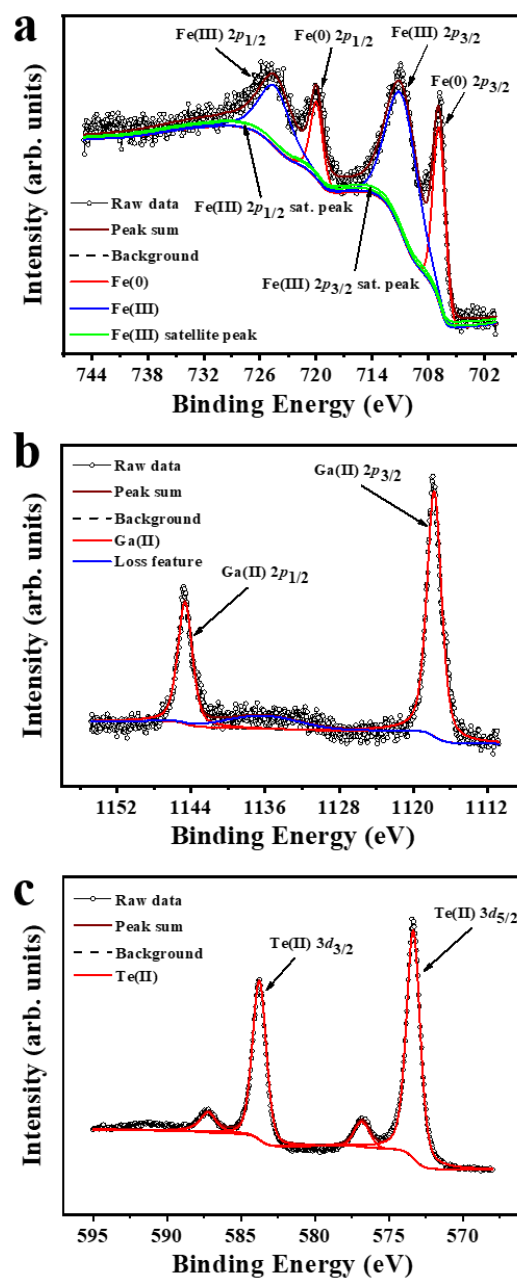
where ρ_{AH} is the anomalous Hall resistivity, ρ_{xx} is the longitudinal resistivity, σ_{xx} is the longitudinal conductivity.



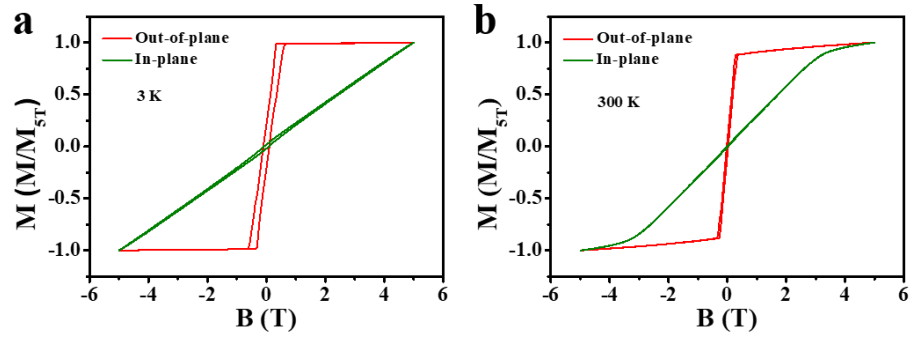
Supplementary Fig. 1. Raman characterization of the Fe_3GaTe_2 nanosheets with different thickness. (a-d) Raman spectra of the Fe_3GaTe_2 nanosheets with different thickness. Insets show the corresponding optical micrographs. (e-h) Corresponding AFM topographies and thickness of the as-tested Fe_3GaTe_2 nanosheets.



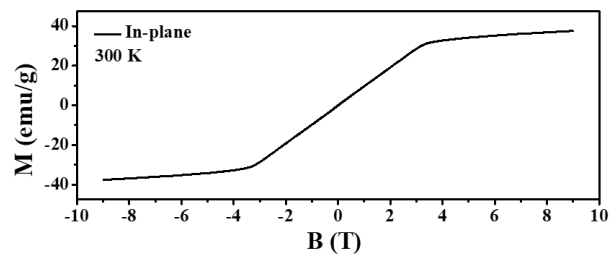
Supplementary Fig. 2. TEM characterization of a Fe_3GaTe_2 nanosheet. (a, b) Dark-field image and corresponded high-resolution element mapping image of a Fe_3GaTe_2 nanosheet. (c, d) HRTEM image and corresponded SAED pattern of the Fe_3GaTe_2 nanosheet.



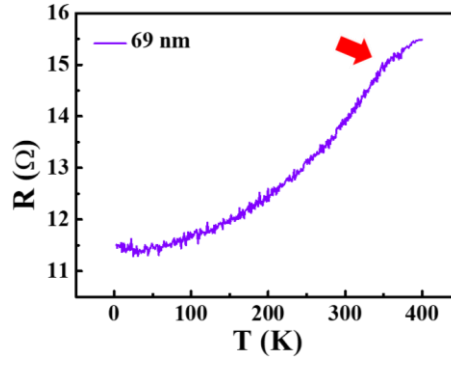
Supplementary Fig. 3. XPS analysis of the exfoliated fresh Fe_3GaTe_2 crystal surface. (a) Fe. (b) Ga. (c) Te.



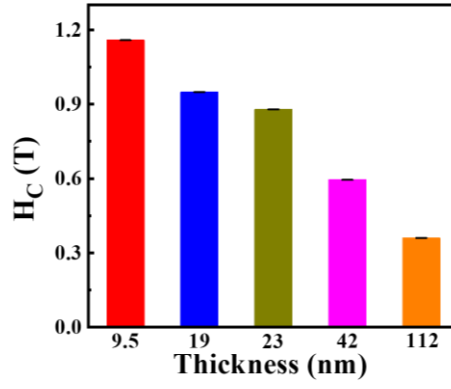
Supplementary Fig. 4. Magnetic anisotropy of Fe₃GaTe₂ bulk crystals by VSM. (a, b) M-H curves for out-of-plane and in-plane orientation with normalized moment at 3 K (a) and 300 K (b).



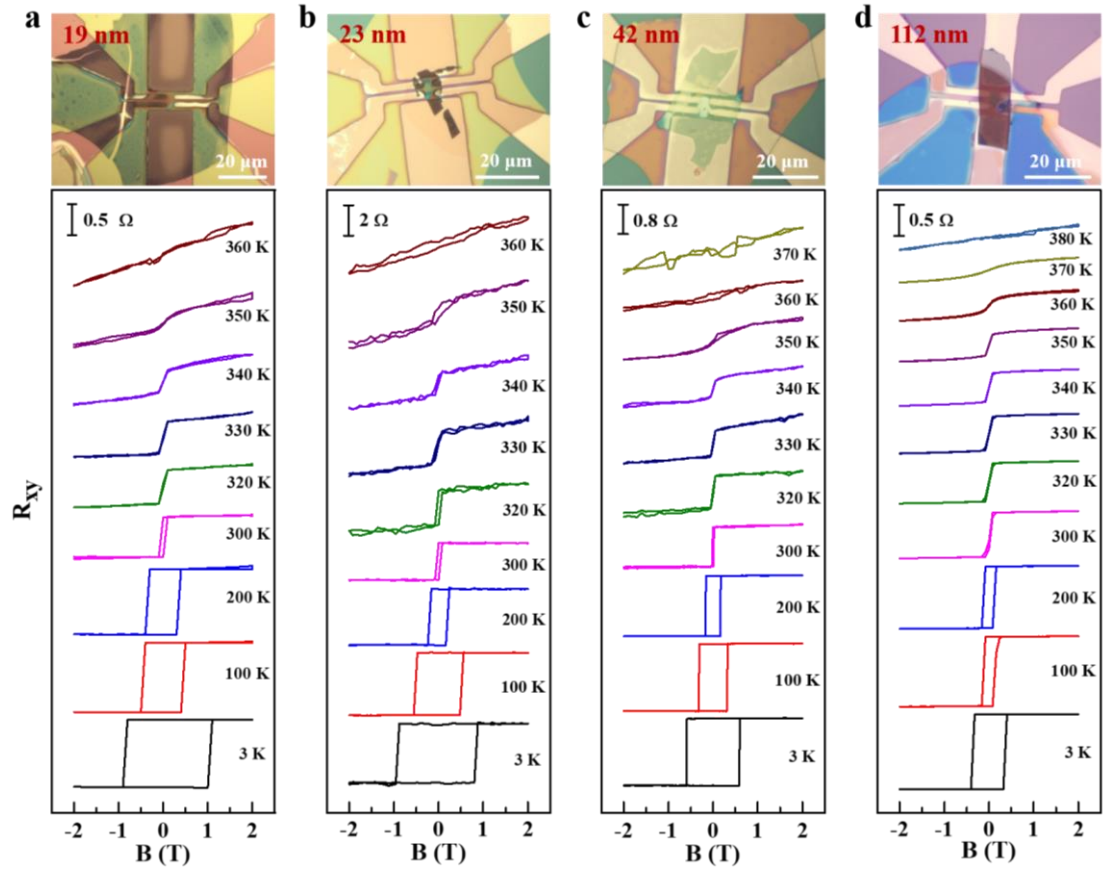
Supplementary Fig. 5. M-H curve of the Fe_3GaTe_2 bulk crystals under in-plane magnetic field of -9 to 9T at 300 K.



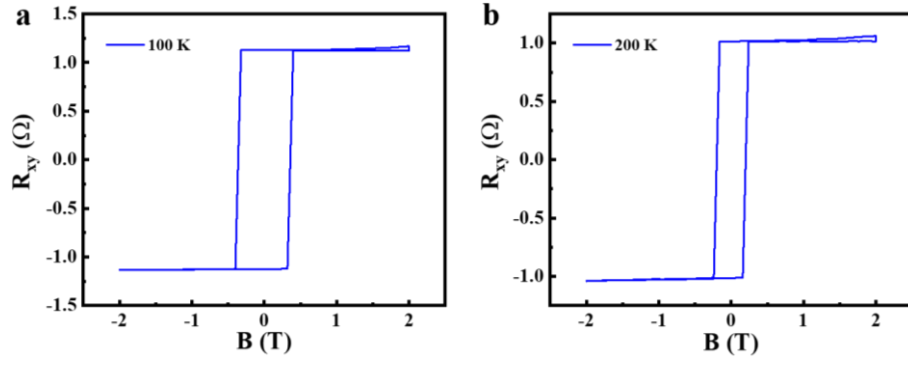
Supplementary Fig. 6. Temperature-dependent longitudinal resistance (R-T) curve of a 69 nm Fe_3GaTe_2 nanosheet from 3 to 400 K. The red arrow shows a kink, which are characteristic of the established phase transition from paramagnetism to ferromagnetism.



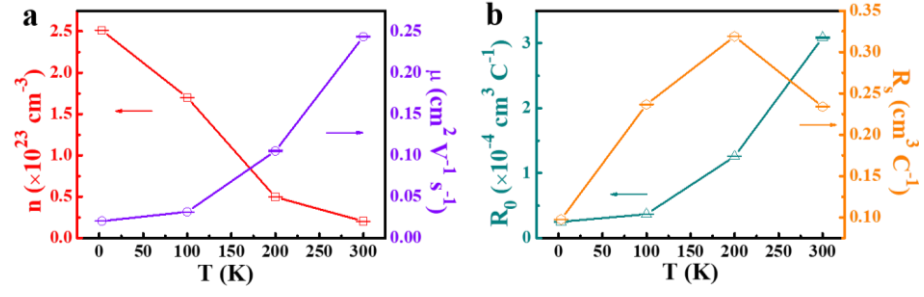
Supplementary Fig. 7. Thickness-dependent H_C histogram of Fe_3GaTe_2 nanosheets at 3 K. The data are obtained from Fig. 3d in main text. Error bars s.d., $N=25$.



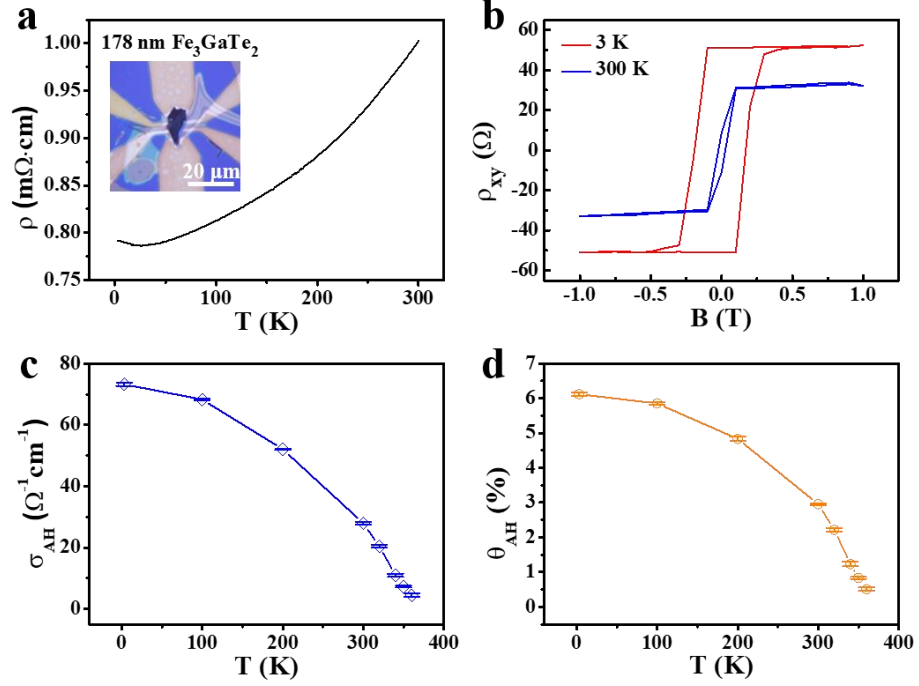
Supplementary Fig. 8. Optical images of 19 (a), 23 (b), 42 (c) and 112 nm (d) Fe_3GaTe_2 Hall devices and corresponded temperature-dependent AHE.



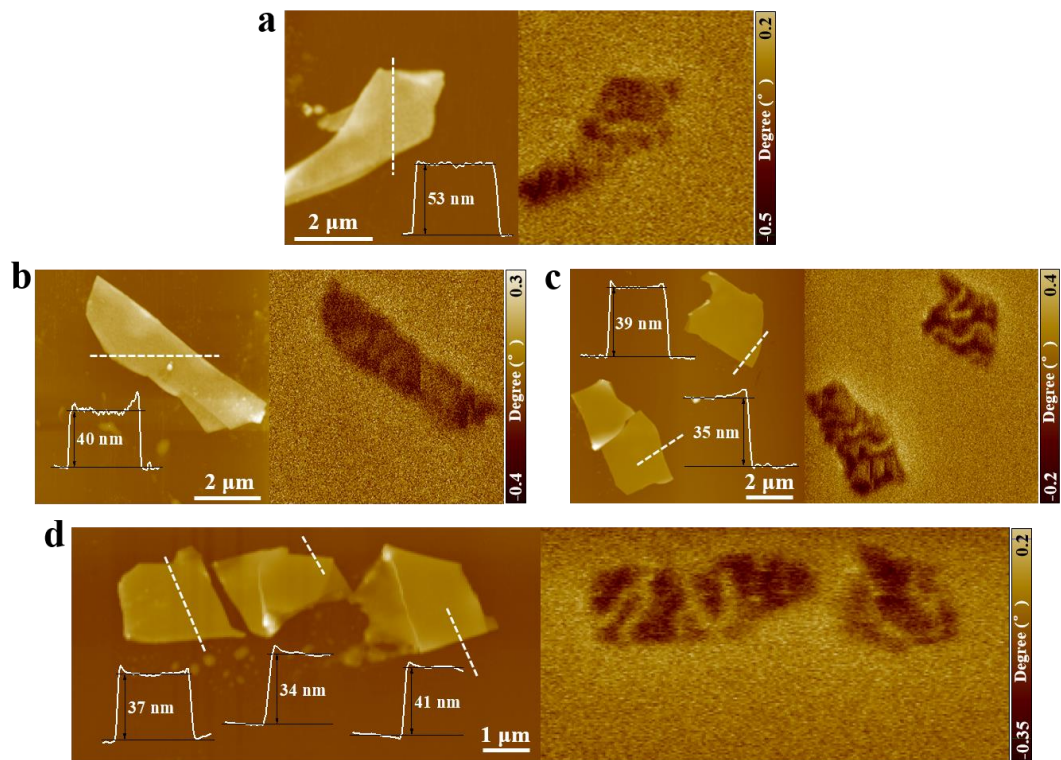
Supplementary Fig. 9. Additional AHE data in 9.5 nm few-layer Fe₃GaTe₂ nanosheet at 100 K (a) and 200 K (b). Other AHE data at 3, 300, 320-350 K can be found in Fig. 3d, 3e, 3f in main text, respectively.



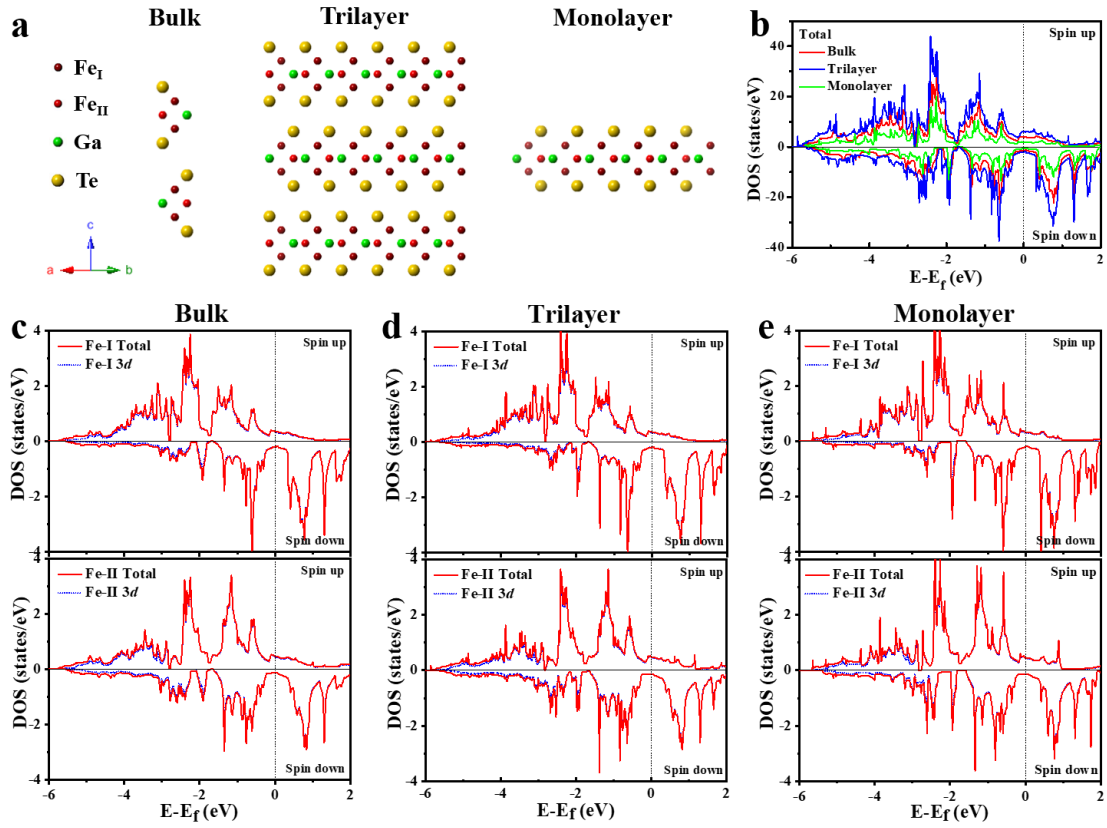
Supplementary Fig. 10. Basic parameter of a 9.5 nm Fe₃GaTe₂ few-layer nanosheet from magneto-transport measurement. (a) Temperature-dependent carrier density (n) and mobility (μ). **(b)** Temperature-dependent normal Hall coefficient (R_0) and anomalous Hall coefficient (R_s). Error bars s.d., N=25. See data in Supplementary Table 5.



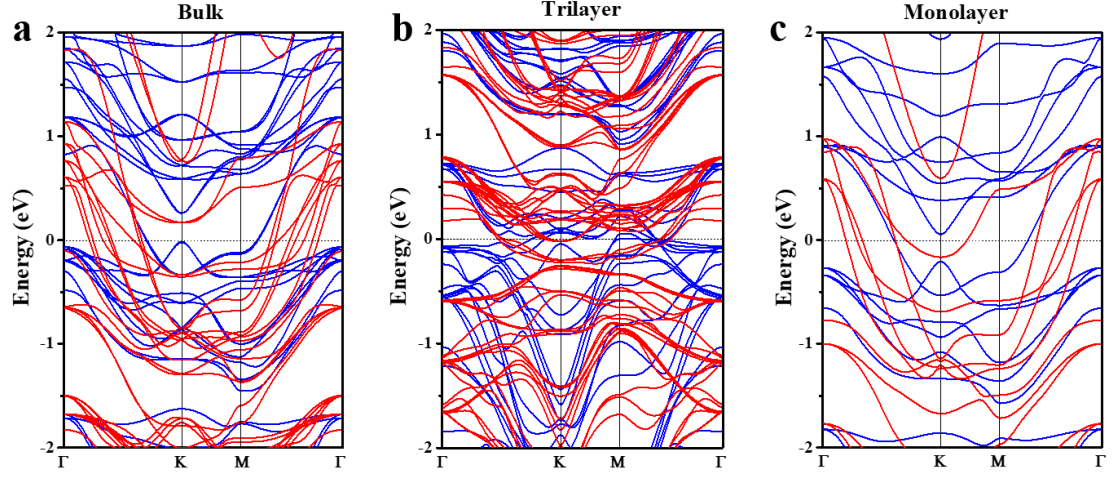
Supplementary Fig. 11. Magneto-transport measurement of a 178 nm Fe_3GaTe_2 nanosheet. (a) Temperature-dependent longitudinal resistivity (ρ - T) curve. Inset shows the optical image of the as-tested Fe_3GaTe_2 Hall device. (b) Temperature-dependent magnetic field (out-of-plane) sweeps of the Hall resistivity (ρ_{xx}) at 3 K and 300 K. (c, d) Temperature-dependent anomalous Hall conductivity (σ_{AH}) (c) and anomalous Hall angle (θ_{AH}) (d). Error bars s.d., $N=25$.



Supplementary Fig. 12. Direct imaging intrinsic ferromagnetism and magnetic domains of Fe_3GaTe_2 nanosheets with different thickness at 300 K by MFM without external magnetic field.



Supplementary Fig. 13. Spin-resolved density of states (DOSs) of the vdW Fe₃GaTe₂ ferromagnetic bulk and few-layer crystals. (a) Supercells of the bulk, trilayer and monolayer vdW Fe₃GaTe₂ crystals. (b) Total DOSs in bulk, trilayer and monolayer vdW Fe₃GaTe₂ crystals. (c-e) Total and partial DOSs of two types of Fe 3d states in bulk (c), trilayer (d) and monolayer (e) vdW Fe₃GaTe₂ crystals. The vertical dash lines denote the position of Fermi level.



Supplementary Fig. 14. Spin-resolved band structures of the bulk (a), trilayer (b) and monolayer (c) $\text{vdW Fe}_3\text{GaTe}_2$ ferromagnetic bulk and few-layer crystals. The red and blue solid lines represent the majority-spin and minority-spin bands, respectively. The horizontal dash lines denote the position of Fermi level.

Supplementary Table 1. Element analysis of the Fe₃GaTe₂ by EDS and XPS measurements.

Method	Elements	Atomic Conc (%)	Error (%)
EDS-1	Fe K	48.75	-
	Ga K	18.95	-
	Te K	32.30	-
EDS-2	Fe K	51.01	-
	Ga K	16.52	-
	Te K	32.47	-
EDS-3	Fe K	50.09	-
	Ga K	17.09	-
	Te K	32.82	-
EDS-Ave	Fe K	49.95	Sd. 0.93
	Ga K	17.52	Sd. 1.04
	Te K	32.53	Sd. 0.22
XPS	Fe 2 <i>p</i>	49.90	3.02
	Ga 2 <i>p</i>	17.60	1.44
	Te 3 <i>d</i>	32.50	2.02

Notes for Supplementary Table 1:

For the EDS measurement, we measured 3 points on an exfoliated Fe₃GaTe₂ thin nanosheet. The average atomic ratio of Fe: Ga: Te was 2.94±0.05: 1.03±0.06: 1.91±0.01, which is very close to 3: 1: 2. Additionally, we also performed the XPS measurement on an exfoliated fresh Fe₃GaTe₂ crystal surface. The atomic ratio of Fe: Ga: Te was 2.94: 1.04: 1.91, which is also very close to 3: 1: 2. The above EDS and XPS measurements were performed in the vacuum TEM and XPS chambers, respectively.

Supplementary Table 2. Saturation magnetic moment (M_{sat}) and coercivity (H_C) of bulk Fe₃GaTe₂ with out-of-plane magnetic field at different temperatures.

T (K)	M_{sat} (emu/g)	M_{sat} (μ B/Fe)	H_C (Oe)
3	57.18 ($\pm 3.12 \times 10^{-4}$)	1.68 ($\pm 2.75 \times 10^{-5}$)	1014.19 (± 0.62)
100	55.39 ($\pm 5.29 \times 10^{-4}$)	1.63 ($\pm 4.67 \times 10^{-5}$)	378.23 (± 0.09)
200	50.15 ($\pm 1.09 \times 10^{-4}$)	1.47 ($\pm 9.61 \times 10^{-5}$)	287.87 (± 0.36)
300	40.11 ($\pm 2.82 \times 10^{-4}$)	1.18 ($\pm 2.49 \times 10^{-5}$)	248.71 (± 0.58)
350	31.05 ($\pm 3.33 \times 10^{-4}$)	0.91 ($\pm 2.94 \times 10^{-5}$)	209.13 (± 0.17)

Supplementary Table 3. Ferromagnetic properties of Fe₃GaTe₂ in comparison with other vdW intrinsic ferromagnetic crystals.

Materials	Type	Easy axis	M_{sat} (uB)	T_C (K)	Ref.
CrI ₃	Semi	OOP	3.1 @ 2K&6T	61	4
Cr ₂ Ge ₂ Te ₆	Semi	OOP	2.39 @ 2K&5T	66	5
			2.4 @ 5K&1T	61	6
CrSiTe ₃	Semi	OOP	3.08 @ 2K&6T	33	7
CrBr ₃	Semi	OOP	2.85 @ 2K&2T	33	8
VI ₃	Semi	OOP	1.25 @ 1.8K&9T	49	9
			2.47 @ 1.8K&5T	50	10
			1.31 @ 2K&7T	50	11
AgVP ₂ Se ₆	Semi	OOP	1.38 @ 2K&1.5T	18.5	12
CrTe ₂	Metal	IP	1.81 @ 2K&7T	310	13
Cr _{1.33} Te ₂	Metal	OOP	2.66 @ 3K&7T	312	14
Fe ₃ GeTe ₂	Metal	OOP	1.6 @ 10K&5T	220	15
Fe ₄ GeTe ₂	Metal	OOP	1.8 @ 2K&1T	270	16
Fe ₅ GeTe ₂	Metal	IP	2.1 @ 3K&6T	317	17
Fe _{1/4} TaS ₂	Metal	OOP	3 @ 2K&7T	160	18
Cr _{1/3} TaS ₂	Metal	IP	1.85 @ 10K&5T	110	19
Cr _{1/3} NbS ₂	Metal	IP	2.9 @ 4.2K&1.5T	127	20
Mn _{1/3} TaS ₂	Metal	IP	2.68 @ 2K&6T	70	21
Ni ₂ FeGaTe ₂	Metal	-	0.6 @ 4K&5T	75	22
Fe _{2.7} AsTe ₂	Metal	-	0.1 @ 2K&7T	123	23
Fe _{2.9} Ge _{1-x} As _x Te ₂	Metal	-	1.1-0.25 @ 10K&5T	177-33	24
(Fe,Ni)GeTe ₂	Metal	-	2.11 @ 1.8K&5T	279	25
Fe₃GaTe₂	Metal	OOP	1.68 @ 3K&5T	367	This work

Notes for Supplementary Table 3:

OOP is out-of-plane, IP is in-plane. Fe₃GaTe₂ possess record-high T_C compared with other vdW intrinsic ferromagnets, which is significant for room-temperature vdW-integrated spintronic device. This table only compares data for bulk crystals.

Supplementary Table 4. Comparison of magnetic anisotropy energy density K_u in some classical PMA systems at room temperature from literatures.

Structure	Materials	Sample Information	K_u ($\times 10^5$ J/m ³)	Ref.
Non-vdW	CoFeB	1.3 nm thin film	2.10	¹
	Co ₂ FeAl	2 nm thin film	1.30	²⁶
	Fe ₂ CrSi	0.8 nm thin film	2.80	²⁷
VdW	CrTe ₂	7 layers thin film	0.49	²⁸
	Cr-CrTe ₂	Bulk crystals	0.76	¹⁴
	Fe₃GaTe₂	Bulk crystals	4.79	This
		9.5 nm nanosheet	3.88	work

Notes for Supplementary Table 4:

The K_u in Fe₃GaTe₂ is comparable to the conventional ferromagnetic thin film and one order of magnitude larger than recently discovered vdW ferromagnetic crystals. The large K_u value ($\sim 4.79 \times 10^5$ J/m³ for bulk crystals and $\sim 3.88 \times 10^5$ J/m³ for 2D few-layer nanosheet) in Fe₃GaTe₂ indicate the strong and robust PMA, which is of crucial importance to maintain high thermal stability for long-term data retention in high-density magnetic data-storage devices.

Supplementary Table 5. Normal Hall coefficient (R_θ), anomalous Hall coefficient (R_s), conductivity (σ), carrier density (n) and mobility (μ) of the 9.5 nm Fe₃GaTe₂ few-layer nanosheet at different temperatures.

T (K)	R_θ (cm ³ C ⁻¹)	R_s (cm ³ C ⁻¹)	σ (S cm ⁻¹)	n (cm ⁻³)	μ (cm ² V ⁻¹ s ⁻¹)
3	2.49×10^{-5}	0.09759	830.74	2.51×10^{23}	0.02
100	3.68×10^{-5}	0.23664	861.07	1.70×10^{23}	0.03
200	1.26×10^{-4}	0.3189	838.35	4.98×10^{22}	0.11
300	3.09×10^{-4}	0.23396	789.30	2.03×10^{22}	0.24

Supplementary Table 6. Electric conductivity (σ_{xx}) and anomalous Hall angle (θ_{AH}) of Fe₃GaTe₂ in comparison with other ferromagnets.

Materials	Sample Information	σ_{xx} ($10^4 \Omega^{-1} \text{ cm}^{-1}$)	θ_{AH} (%)	Ref.
Co ₃ Sn ₂ S ₂	Single crystal, 130 K	0.57	19.8	29
Fe ₃ GeTe ₂	Single crystal, 5 K	0.67	9.0	30
Fe _{0.28} TaS ₂	Single crystal, 4.2 K	0.90	3.7	31
(Ga, Mn)As	50 nm thin film, 2 K	0.22	6.6	32
Fe-Co alloy	0.25 wt% Co-Fe, 4.2 K	438.00	1.5	33
Fe _{0.99} Co _{0.01}	Bulk, 5 K	110.00	1.2	34
Fe ₃ Sn ₂	Single crystal, 10 K	12.50	1.0	35
Fe	Thin film, 5 K	4.36	2.6	36
Gd	Thin film, 5 K	50.00	0.2	36
CrTe ₂	170 nm, 2 K	133.08	5.5	37
	170 nm, 300 K	<1.00	<0.5	
Fe₃GaTe₂	178 nm, 3 K	0.12	6.1	This
	178 nm, 300 K	0.09	3.0	work

Supplementary Table 7. Calculated atomic magnetic moment (μ_B) of the bulk, trilayer and monolayer Fe_3GaTe_2 .

Element	Bulk	Trilayer	Monolayer
Fe-I	2.356	2.327	2.373
Fe-II	1.459	1.454	1.421
Ga	-0.114	-0.112	-0.113
Te	-0.09	-0.088	-0.089

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