

Robust ferromagnetism in wafer-scale Fe₃GaTe₂ above room-temperature

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This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

In this manuscript, the authors successfully synthesized few-layer Fe₃GaTe₂ via MBE method. The Curie temperature (T_c) of this van der Waals (vdW) magnet is much higher ($T_c=457K$) than that from previously report and the T_c of monolayer Fe₃GaTe₂ in this work is higher than room-temperature. The room-temperature ferromagnetism in monolayer vdW magnet is already reported in MnSe case [Nano Lett. 18, 3125 (2018)], considering Fe₃GeTe₂ and Fe₅GeTe₂ case that their monolayer exhibit lower T_c compared to its bulk crystal, high T_c of monolayer Fe₃GaTe₂ is not only scientifically interesting, but also can be utilized in realization of all-vdW spintronic device functioning in room-temperature. Given that, the verification of robust FM characteristic of Fe₃GaTe₂ itself deserves to be published in Nature Communication. However, several critical issues related to their experimental condition and obtained data must be clarified. If those issues are not handled properly and the quality of the manuscript isn't improved, even though the result itself is intriguing, I don't recommend to publish this work in Nature Communication.

Comments are listed below,

1. There is non-negligible mismatch between lattice parameters of Fe₃GaTe₂ listed in this work and those from previous report. The lattice parameters from XRD measurement and TEM measurement also don't match to each other. This inconsistency may originate from the thickness of samples, since the thicker sample is less affected by strain. Also, as mentioned by the authors, non-negligible strain effect in those MBE-grown Fe₃GaTe₂ exists through all samples. However, the effect of this strain to the magnetic properties hasn't been mentioned clearly in this manuscript. It is well-known that strain can greatly modulate the magnetic properties of vdW magnets, this strain effect on the magnetic properties of Fe₃GaTe₂ should be clarified via DFT calculation or measuring their magnetic properties in other substrate.
2. How the large lattice mismatch is believed to trigger the vdW growth mode of Fe₃GaTe₂?
3. The magnetic moment of magnetic material is proportional to its volume. Therefore, the magnetic moment of 9 ML should be at least 9 times higher than that of 1 ML, or at least there should be deviation between them. However, in fig. 3a and fig. 3d, there is no deviation between them at applied field, which seems to be unrealistic. Also, due to less magnetic volume, the M-T curve of 1 ML Fe₃GaTe₂ is supposed to be noisy compared to 9 ML, like fig. 3c and fig. 3f, which is the opposite situation of fig. 3a and fig. 3d.
4. It is very unrealistic that saturation magnetic moment per unit mass of 1 ML is larger than 9 ML. It is widely known that interlayer interaction in vdW materials is considerably weak, still magnetic properties of vdW magnets are greatly affected by the number of layer. This tendency is experimentally shown in several works, the critical temperature of vdW magnets are decreasing with decreasing number of layer, which indicate the weakening of magnetic properties due to thermal fluctuation. The increase of saturation magnetic moment in this manuscript is the opposite situation. Can the authors give any supporting data or simulation result to back up their observance? If saturation magnetic moment is mis-obtained, one of possible reasons is maybe the uncertainty of volume or mass obtained during calculating it. Since MBE-grown Fe₃GaTe₂ is directly grown on substrate, it is very challenging to measure the mass or the volume of thin film precisely. This uncertainty may result to inaccurate value of saturation magnetization. Anyhow, the authors should clarify this issue, 1) Is the increase of saturation magnetization not mis-obtained result? 2) If so, then what is origin of increase of saturation magnetization at 1 ML.
5. How can the decrease of coercivity from 9 ML to 1 ML be interpreted to the existence of weak interlayer interaction? For me, the weakening of coercivity seems to reveal the non-negligible interlayer interaction between layers. This sentence doesn't harmonize not only with common sense, but also with the flow of the manuscript. Therefore, this sentence must be deleted, in my opinion.
6. The magnetic properties of Fe₃GeTe₂ and Fe₅GeTe₂ are shown to be greatly affected by proximity effect. Therefore, the

argument what authors have written related to substrate effect is almost not persuasive to me. To rule out the concern related to substrate effect, the authors is recommended to grow Fe₃GaTe₂ on other substrate, which will rule out substrate and strain effects.

7. There are multiple plateaus in fig. 3e. It is presumably due to the formation of multi-domain, but the authors didn't mention about it in the manuscript. This step-like hysteresis loop is hardly observed in typical FM hysteresis loop, so the authors should clarify the origin of this behavior.

8. In fig. 3i, negative stray field is dominant in upper region and positive stray field is comparably dominant in lower region. This result can be misinterpreted as that Fe₃GaTe film possesses non-uniform magnetic domain structure. The authors explained that stray field mapping is shown to prove robust FM characteristic even in room-temperature, but I think presented magnetization measurement data and transport data is enough. Thus, I recommend to remove figs. 3h and i or present mapping data which is measured at the magnetic field strong enough to saturate Fe₃GaTe₂ film.

9. The authors mentioned that T_c of 1 ML is higher than 400 K, based on anomalous Hall effect (AHE) measurement. However, in fig. 3c, RH-H of 1 ML at 400 K is almost linear and AHE is hardly observed. Therefore, the T_c of 1 ML is lower than 400 K, which can also be proved by fig. 3d. Based on AHE measurement, the T_c of 1 ML is in the range of 300~350 K. Therefore, the argument related to T_c of 1 ML Fe₃GaTe₂ based on AHE measurement should be revised.

10. The RH values of 9 ML Fe₃GaTe₂ is much smaller than that of 1 ML Fe₃GaTe₂ in fig. 4d. There is no direct correlation between anomalous Hall resistance and magnetization, still one can misinterpret that the magnetization of 1 ML Fe₃GaTe₂ is larger than that of 9 ML Fe₃GaTe₂. Therefore, I strongly recommend to convert RH to anomalous Hall conductivity, which is directly proportional to magnetization of FM material.

Minor issues are listed below,

1. The authors mention the bulk lattice parameters from [Nat. Commun. 13, 5067 (2022)] in manuscript, but didn't refer this previous work in first sentence of second paragraph in pg. 2. The authors should refer this paper in this specific sentence.
2. How did the authors define the T_c? It is pointed in fig. 3a for the 9ML Fe₃GaTe₂, it is too roughly defined. There are many ways to define T_c, none of them are explained in this paper. The authors should decide at least one criterion to define the T_c of Fe₃GaTe₂ films.
3. Based on [Nanoscale, 15, 11290 (2023)], one can realize that c lattice parameter is critical to magnetic property of Fe₃GaTe₂. The mismatch between c lattice parameters from different measurement method is acceptable, still it seems better for the authors to mention that c lattice parameter may have some error range due to different measurement is acceptable with referring it for excuse.
4. How did the authors calculate the lattice parameter of 9 ML Fe₃GaTe₂ film?
5. What is the origin of up-turn of magnetization in M-T curve at low temperature region in figs. 3a and d? It may originate from substrate, but it wasn't mentioned in the manuscript. It's better for the authors to mention the origin of it.
6. Refs. 7 and 8 are duplicated at refs. 25 and 34, respectively.
7. The authors mentioned that SQUID is indirect experimental method. However, SQUID measurement is one of the experimental methods which can directly measure magnetic moment of the sample. I cannot agree with this sentence, except it is reflecting macroscopic magnetic properties. The authors are recommended to re-write this sentence, removing indirect experimental method and only mentioning about the limit of the macroscopic measurement.

Reviewer #2

(Remarks to the Author)

In this manuscript, the authors grow Fe₃GaTe₂ (FGT) thin films by using MBE for the first time. The as-grown FGT thin films show very high T_c and large PMA, which is very crucial for 2D spintronics. They try to characterize and discuss the structure and magnetism of FGT films and some results are presented. However, I have important concerns and doubts regarding the magnetism and crystal characterizations, and some statements are lack evidence or references. Meanwhile, the underlying mechanisms about high T_c and variation trend with thickness are not clearly demonstrated. Therefore, I cannot recommend this manuscript for publication before the authors reply satisfactorily to the comments below.

Major issues:

1. In Fig 2a and 2c, cross-sectional TEM images are presented. Are they HRTEM images or HAADF images? In the caption, the authors claim "HRTEM" image, but in the manuscript, the authors claim "HAADF" image, and I am confused about it. To my knowledge, they should look like HAADF images, as the authors clearly indicate the atoms in Fig 2c, and Te atoms have the brightest contrast due to the largest atomic number. However, in Fig 2a, the Te capping layer and F-mica substrate (with relatively small atomic numbers) are clearly observed and bright, but all FGT region is very dark. I am very confused about it because the atoms in FGT are heavier than that in F-mica, and there are many Te layers contained in FGT.

----Moreover, the Fe and Ga atoms in Fig 2c are not very clear. I think they should provide a clearer cross-sectional TEM image to replace current Fig 2c.

----What is the atomic ratio of the as-grown FGT film? EDX test can provide a specific ratio of Fe, Ga, and Te.

2. In Fig 3a, the authors identifies the temperature at which the magnetization decreased to zero as T_c. Based on this, the T_c of the 1 ML and 3 ML samples seems to be ~370 K (Fig 3d and Fig S1), and thus the AHE in the 1-4 ML FGT films at 400 K is negligible (Fig 4c and Fig S6a-c). But the hysteresis loops still exists at 400 K (Fig 3e and Fig S3a). Can the authors give a reasonably explanation? I think the current identification method for T_c is not ideal. Can the author use a unified standard method to determine T_c from M-T or AHE curves in each samples (e.g. dM/dT-T plots or Arrott plots)?

3. The T_c of as-grown 9 ML and 1 ML FGT films are respectively much higher than the T_c of exfoliated ~9.5 nm FGT flake

($T_c \sim 350$ K, Nat. Commun. 13, 5067, 2022) and monolayer one ($T_c \sim 200$ K, Nano Lett. 2024, 24, 993) in previous works. What causes such higher T_c in this work?

----Initially, I think it is attributed to the tensile strain due to the lattice mismatch between few-layer FGT films and substrate. Because the larger in-plane lattice constant of as-grown FGT films compared with bulk value demonstrates the existence of tensile strains. However, the authors think the substrate effect on the ferromagnetic phenomena is negligible because of the weak layer-dependent T_c , and thus the FGT films they grown can be regarded as an isolated magnet. I am very curious about this. If they are "isolated magnets", they should have ferromagnetic properties similar to exfoliated FGT flakes (e.g. T_c). Thus, I think the underlying mechanisms of such high T_c should be fully addressed, and additional results for supporting their mechanisms are needed.

4. The weak layer-dependent T_c is an unusual phenomenon in vdW magnets especially in 2D limit. The authors use weak interlayer exchange coupling to explain the small variation of T_c with thickness in FGT, and they use isostructural Fe_3GeTe_2 as an example to support their opinion. However, for Fe_3GeTe_2 , the variation of T_c with thickness is significant, such as from 175 K in 10 ML to 75 K in 1 ML (a decrease of 57%, npj 2D Mater Appl 4, 33, 2020). Even for FGT flake, the T_c is from ~ 300 K in 10 nm to ~ 200 K in 1 nm (a decrease of 33%, Nano Lett. 2024, 24, 993). In contrast, the larger thermal fluctuations at higher temperature should have a larger impact on T_c , but the T_c of FGT films persist over 400 K and only decreased by $\sim 12\%$ from 9 ML to 1 ML. Thus, I am not convinced this explanation for weak layer-dependent T_c , or the authors should provide more solid evidences to explain this unusual phenomenon.

5. The authors use the saturation magnetic moment per unit mass (emu/g) for comparison in the manuscript. How is the mass of ultrathin FGT films obtained? Moreover, why does the M_s increases more than twice at 300 K as the thickness decreases from 9 ML to 1 ML? Does this phenomenon indicates the influence of substrate effect on ferromagnetism?

6. The author believes that the decrease of H_c with decreasing thickness indicates weak interlayer coupling. Can the author provide a more detailed explanation to reveal the relationship between thickness-dependent H_c and interlayer coupling strength?

7. For the NV spin magnetometry tests, the magnetic domain structures of 1 ML FGT seems to be different from that of 9 ML FGT. Can the authors comment on it? Moreover, under zero bias magnetic field, the stray magnetic field intensity is almost randomly and uniformly distributed, but when a 500 Oe magnetic field is applied, their distribution becomes very uneven (in the Fig 3i, the upper half is blue, and the lower half is red), which is very curious. How do the authors explain this? Also, I suggest the authors should continue to increase the bias magnetic field to saturate the 1 ML FGT and demonstrate a complete field-driven domain evolution.

8. The authors claim "The anomalous temperature dependence of carrier density and mobility could be related to the increase in density of state at Fermi level and effective mass of electron due to the variation of band structure with temperature." Can the authors provide some evidences to support this statement?

Minor issues:

1. I suggest they provide a photograph of as-grown wafer-scale FGT thin film (at least 1 inch) to support the so called "wafer-scale" in the title.

2. Why does the ZFC moments in Fig S1 become negative within a certain temperature range?

3. The calculation method for PMA constants should be disclosed.

4. Is there any literature indicating that FGT is an itinerant ferromagnet? Please cite it.

Reviewer #3

(Remarks to the Author)

In this study led by Shuxiang Wu et al., the investigation of MBE-grown Fe_3GaTe_2 monolayer and multilayer ferromagnetic thin films is presented. However, the claim of a significantly higher Curie temperature (T_c) above 400 K for Fe_3GaTe_2 compared to bulk samples and other 2D ferromagnets requires further clarification. This is especially notable given that bulk Fe_3GaTe_2 has a T_c of 380 K, raising concerns about the assumption that thin films inherently possess smaller T_c . Moreover, the 9ML sample (457 K) in this manuscript exhibits a considerably higher T_c than the 1ML sample (400 K), adding complexity to the understanding of T_c evolution with thickness. The authors delve into the lower T_c in 2D ferromagnets on the theoretical side as well.

The investigation of potential excess Fe in Fe_3GaTe_2 thin films as a contributing factor to the observed higher T_c warrants exploration. Detecting an (e.g up to 20%) excess of Fe using the provided characterization methods, like STEM, might pose challenges due to potential limitations in resolving excess Fe. The exceedingly high carrier densities, akin to copper metal, as measured in this manuscript, could also be attributed to an excess of Fe. If the elevated T_c is indeed linked to excess Fe, it questions the validity of the paper's claim of "intrinsic" ferromagnetism and high Curie temperatures.

To enhance the manuscript, the inclusion of RHEED oscillations during film growth would be valuable. Additionally, comparing the period of these oscillations with the calibration could provide further insights. Given the potential instability in the supply of Fe from the ebeam source, this aspect should be considered and discussed, acknowledging its impact on the

results.

Moreover, for a more comprehensive understanding, control samples subjected to different Fe supplies during the growth processes should be examined. This could provide valuable data on how variations in Fe supply influence the properties of the thin films.

Furthermore, the authors are advised to conduct X-ray Absorption Spectroscopy (XAS) on Fe edges of the thin films. Utilizing Fe valences of 2+ and 3+ in Fe₃GaTe₂, XAS has the potential to identify excess Fe, which may be closer to Fe 0+, offering valuable insights into its quantity relative to Fe in the films.

Additional points for improvement:

1. Impact of Capping Layers on NV Center Measurements:

If the NV center measurements are conducted ex situ, it is crucial to discuss the potential impact of capping layers on these measurements.

A brief commentary on how capping layers might affect the accuracy and reliability of NV center measurements would enhance the comprehensibility of the study.

2. Arrott Plot Analysis:

Including Arrott plot analysis is recommended for an accurate determination of the Curie temperatures.

Arrott plots can provide valuable insights into the ferromagnetic phase transitions and improve the reliability of the reported Curie temperatures.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

I would like to sincerely thank the authors for their careful consideration of the referees' comments and for additional works have been done. The manuscript has somewhat improved compared to submitted version. Still, I have several major comments that detrimentally affect the quality of paper and hinder me from recommending this manuscript to be published in Nature Communications. Also, there are still many typos and mistakes with figures and numbering of them. Therefore, I would like to recommend them to fully rewrite the manuscript, references, and supplementary information. These issues can be thought to be trivial, considering the high standard of Nature Communications, the authors should also care to these non-scientific issues with great attention. If these kind of issues are found again, even though their work deserves to be published in Nature Communications, I would not recommend it.

Major comments

1. In the 2nd paragraph of page 8, the authors wrote that "A unique nature of layered vdW ferromagnets is that interlayer interactions are comparatively weak, ~". In the 1st paragraph of page 9, however, the authors mentioned that "~the corresponding H_c dramatically decreases from 2360 Oe to 230 Oe at 300K, revealing large interlayer interaction." These two sentences are not aligned to each other, and the authors must revise them. I guess, it may be more plausible to argue that the origin of decrease of H_c during decreasing thickness is due to the weakening of magnetic anisotropy or others.
2. The authors must provide the AFM measurement data of samples with various thickness. The authors have confused the unit cell and monolayer and I think it is better to clarify this issue by providing AFM images.

Minor comments

1. The electrical measurement data is shown in supplementary figure 8, not 6.
2. The resistivity of Fe₃GaTe₂ with various devices are shown in supplementary figure 10, not 9.
3. The author name of ref. 15 is miswritten.
4. The color legend for figure 4e is not consistent with its graph. Please change the color.
5. The caption for Supplementary figure 14 is not fully written. Also, the color legend for supplementary figure 14.c is different with its graph.

Reviewer #2

(Remarks to the Author)

I acknowledge that the authors have made many efforts to improve their paper. Indeed, some important concerns have been resolved. However, there are still some important issues that should be addressed. The comments are as follow:

(1) The authors use the dM/dT plot to determine the T_c from M-T curves, which is good. However, if the T_c of 1 u.c. FGT is about 350 K (Figure 3d), why does the clear hysteresis loop still maintain at 400 K (Figure 3e)?

(2) If the authors acknowledge that the significantly higher Curie temperature of epitaxial FGT comes from strain effects, the title should not be referred to as "intrinsic ferromagnetism". It is not correct to claim intrinsic since there is strong extrinsic effect. The relevant statements in the main text should also be revised.

(3)The proof for the existence of strain effect and the effect to induce the high T_c is not solid. More experimental evidence for the existence of strain effect should be given.

Reviewer #3

(Remarks to the Author)

The authors have revised their explanation for the enhanced Curie temperatures (T_C) of Fe_3GaTe_2 (FGT), moving away from the interlayer coupling hypothesis. The new DFT calculations suggesting that tensile strain leads to an enhanced T_C are reasonable, yet there are several details that require further consideration and clarification.

First of all, I would like to thank the authors for attempting the Arrott plot analysis, which has corrected the T_C for the 1 u.c. sample from 380K to 345K. It would be beneficial to apply this method to all samples to ensure consistency and accuracy of the T_C values.

The observed increase in T_C from 1 u.c. to 9 u.c., and the peak T_C at 9 u.c., remains unclear. This aspect is a key highlight in the abstract and warrants a thorough explanation. Given that strain typically decreases with increasing thickness, it is crucial to examine the relationship between T_C , the number of u.c., and the strain percentage in detail for all 9 thickness. A table summarizing these parameters for all thicknesses would be necessary to elucidate this relationship. Furthermore, it is important to explore and discuss any additional factors beyond strain that could contribute to the observed enhancement in T_C . Such a discussion would provide a more comprehensive understanding of the underlying mechanisms at play.

Version 2:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors addressed all of the concerns well, and it is now publishable.

Reviewer #2

(Remarks to the Author)

The authors have addressed all the questions and the paper can be accepted now.

Reviewer #3

(Remarks to the Author)

The authors have adequately addressed my previous comments in this version.

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Reply to reviewers' comments

Manuscript Number: NCOMMS-23-51330

We are appreciative of the constructive suggestions and comments from all the reviewers. Accordingly, we have revised the manuscript carefully to improve the quality of the manuscript. Our point-by-point Replies are presented below in detail. we also provide a marked version of the revised manuscript in which all the changes are highlighted in yellow.

Reviewer #1

Comment 1: *In this manuscript, the authors successfully synthesized few-layer Fe₃GaTe₂ via MBE method. The Curie temperature (T_c) of this van der Waals (vdW) magnet is much higher ($T_c=457K$) than that from previously report and the T_c of monolayer Fe₃GaTe₂ in this work is higher than room-temperature. The room-temperature ferromagnetism in monolayer vdW magnet is already reported in MnSex case [Nano Lett. 18, 3125 (2018)], considering Fe₃GeTe₂ and Fe₅GeTe₂ case that their monolayer exhibit lower T_c compared to its bulk crystal, high T_c of monolayer Fe₃GaTe₂ is not only scientifically interesting, but also can be utilized in realization of all-vdW spintronic device functioning in room-temperature. Given that, the verification of robust FM characteristic of Fe₃GaTe₂ itself deserves to be published in Nature Communication. However, several critical issues related to their experimental condition and obtained data must be clarified. If those issues are not handled properly and the quality of the manuscript isn't improved, even though the result itself is intriguing, I don't recommend to publish this work in Nature Communication.*

Reply: We thank the reviewers for the positive evaluation of our work. Following the referee's constructive suggestions, the manuscript have been extensively improved. In the revised manuscript, to avoid making confusion on the definition of monolayer materials via MBE and mechanical exfoliation, we changed the term of "monolayer (ML)" to "unit cell (u.c.)". We have conducted density functional theory (DFT) calculations using the Vienna Ab-initio Simulation Package (VASP) software and classical Heisenberg model Monte Carlo (MC) simulations implemented in the VAMPIRE software package (*Physical Review B* **91**, 144425 (2015)) to verify the strain

effect on the enhancement of T_c . We have retested the SQUID measurements and normalized the data for magnetic properties. In addition, we have precisely determined the Curie temperature by taking first derivative of the magnetization as a function of temperature (dM/dT) and Arrott plots.

Comment 2: *There is non-negligible mismatch between lattice parameters of Fe_3GaTe_2 listed in this work and those from previous report. The lattice parameters from XRD measurement and TEM measurement also don't match to each other. This inconsistency may originate from the thickness of samples, since the thicker sample is less affected by strain. Also, as mentioned by the authors, non-negligible strain effect in those MBE-grown Fe_3GaTe_2 exists through all samples. However, the effect of this strain to the magnetic properties hasn't been mentioned clearly in this manuscript. It is well-known that strain can greatly modulate the magnetic properties of vdW magnets, this strain effect on the magnetic properties of Fe_3GaTe_2 should be clarified via DFT calculation or measuring their magnetic properties in other substrate.*

Reply: Thanks for the comment. The thickness of Fe_3GaTe_2 (FGT) film is ~ 28 nm (18 u.c.) determined by STEM images. The out-of-plane lattice parameter c of such a thick film is almost the same as the bulk value. We note that the discrepancy of the out-of-plane lattice parameter deduced from XRD and TEM is due to the difference on resolution. We measured the in-plane lattice parameter of the 9 u.c. FGT using the in-situ RHEED patterns [Fig. 1b], which demonstrate that the film is under tensile strain from the F-mica substrate.

We fully agree that the strain significantly impacts on the magnetic properties of vdW magnets, especially in the aspect of enhancing Curie temperature in our FGT films in comparison with the mechanically exfoliated samples. To clarify the strain effect, we performed density functional theory (DFT) calculations using Vienna Ab-initio Simulation Package (VASP) software and classical Heisenberg model Monte Carlo (MC) simulations implemented in the VAMPIRE software package (*Physical Review B* **91**, 144425 (2015)). Fig. R1a shows the calculated normalized magnetic moment as a function of temperature under varied strain states. The T_C is determined by fitting the data using the classical spin model $m(T) = (1 - T/T_C)^\gamma$, as plotted in the red solid lines. For the 1 u.c. FGT without strain, the T_C is calculated to be 303 K, which is consistent with

the reported value (approximately 295 K) of exfoliated FGT with same thickness (*npj 2D Materials and Applications*, 8:22, 2024). As shown in **Fig. R1a**, with 0.1% strain, the T_C dramatically increases to 345 K, and with a further increase to 0.4%, the T_C rises to 396 K. The theoretical results reveal that the strain effect would significantly enhance the T_C and ferromagnetism of FGT. We have added the computational details and **Fig. R1** in the revised manuscript (Page. 14 and line 28).

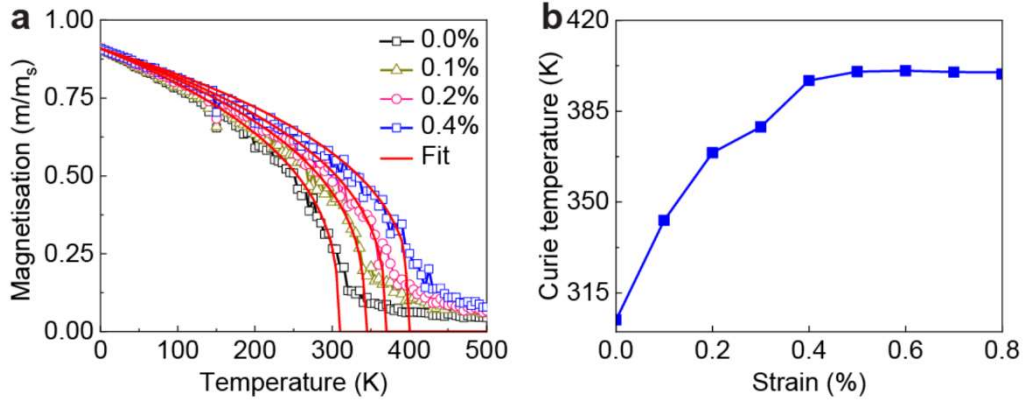


Fig. R1 (a) Simulated temperature-dependent magnetization for the 1 u.c. FGT under different strain states. (b) T_C is dramatically enhanced with increasing strain.

Comment 3: *How the large lattice mismatch is believed to trigger the vdW growth mode of Fe₃GaTe₂?*

Reply: Thanks for pointing out. We have reconsidered the lattice mismatch between the FGT and the F-mica, which have in-plane lattice parameter of approximately 3.986 and 5.304 Å, respectively. As schematically shown in **Fig. R2** and **Supplementary Figure 1**, the in-plane epitaxial relation should be a 4×4 supercell of the FGT on a 3×3 supercell of the F-mica, resulting in an in-plane lattice mismatch is around ~ 0.2 %.

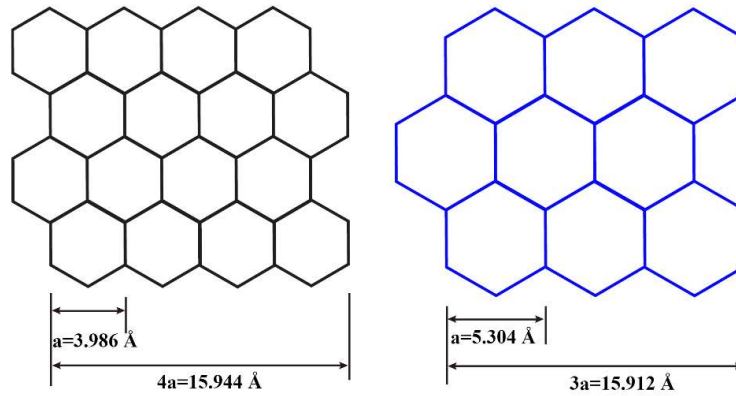


Fig. R2 The in-plane epitaxial relation between a 4×4 supercell FGT and a 3×3 supercell F-mica, results in an in-plane lattice mismatch of approximately 0.2 %.

Therefore, we have deleted the misleading sentence “The large lattice mismatch is believed to trigger the vdW growth mode of FGT” and added the above description in the revised manuscript (Page 5 and Line 4).

Comment 4: *The magnetic moment of magnetic material is proportional to its volume. Therefore, the magnetic moment of 9 ML should be at least 9 times higher than that of 1 ML, or at least there should be deviation between them. However, in fig. 3a and fig. 3d, there is no deviation between them at applied field, which seems to be unrealistic. Also, due to less magnetic volume, the M-T curve of 1 ML Fe₃GaTe₂ is supposed to be noisy compared to 9 ML, like fig. 3c and fig. 3f, which is the opposite situation of fig. 3a and fig. 3d.*

Reply: Thanks for pointing out. Indeed, the magnetic moment of magnetic material should be proportional to its volume. The main reason for the anomaly you pointed out is that the sample size is different. We measured the magnetization (M) above 395 K using the high-temperature (HT) oven component of MPMS-3 (**Fig. 3a**). To prepare the sample, the film should be cut into a piece with the largest size $\sim 2 \times 2 \text{ mm}^2$ and loaded into the brass tube. Then we carried out the M measurements below 395 K on this same piece but need to change the brass tube to quartz rod under the regular S-VSM mode. While the MT data shown in **Fig. 3d** is from the 1 u.c. FGT with the size around $3 \times 5 \text{ mm}^2$. We also need to point out that since the F-mica is soft and fragile, it is hard to cut the FGT/F-mica samples into the same size. To address these discrepancies, we have recalculated the M by normalizing area and changed **Fig.3 a and d** accordingly in the revised manuscript.

Comment 5: *It is very unrealistic that saturation magnetic moment per unit mass of 1 ML is larger than 9 ML. It is widely known that interlayer interaction in vdW materials is considerably weak, still magnetic properties of vdW magnets are greatly affected by the number of layer. This tendency is experimentally shown in several works, the critical temperature of vdW magnets are decreasing with decreasing number of layer, which indicate the weakening of magnetic properties due to thermal fluctuation. The increase of saturation magnetic moment in this manuscript is the opposite situation. Can the authors give any supporting data or simulation result to back up their observance? If saturation magnetic moment is mis-obtained, one of possible reasons is maybe the*

uncertainty of volume or mass obtained during calculating it. Since MBE-grown Fe_3GaTe_2 is directly grown on substrate, it is very challenging to measure the mass or the volume of thin film precisely. This uncertainty may result to inaccurate value of saturation magnetization. Anyhow, the authors should clarify this issue, 1) Is the increase of saturation magnetization not mis-obtained result? 2) If so, then what is origin of increase of saturation magnetization at 1 ML.

Reply: As briefed in the *reply-to-Comment 4*, we have recalculated the magnetic data by normalizing the volume. The corresponding MH curves for FGT with different thicknesses are shown in **Fig. R3**. For the 1 u.c.-thick sample, to guarantee the electrical transport results the first unit cell FGT should be fully covered the substrate. However, it is noted that achieving the precise coverage of the first unit cell is challenging, often resulting in partial coverage of the second unit cell (e.g. the actual thickness exceeding 1 u.c.). The uncertainty in the coverage of the second unit cell of FGT could lead to deviation of magnetic moment ratio between the 9 u.c. FGT and 1 u.c. sample. Moreover, as discussed in *Reply-to-comment-2* and **Fig. R1**, the strain effect would significantly enhance the ferromagnetism in FGT, which is more pronounced in 1 u.c.-thick sample. This could also contribute to the deviation of magnetic moment. We have added the above discussion in the revised manuscript (Page 9, line 25).

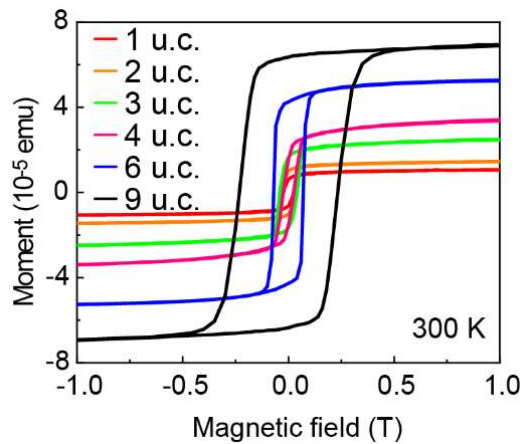


Fig. R3 M - H curves for FGT with different thicknesses at 300 K.

Comment 6: *How can the decrease of coercivity from 9 ML to 1 ML be interpreted to the existence of weak interlayer interaction? For me, the weakening of coercivity seems to reveal the non-negligible interlayer interaction between layers. This sentence doesn't harmonize not only with common sense, but also with the flow of the manuscript.*

Therefore, this sentence must be deleted, in my opinion.

Reply: Thanks for the suggestion. Based on our DFT calculations and MC simulations, we reveal that the strain would significantly affect the magnetic properties of FGT. We have deleted this description related to “weak interlayer interaction” in the revised manuscript.

Comment 7: *The magnetic properties of Fe₃GeTe₂ and Fe₅GeTe₂ are shown to be greatly affected by proximity effect. Therefore, the argument what authors have written related to substrate effect is almost not persuasive to me. To rule out the concern related to substrate effect, the authors is recommended to grow Fe₃GaTe₂ on other substrate, which will rule out substrate and strain effects.*

Reply: Thanks for this constructive suggestion. To elucidate the proximity effect and strain effect, we have grown a 1 u.c. FGT film on F-mica with a 1 u.c. PtTe₂ buffer layer. The PtTe₂ has a hexagonal lattice symmetry with lattice constants $a = b = 4.026 \text{ \AA}$ and $c = 5.221 \text{ \AA}$ [Nature Communications 15, 2605 (2024)]. The lattice mismatch between FGT and PtTe₂ is $\sim 0.1 \%$, half smaller than that ($\sim 0.2 \%$) between 4×4 supercell FGT and 3×3 supercell F-mica. **Figure R4** shows the M - H loops at various temperatures and dM/dT for both FGT/PtTe₂/F-mica and FGT/F-mica. As compared in **Fig.R4(d)** and **R4 (b)**, the T_C of the FGT/PtTe₂/F-mica is approximately 330K, lower than that ($T_C = 345 \text{ K}$) of FGT on F-mica. This, in conjunction with our theoretical calculations [Reply-to-comment-2 and **Fig. R1**], reveals that the strain effect would significantly enhance the ferromagnetism of Fe₃GaTe₂. We have added the above results and descriptions into the revised supporting information (**Supplementary Figure 14**), and have changed the corresponding description in the revised manuscript (Page 12 Line 22).

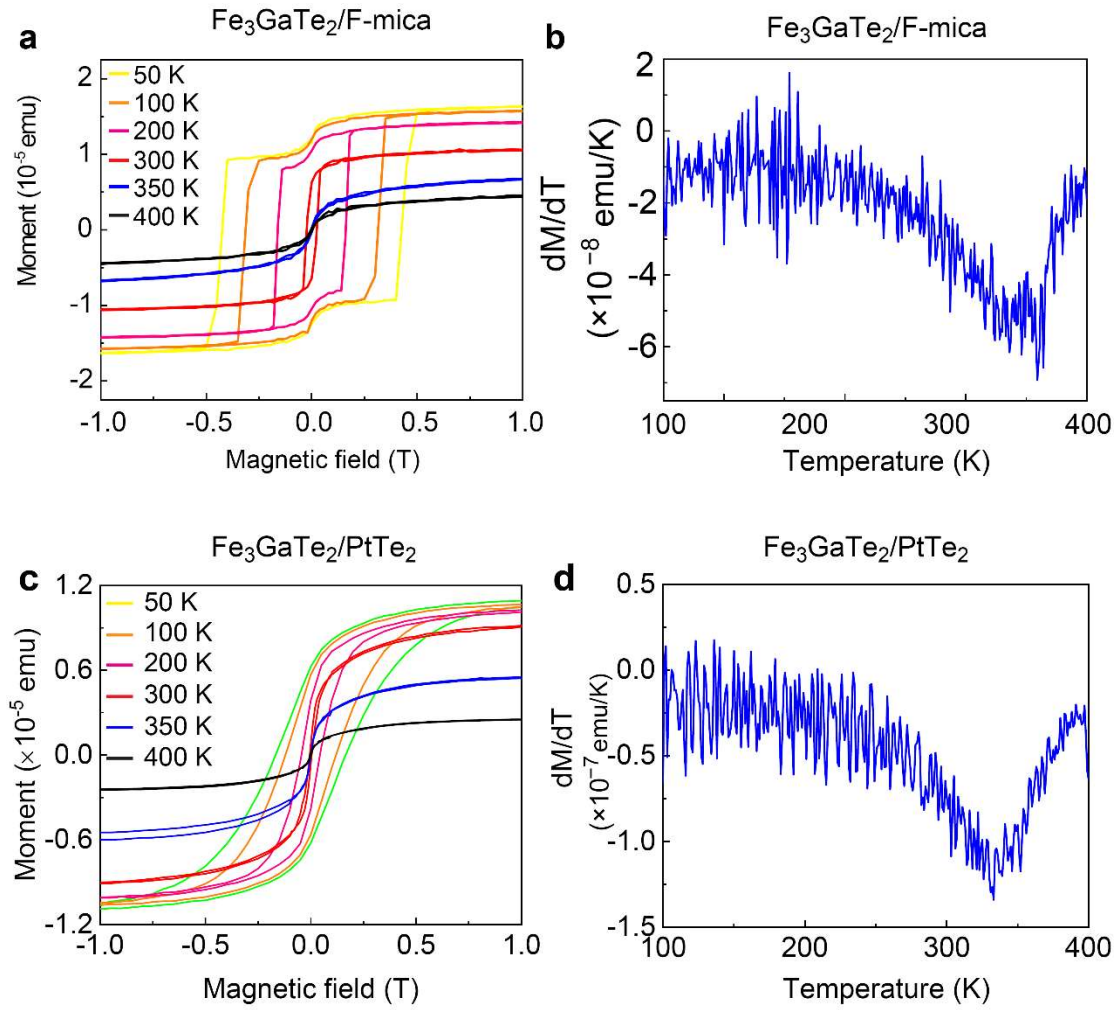


Fig. R4. M - H loops at various temperatures and dM/dT for FGT/F-mica and FGT/PtTe₂/F-mica.

Comment 8: *There are multiple plateaus in fig. 3e. It is presumably due to the formation of multi-domain, but the authors didn't mention about it in the manuscript. This step-like hysteresis loop is hardly observed in typical FM hysteresis loop, so the authors should clarify the origin of this behavior.*

Reply: We agree that the multiple plateaus is presumably from the formation of multi-domain. We note that, compared to 9 u.c.-thick sample, such multiple plateaus are more pronounced in the 1 u.c.-thick sample. As briefed in the Reply-to-Comment 5, this observation could be due to the thickness inhomogeneity, which is more significant in the thinner samples. We have clarified the possible origin in the revised manuscript (page 9, line 3).

Comment 9: *In fig. 3i, negative stray field is dominant in upper region and positive stray field is comparably dominant in lower region. This result can be misinterpreted*

as that Fe₃GaTe film possesses non-uniform magnetic domain structure. The authors explained that stray field mapping is shown to prove robust FM characteristic even in room-temperature, but I think presented magnetization measurement data and transport data is enough. Thus, I recommend to remove figs. 3h and i or present mapping data which is measured at the magnetic field strong enough to saturate Fe₃GaTe₂ film.

Reply: Thanks for the suggestion. Currently, the maximum external magnetic field of the scanning NV spin magnetometry is limited to be 500 Oe, which is not strong enough to completely magnetize the FGT samples. In the revised manuscript, we have removed the NV center results and corresponding descriptions.

Comment 10: The authors mentioned that T_c of 1 ML is higher than 400 K, based on anomalous Hall effect (AHE) measurement. However, in fig. 3c, R_H - H of 1 ML at 400 K is almost linear and AHE is hardly observed. Therefore, the T_c of 1 ML is lower than 400 K, which can also be proved by fig. 3d. Based on AHE measurement, the T_c of 1 ML is in the range of 300~350 K. Therefore, the argument related to T_c of 1 ML Fe₃GaTe₂ based on AHE measurement should be revised.

Reply: Thanks for the insightful suggestions. To accurately determine the Curie temperature of 1 u.c. FGT, we did the Arrott plots (**Fig. R5**). We replot the Hall data in plots of R_H^2 as a function of $\mu_0 H/R_H$, where $\mu_0 H$ is magnetic field, and R_H is Hall resistance. The Curie temperature can be extrapolated by the Arrott plots intercepts through fitting high-field linear part of the R_H^2 axis [*Nature* **563**, 94–99 (2018)]. Therefore, the T_C of 1 u.c. FGT is deduced to be ~ 345 K. We have corrected the result in the revised manuscript.

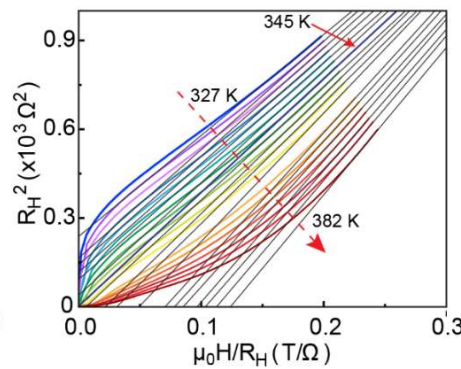


Fig. R5. The Curie temperature of the 1 u.c. FGT can be extrapolated by the Arrott plots intercepts through fitting high-field linear part of the fitting high-field linear part.

Comment 11: *The R_H values of 9 ML Fe_3GaTe_2 is much smaller than that of 1 ML Fe_3GaTe_2 in fig. 4d. There is no direct correlation between anomalous Hall resistance and magnetization, still one can misinterpret that the magnetization of 1 ML Fe_3GaTe_2 is larger than that of 9 ML Fe_3GaTe_2 . Therefore, I strongly recommend to convert R_H to anomalous Hall conductivity, which is directly proportional to magnetization of FM material.*

Reply: Thanks for the constructive suggestion. We have converted R_H to anomalous Hall conductivity in the revised manuscript.

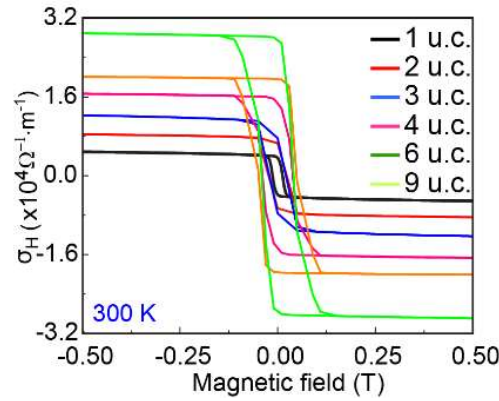


Fig. R6. Anomalous Hall conductivity as function of magnetic field in the FGT film with different thickness.

Comment 12: *The authors mention the bulk lattice parameters from [Nat. Commun. 13, 5067 (2022)] in manuscript, but didn't refer this previous work in first sentence of second paragraph in pg. 2. The authors should refer this paper in this specific sentence.*

Reply: Thanks for pointing out. We have made the correction in the revised manuscript (page 3, line 25).

Comment 13: *How did the authors define the T_C ? It is pointed in fig. 3a for the 9ML Fe_3GaTe_2 , it is too roughly defined. There are many ways to define T_C , none of them are explained in this paper. The authors should decide at least one criterion to define the T_C of Fe_3GaTe_2 films.*

Reply: Thanks for pointing out the important issue. We use the dM/dT to determine the T_C of 9 u.c. FGT. As shown in **Fig. R7 a and b**, the T_C of 9 u.c. and 1 u.c.) FGT is extracted to be ~ 420 K and 350 K, respectively. Moreover, Arrott plots of R_H^2 as a function of $\mu_0 H/R_H$ further precisely determines that T_C of 1 u.c. FGT is ~ 345 K, which

has been briefed in *Reply-to-Comment 10*.

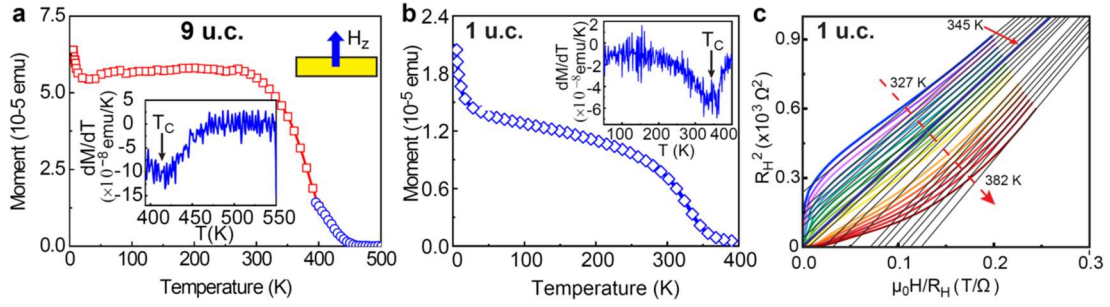


Fig. R7 The T_C of the epitaxial FGT with (a) 9 u.c. and (b) 1 u.c. can be determined by dM/dT and (c) Arrott plots.

Comment 14: Based on [Nanoscale, 15, 11290 (2023)], one can realize that c lattice parameter is critical to magnetic property of Fe_3GaTe_2 . The mismatch between c lattice parameters from different measurement method is acceptable, still it seems better for the authors to mention that c lattice parameter may have some error range due to different measurement is acceptable with referring it for excuse.

Reply: Thanks for the kind suggestion. We agree that the mismatch between c lattice parameters from different measurement methods should be due to the error range of these measurements. We have referred this paper in our revised manuscript to explain the discrepancy (Page 6, line 13).

Comment 15: How did the authors calculate the lattice parameter of 9 ML Fe_3GaTe_2 film?

Reply: In-plane lattice constants of 9 u.c. FGT films were calculated by measuring the streak spacing of RHEED patterns via taking streak spacing and in-plane lattice constant of F-mica for reference.

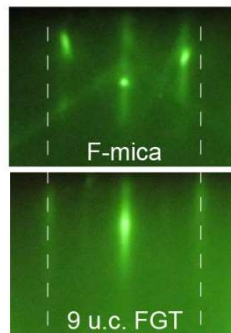


Fig. R8. RHEED patterns of F-mica and 9 u.c. FGT films

Comment 16: What is the origin of up-turn of magnetization in M - T curve at low

temperature region in figs. 3a and d? It may originate from substrate, but it wasn't mentioned in the manuscript. It's better for the authors to mention the origin of it.

Reply: The up-turn of magnetization in M - T curve at low temperature region could be originated from the impurities of the substrate.

Comment 17: *Refs. 7 and 8 are duplicated at refs. 25 and 34, respectively.*

Reply: Thanks for pointing out the mistakes. We have done the corresponding correction in the revised manuscript.

Comment 18: *The authors mentioned that SQUID is indirect experimental method. However, SQUID measurement is one of the experimental methods which can directly measure magnetic moment of the sample. I cannot agree with this sentence, except it is reflecting macroscopic magnetic properties. The authors are recommended to re-write this sentence, removing indirect experimental method and only mentioning about the limit of the macroscopic measurement.*

Reply: Thanks for the suggestion. We have deleted the sentence in the revised manuscript.

Reviewer #2

Comment 1: *In this manuscript, the authors grow Fe₃GaTe₂ (FGT) thin films by using MBE for the first time. The as-grown FGT thin films show very high T_c and large PMA, which is very crucial for 2D spintronics. They try to characterize and discuss the structure and magnetism of FGT films and some results are presented. However, I have important concerns and doubts regarding the magnetism and crystal characterizations, and some statements are lack evidence or references. Meanwhile, the underlying mechanisms about high T_c and variation trend with thickness are not clearly demonstrated. Therefore, I cannot recommend this manuscript for publication before the authors reply satisfactorily to the comments below.*

Reply: We thank the reviewer for the thoughtful review of our manuscript. To elucidate the origin of the high- T_c observed in the MBE-grown FGT films, we have performed theoretical calculations and grown the FGT with a PtTe₂ buffer layer to reduce the lattice mismatch, indicating that the high- T_c of the epitaxial FGT could be originated from the strain effect.

Comment 2: *In Fig 2a and 2c, cross-sectional TEM images are presented. Are they HRTEM images or HAADF images? In the caption, the authors claim “HRTEM” image, but in the manuscript, the authors claim “HAADF” image, and I am confused about it. To my knowledge, they should look like HAADF images, as the authors clearly indicate the atoms in Fig 2c, and Te atoms have the brightest contrast due to the largest atomic number. However, in Fig 2a, the Te capping layer and F-mica substrate (with relatively small atomic numbers) are clearly observed and bright, but all FGT region is very dark. I am very confused about it because the atoms in FGT are heavier than that in F-mica, and there are many Te layers contained in FGT. Moreover, the Fe and Ga atoms in Fig 2c are not very clear. I think they should provide a clearer cross-sectional TEM image to replace current Fig 2c. What is the atomic ratio of the as-grown FGT film? EDX test can provide a specific ratio of Fe, Ga, and Te.*

Reply: Thanks for pointing out the problems. We have revised the manuscript to address your concerns. Figure 2a depicts a TEM image acquired in TEM mode, where an aperture was utilized for enhancing the contrast and was set to auto-contrast mode. In this TEM mode, image contrast can be affected by many factors, including the focal

plane, the thickness of the sample piece (the piece after cutting via FIB), and the positioning of the aperture's center. This makes it impractical to conclusively determine the elemental composition solely from TEM images. Figure 2b presents a HAADF-STEM image obtained in STEM mode. The intensity of the signal in HAADF images is approximately proportional to the square of the atomic number. Therefore, elements with higher atomic numbers scatter electrons more strongly and appear brighter in the image. Consequently, Te atoms, having a higher atomic number, appear largest and brightest in the image.

For clearer clarity, we have modified the caption of Figure as follows: **STEM images and EDS mapping. a.** Cross-section STEM images taken from the interface area of the FGT thin film grown on F-mica with the scale bar of 20 nm. **b.** EDS mapping display uniform distribution of Fe, Ga, and Te atoms in the film, marked as the region between the white dash lines. **c.** HAADF-STEM image of the FGT confirms the P63/mmc space group of the epitaxial FGT.”

Obtaining higher-resolution cross-sectional STEM images poses significant challenges in our study. Since the substrate is not stable under the electron beam due to its layered nature. The continuous exposure to the electron beam during image acquisition can result in significant damage to both the substrate and the sample, even during the image snapshot process. Lowering the operating voltage of the STEM could lessen the damage to the sample but lowers the resolution of the imaging. In response

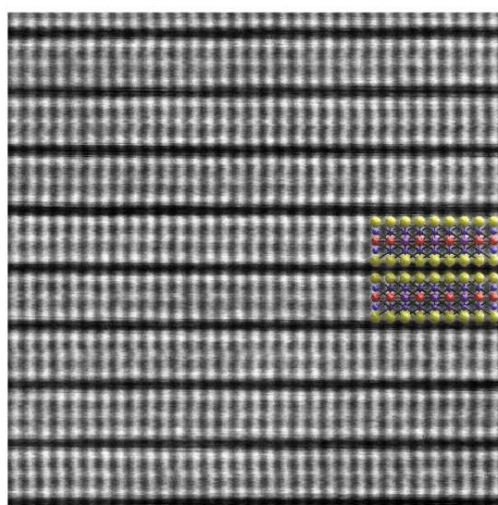


Fig. R9. HAADF-STEM image of the FGT confirms the P63/mmc space group of the epitaxial FGT.

to these challenges and in line with your suggestions, we have made concerted efforts to enhance the resolution of our STEM images. We have managed to obtain a new STEM image that offers improved resolution and clarity. Fig.R9 shows the HAADF-STEM image of FGT (~ 28 nm) on F-mica. This high-resolution image has been added into the revised manuscript (Fig. 2b).

Regarding elemental analysis, while Energy-Dispersive X-ray Spectroscopy (EDS) can indeed provide an atomic ratio of elements. Our measurements present certain challenges that affect the accuracy. We have performed the EDS measurements many times, the atomic ratio of Fe, Ga, and Te show considerable variability. This region as shown in Figure 2(a) of the manuscript includes a Te capping layer, which artificially elevates the apparent concentration of Te. Additionally, Ga, being an element with a low melting point, is particularly susceptible to beam damage during EDS mapping under high-energy electron beams (200 kV). This might result in a reduced apparent concentration of Ga in the analyses spectrum. These factors demonstrates that EDS may not be the most reliable method for determining specific atomic ratios in our FeGaTe (FGT) films. To overcome these limitations, we carried out element concentration analysis by photoelectron spectroscopy (XPS), as shown in **Supplementary Figure 3**. The element analysis gives the Fe: Ga: Te concentration ration $\sim 3:1:2$.

Comment 3: *In Fig 3a, the authors identify the temperature at which the magnetization decreased to zero as T_c . Based on this, the T_c of the 1 ML and 3 ML samples seems to be ~ 370 K (Fig 3d and Fig S1), and thus the AHE in the 1-4 ML FGT films at 400 K is negligible (Fig 4c and Fig S6a-c). But the hysteresis loops still exists at 400 K (Fig 3e and Fig S3a). Can the authors give a reasonably explanation? I think the current identification method for T_c is not ideal. Can the author use a unified standard method to determine T_c from M - T or AHE curves in each samples (e.g. dM/dT - T plots or Arrott plots)?*

Reply: Thanks for pointing out the important issue. We use the unified method to determine T_c (dM/dT - T plots). As shown in **Fig. R10 a and b**, the determined T_c from the dM/dT curves is ~ 420 K and ~ 350 K for 9 u.c. and 1 u.c. FGT, respectively. Following the suggestions from Reviewer #1 (see *reply-to-comment 10&13*), we also

did the Arrott plots to accurately determine the T_C of 1 u.c. FGT, as shown in **Fig. R10 c**, which gives T_C of ~ 345 K, consistent with the value determined by dM/dT - T plots. We have added the following description and **Fig. R10** into the revised manuscript (Page 6, line 20 and Page 9, line 20).

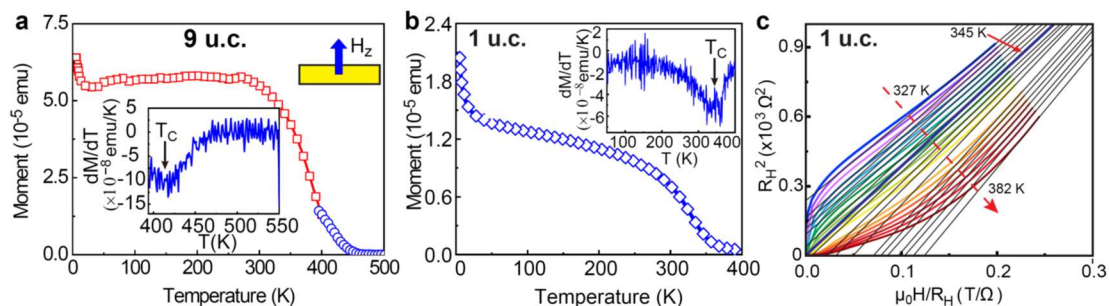


Fig. R10 The T_C of the epitaxial FGT with (a) 9 u.c. and (b) 1 u.c. can be determined by dM/dT and (c) Arrott plots.

Comment 4: *The T_c of as-grown 9 ML and 1 ML FGT films are respectively much higher than the T_c of exfoliated ~ 9.5 nm FGT flake ($T_c \sim 350$ K, Nat. Commun. 13, 5067, 2022) and monolayer one ($T_c \sim 200$ K, Nano Lett. 2024, 24, 993) in previous works. What causes such higher T_c in this work?*

----Initially, I think it is attributed to the tensile strain due to the lattice mismatch between few-layer FGT films and substrate. Because the larger in-plane lattice constant of as-grown FGT films compared with bulk value demonstrates the existence of tensile strains. However, the authors think the substrate effect on the ferromagnetic phenomena is negligible because of the weak layer-dependent T_c , and thus the FGT films they grown can be regarded as an isolated magnet. I am very curious about this. If they are “isolated magnets”, they should have ferromagnetic properties similar to exfoliated FGT flakes (e.g. T_c). Thus, I think the underlying mechanisms of such high T_c should be fully addressed, and additional results for supporting their mechanisms are needed.

Reply: We totally agree this opinion. The significantly higher Curie temperature of the epitaxial FGT on F-mica in comparison with bulk or exfoliated FGT should be due to the strain effect. To understand the large enhancement of Curie temperature of the epitaxial FGT on F-mica substrate, we performed density functional theory (DFT) calculations with Vienna Ab-initio Simulation Package (VASP) software and the Monte Carlo (MC) simulation implemented in the VAMPIRE software package. The computational details have added in the revised manuscript. As shown in **Fig. R11** For the 1 u.c. FGT without strain, the T_C is calculated to be 303 K, which is consistent with

reported value (approximately 295 K) of exfoliated FGT with same thickness (*npj 2D Materials and Applications*, 8:22, 2024). However, with 0.1% strain, the T_C is dramatically enhanced to 345 K. With the strain increasing to 0.4%, the T_C is up to 396 K. The theoretical results reveal that the strain effect would significantly enhance the T_C and ferromagnetism of FGT.

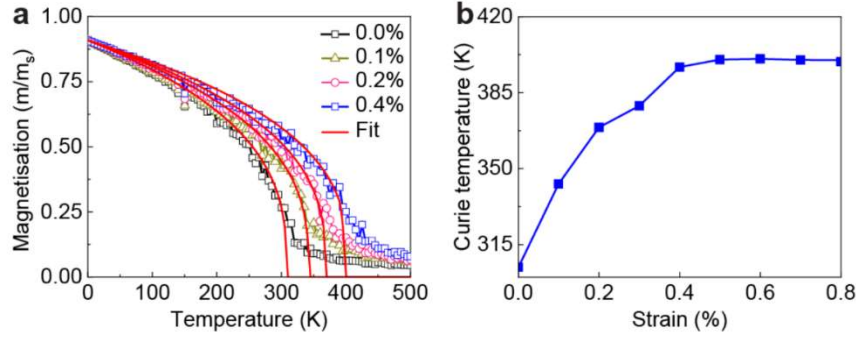


Fig. R11 Classical Heisenberg model Monte Carlo simulations. With the strain increasing, the T_C is dramatically enhanced.

Comment 5: *The weak layer-dependent T_C is an unusual phenomenon in vdW magnets especially in 2D limit. The authors use weak interlayer exchange coupling to explain the small variation of T_C with thickness in FGT, and they use isostructural Fe_3GeTe_2 as an example to support their opinion. However, for Fe_3GeTe_2 , the variation of T_C with thickness is significant, such as from 175 K in 10 ML to 75 K in 1 ML (a decrease of 57%, *npj 2D Mater Appl* 4, 33, 2020). Even for FGT flake, the T_C is from ~300 K in 10 nm to ~200 K in 1 nm (a decrease of 33%, *Nano Lett.* 2024, 24, 993). In contrast, the larger thermal fluctuations at higher temperature should have a larger impact on T_C , but the T_C of FGT films persist over 400 K and only decreased by ~12% from 9 ML to 1 ML. Thus, I am not convinced this explanation for weak layer-dependent T_C , or the authors should provide more solid evidences to explain this unusual phenomenon.*

Reply: We indeed observe an unusual weak layer-dependent T_C in the FGT films. Combined the DFT calculations and controlled samples, the former assumption, that small variation of T_C with thickness in FGT is due to weak interlayer exchange coupling, is incorrect. Since the strain effect is more pronounced with the thinner films, the small variation of T_C with thickness is another evidence to support the high T_C and enhanced ferromagnetism is originated from the strain effect.

Comment 6: *The authors use the saturation magnetic moment per unit mass (emu/g) for comparison in the manuscript. How is the mass of ultrathin FGT films obtained? Moreover, why does the M_s increases more than twice at 300 K as the thickness decreases from 9 ML to 1 ML? Does this phenomenon indicates the influence of substrate effect on ferromagnetism?*

Reply: From the evolution of RHEED patterns of FGT during the whole growth, we can observe that the streak spacing remain unchanged with thickness increasing. The result revealed that the in-plane lattice constant of FGT with different thickness would keep invariable, which should be ascribed to the small lattice mismatch and weak Van der Waals force between the epitaxial FGT and the F-mica substrate. Therefore, we could obtain the volume and corresponding mass of the ultrathin FGT films.

Indeed, the magnetic moment of magnetic material should be proportional to its volume. The main reason for the anomaly you pointed out is that the sample size is different. We measured the magnetization (M) above 395 K using the HT oven component of MPMS-3 (**Fig. 3a**). To prepare the sample, the film should be cut into a piece with the largest size $\sim 2 \times 2 \text{ mm}^2$ and loaded into the brass tube. Then we carried out the M measurements below 395 K on this same piece but need to change the brass tube to quartz rod under the regular S-VSM mode. While the MT data shown in **Fig. 3d** is from the 1 u.c. FGT with the size around $3 \times 5 \text{ mm}^2$. In addition, since the F-mica is soft and fragile, it is hard to cut the FGT/F-mica samples into the same size. We have recalculated the M by normalizing area and changed **Fig.3 a and d** in the revised manuscript. The corresponding MH curves for FGT with different thicknesses is shown in **Fig. R12**. For the 1 u.c. sample, according to the *in-situ* RHEED patterns, it difficult to precisely control the thickness, and therefore the actual thickness is larger than 1 u.c.). The uncertain coverage of the second unit cell of FGT could lead to deviation of magnetic moment ratio between 9 u.c. and 1 u.c. FGT. Moreover, magnetic moment can be largely enhanced by strain-induced lattice deformation in the vdW ferromagnet due to the competitive effects of direct exchange and super-exchange interaction. The enhancement could be more pronounced in 1 u.c. FGT. These factors could also make the deviation of magnetic moment. We have added the above discussion in the revised

manuscript (Page 8, line 18).

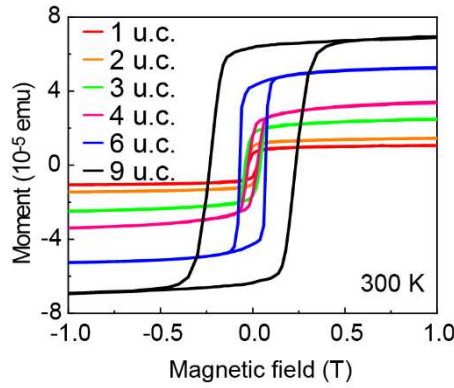


Fig. R12 *M-H* curves measured at 300 K for FGT films with different thicknesses.

Comment 7: *The author believes that the decrease of H_c with decreasing thickness indicates weak interlayer coupling. Can the author provide a more detailed explanation to reveal the relationship between thickness-dependent H_c and interlayer coupling strength?*

Reply: Thanks for pointing out this issue. We have recalculated the Curie temperature by first derivative of the magnetization as a function of temperature (dM/dT), the Curie temperature of 9 u.c. and 1 u.c. FGT was extracted at around 420K and 350 K, respectively. Moreover, as the thickness being reduced from 9 u.c. to 1 u.c., the corresponding H_C dramatically decreases from 2360 Oe to 230 Oe at 300 K, revealing large interlayer interaction. The significant dependence of Currie temperature and coercive field on thickness reveal that the interlayer coupling should be strong. We have corrected the explanation of weak interlayer coupling in the revised manuscript.

Comment 8: *For the NV spin magnetometry tests, the magnetic domain structures of 1 ML FGT seems to be different from that of 9 ML FGT. Can the authors comment on it? Moreover, under zero bias magnetic field, the stray magnetic field intensity is almost randomly and uniformly distributed, but when a 500 Oe magnetic field is applied, their distribution becomes very uneven (in the Fig 3i, the upper half is blue, and the lower half is red) , which is very curious. How do the authors explain this? Also, I suggest the authors should continue to increase the bias magnetic field to saturate the 1 ML FGT and demonstrate a complete field-driven domain evolution.*

Reply: Thanks for the suggestion. Currently, the maximum external magnetic field of the scanning NV spin magnetometry is limited to 500 Oe, which is not strong enough

to completely magnetize the monolayer FGT. Following the suggestion from Reviewer #1 (*Comment 9*), the present magnetization measurement and transport data are enough to draw the conclusion that the enhanced ferromagnetism with high- T_C (above RT) in epitaxial growth FGT is driven by the tensile strain effect. Consideration that other reviewers' suggestion and limitation of the scanning NV spin magnetometry, we prefer to remove this part in the revised manuscript.

Comment 9: *The authors claim “The anomalous temperature dependence of carrier density and mobility could be related to the increase in density of state at Fermi level and effective mass of electron due to the variation of band structure with temperature.” Can the authors provide some evidences to support this statement?*

Reply: Thanks for pointing out. Currently, we don't have evidence to support this assumption. To avoid misunderstanding, we have deleted the statement in the revised manuscript.

Minor issues:

Comment 10: *I suggest they provide a photograph of as-grown wafer-scale FGT thin film (at least 1 inch) to support the so called “wafer-scale” in the title.*

Reply: Thanks for the suggestion. We have provided a photograph (**Fig. R13**) of as-grown wafer-scale 1 u.c. FGT with scale of 3 cm×3 cm in the revised supporting information (**Supplementary Figure 2**).

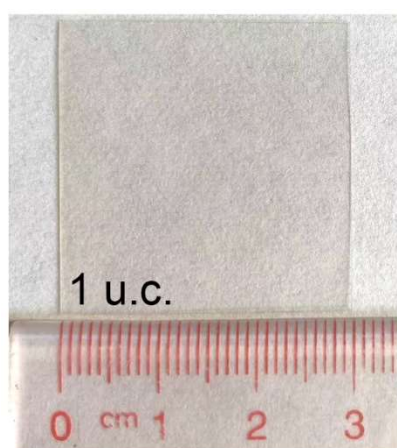


Fig. R13. Photograph of as-grown wafer-scale 1 u.c. FGT with scale of 3 cm×3 cm

Comment 11: *Why does the ZFC moments in Fig S1 become negative within a certain temperature range?*

Reply: The occurrence of negative ZFC moments in [Supplementary Figure 4](#) within a certain temperature range could be related to the large diamagnetism of the F-Mica substrate or demagnetization for the SQUID. To avoid misleading, we have remeasured ZFC-FC curves for the 1 u.c. FGT under 0.2 T, as shown in [Fig. R14](#). It can find that no negative moment within the certain temperature range can be observed.

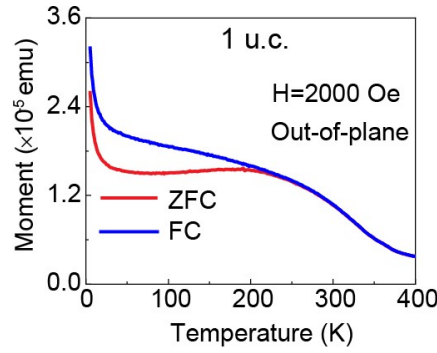


Fig. R14. ZFC-FC curve for 1 u.c. FGT under 0.2 T.

Comment 12: *The calculation method for PMA constants should be disclosed.*

Reply: Thanks for the suggestion. We calculated the PMA constants K_U by $\mu_0 \int_0^{M_s} H(M) dM$ (In plane) $- \mu_0 \int_0^{M_s} H(M) dM$ (out of plane) , where M_s is the saturation magnetic moment. We have showed the calculation detail in Methods of the revised manuscript.

Comment 13: *Is there any literature indicating that FGT is an itinerant ferromagnet? Please cite it.*

Reply: Recent research found that the exfoliated FGT flake is a weak itinerant ferromagnetism [Nano Letters, 24, 993 (2024)]. To avoid misunderstanding the description of itinerant ferromagnet in this work, we have deleted the related sentences in the revised manuscript.

Reviewer #3

Comment 1: *In this study led by Shuxiang Wu et al., the investigation of MBE-grown Fe₃GaTe₂ monolayer and multilayer ferromagnetic thin films is presented. However, the claim of a significantly higher Curie temperature (T_c) above 400 K for Fe₃GaTe₂ compared to bulk samples and other 2D ferromagnets requires further clarification. This is especially notable given that bulk Fe₃GaTe₂ has a T_c of 380 K, raising concerns about the assumption that thin films inherently possess smaller T_c . Moreover, the 9ML sample (457 K) in this manuscript exhibits a considerably higher T_c than the 1ML sample (400 K), adding complexity to the understanding of T_c evolution with thickness. The authors delve into the lower T_c in 2D ferromagnets on the theoretical side as well.*

Reply: To elucidate the origin of the high- T_c observed in the MBE-grown FGT films, we have performed theoretical calculations (See **Reply-to-Comment-2** from Reviewer #1) and grown the controlled sample FGT/PtTe₂/F-mica (See **Reply-to-Comment-7** from Reviewer #1), indicating that the high- T_c is originated from the strain effect. We demonstrate that the magnetism of epitaxial FGT is enhanced by the tensile strain, which is totally different from the magnetic property of the bulk FGT. We have done extensive revisions in the revised manuscript.

Comment 2: *The investigation of potential excess Fe in Fe₃GaTe₂ thin films as a contributing factor to the observed higher T_c warrants exploration. Detecting an (e.g up to 20%) excess of Fe using the provided characterization methods, like STEM, might pose challenges due to potential limitations in resolving excess Fe. The exceedingly high carrier densities, akin to copper metal, as measured in this manuscript, could also be attributed to an excess of Fe. If the elevated T_c is indeed linked to excess Fe, it questions the validity of the paper's claim of "intrinsic" ferromagnetism and high Curie temperatures.*

Reply: Thanks for pointing out the important issue. During the film growth, we have carefully adjusted the flux ratio of Fe and Ga to 3:1. As a result the stoichiometric FGT exhibits highest Currie temperature compared to other nonstoichiometric FGT films, such as Fe-poor FGT and Fe-excess FGT samples. The stoichiometric 2 u.c. FGT shows highest Curie temperature, while for the Fe-poor and Fe-excess FGT samples the Curie temperature is degenerated, respectively, as shown in **Fig. R15**. Therefore, this series

of samples give strong evidence that the high- T_C is not originated from excess Fe (or Fe impurities) in the FGT films. We have added the above description [Fig. R15](#) and [Supplementary Figure 11](#) in the revised Supporting Information. We have done the corresponding revision in the revised manuscript (Page 11, Line 25).

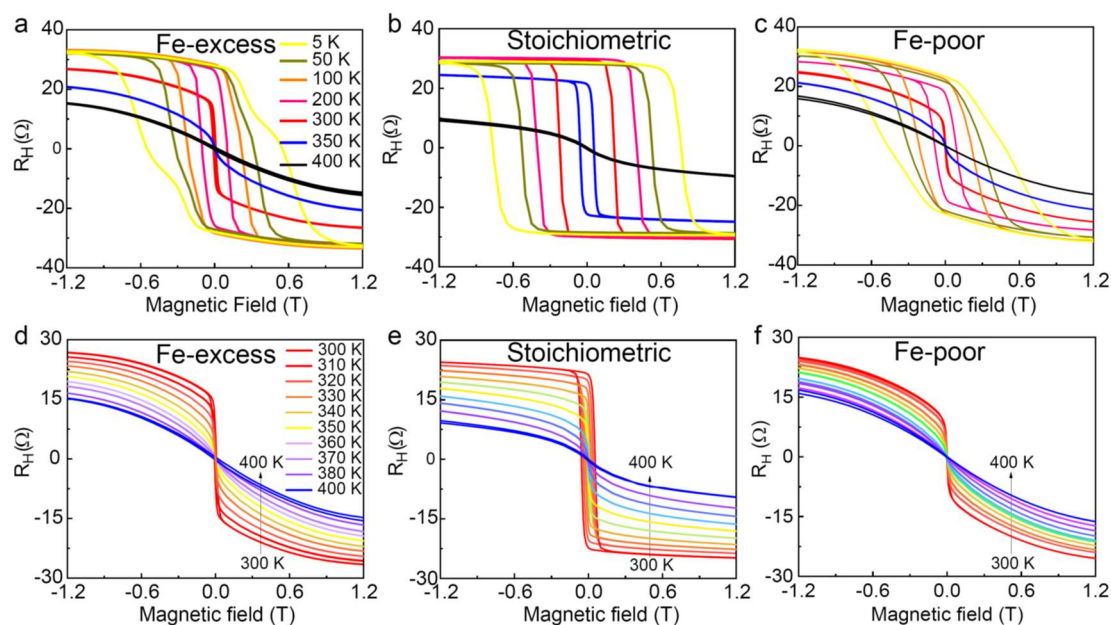


Fig. R15. R_H - H loops of stoichiometric, Fe-poor FGT, and Fe-excess FGT films.

Comment 3: *To enhance the manuscript, the inclusion of RHEED oscillations during film growth would be valuable. Additionally, comparing the period of these oscillations with the calibration could provide further insights. Given the potential instability in the supply of Fe from the e beam source, this aspect should be considered and discussed, acknowledging its impact on the results.*

Reply: That's a good point. It was found that the FGT can be destroyed under illumination of high-energy electron beam of RHEED for long time. So, it's very difficult to acquire the RHEED oscillations during the film growth. In our experiments, we estimated the growth speed by observation of the RHEED pattern every 20 seconds. During growth, the RHEED streaks of F-mica would gradually become a blur and then disappear, while that of the epitaxial film would get clearer. As shown in [Fig. R16](#), the disappearance of the F-mica patterns at approximately 4 min 30s revealed that the surface of F-mica was covered by a monolayer of epitaxial film, corresponding to the growth rate of 1 u.c./4 min 30s. The growth speed was further confirmed by TEM.

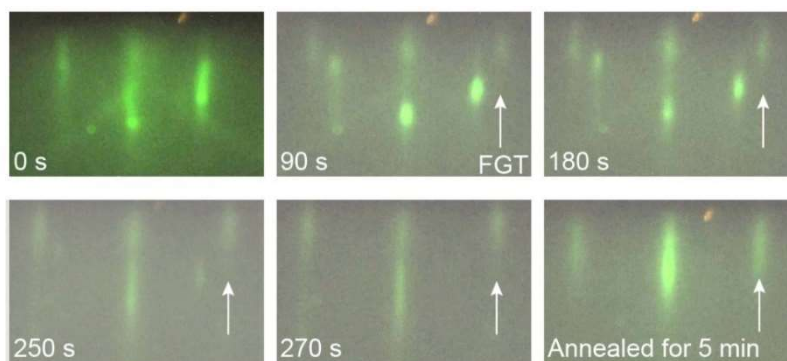


Fig. R16. Estimation of the growth speed by observation of the RHEED patterns.

Comment 4: *Moreover, for a more comprehensive understanding, control samples subjected to different Fe supplies during the growth processes should be examined. This could provide valuable data on how variations in Fe supply influence the properties of the thin films.*

Reply: We agree that the different Fe supplies should influence the magnetic properties of the FGT films. As briefed in *reply-to-comment-2*, we have grown a series of control samples with different Fe: Ga ratio. As shown in **Fig. R15**, the stoichiometric 2 u.c. FGT shows the more squared RH loops and higher T_C than those of Fe-poor or Fe-excess FGT films, indicating the nonstoichiometry would degenerate the magnetism of FGT. Besides, we observed the plateaus in the R_H - H curve at 5 K for the Fe-excess FGT, which could be attributed to the formation of other Fe-Ga-Te phases. This also indicates excess Fe supply would not lead to Fe impurities, behaving like Fe free ions, which dramatically influence the magnetic behaviors. Excess Fe atoms in Fe-Ga-Te structure frame would be bonded to Ga or Te atom, forming $\text{Fe}_{3+x}\text{GaTe}_2$ phases, as reported in Fe_nGaTe_2 ($n = 3, 4, 5$) systems [*Nature Communications*, 14, 2483 (2023)].

Comment 4: *Furthermore, the authors are advised to conduct X-ray Absorption Spectroscopy (XAS) on Fe edges of the thin films. Utilizing Fe valences of 2+ and 3+ in Fe_3GaTe_2 , XAS has the potential to identify excess Fe, which may be closer to Fe 0+, offering valuable insights into its quantity relative to Fe in the films.*

Reply: We thank the constructive suggestion. Soft X-ray absorption spectroscopy (XAS) was conducted at the Hefei Light Source. Fe $L_{2,3}$ -edge was probed at beamlines MCD-A and MCD-B using the Total Electron Yield (TEY) mode. We note that, because of the ultrathin nature of our samples, the obtained XAS signals are noisy. **Fig. R15**

shows the Fe $L_{2,3}$ -edge XAS of the 2 u.c. FGT with different Fe/Ga ratio. The peak positions of Fe L_3 -edge are unchanged with the varied Fe/Ga ratio, indicating the Fe valence state in these samples is the same. Moreover, the line shape of the L_3 -edge is similar to that of the Fe_3GeTe_2 . There are two peaks at 707.6 eV and 709.1 eV, originated from Fe^{2+} and Fe^{3+} states, respectively. [*Phys. Rev. B* **108**, 094408 (2023)]. Moreover, we did not observe excess Fe metal (Fe^0) peaks at 706.7 eV from the XAS curves. Therefore, we could exclude the existence of metal Fe impurities in our FGT films, which would dramatically affect the magnetic properties. We have added **Fig. R17** into the revised Supporting Information (as **Supplementary Figure 12**) and did the corresponding revision in the revised manuscript (Page 12, Line 3).

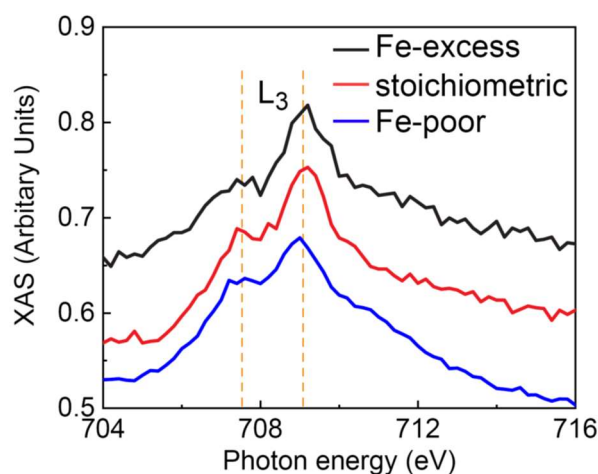


Fig. R17. The Fe $L_{2,3}$ -edge XAS of the 2 u.c. FGT with different Fe/Ga ratio

Additional points for improvement:

Comment 6: *Impact of Capping Layers on NV Center Measurements: If the NV center measurements are conducted ex situ, it is crucial to discuss the potential impact of capping layers on these measurements. A brief commentary on how capping layers might affect the accuracy and reliability of NV center measurements would enhance the comprehensibility of the study.*

Reply: Currently, the maximum external magnetic field of the scanning NV spin magnetometry is limited to 500 Oe, which is not strong enough to completely magnetize the monolayer FGT. Following the suggestion from Reviewer #1 (*Comment 9*), the present magnetization measurement and transport data are enough to draw the conclusion that the enhanced ferromagnetism with high- T_C (above RT) in epitaxial

growth FGT is driven by the tensile strain effect. Consideration that other reviewers' suggestion and limitation of the scanning NV spin magnetometry, we prefer to remove this part in the revised manuscript.

Comment 7: *Arrott Plot Analysis: Including Arrott plot analysis is recommended for an accurate determination of the Curie temperatures. Arrott plots can provide valuable insights into the ferromagnetic phase transitions and improve the reliability of the reported Curie temperatures.*

Reply: Thanks for the insightful suggestions. To accurately determine the T_C of 1 u.c. FGT film, we measured temperature-dependent AHE from 327 K to 382 K with temperature interval of 3 K. We replot the Hall data in plots of R_H^2 as a function of $\mu_0 H/R_H$ (**Fig. R18**), where $\mu_0 H$ is magnetic field, and R_H is Hall resistance. The T_C can be extrapolated by the Arrott plots intercepts through fitting high-field linear part of the R_H^2 axis [*Nature* **563**, 94–99 (2018)]. Therefore, the T_C of 1 u.c. FGT is deduced to be ~ 345 K. We have done corresponding revision in the revised manuscript (Page 9, line 18).

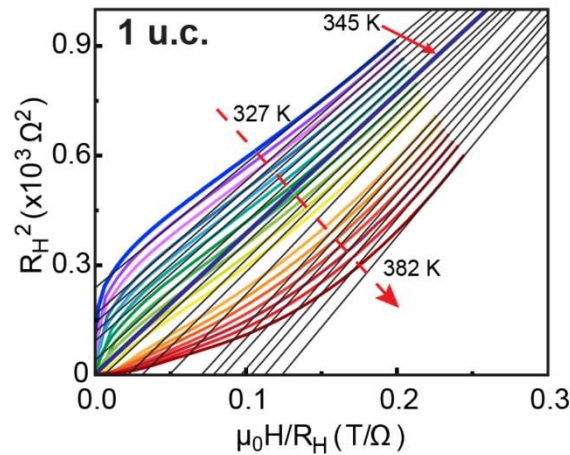


Fig. R18. The T_C can be extrapolated by the Arrott plots intercepts through fitting high-field linear part.

Reply to reviewers' comments

Manuscript Number: NCOMMS-23-51330

We are appreciative of the constructive suggestions and comments from all the reviewers. Accordingly, we have carefully revised the manuscript to improve the quality. Detailed point-by-point Replies to the comments are provided below. We also provide a marked version of the revised manuscript in which all the changes were highlighted in yellow.

Reviewer #1

Comment 1: *In the 2nd paragraph of page 8, the authors wrote that "A unique nature of layered vdW ferromagnets is that interlayer interactions are comparatively weak, ~". In the 1st paragraph of page 9, however, the authors mentioned that "~the corresponding H_C dramatically decreases from 2360 Oe to 230 Oe at 300K, revealing large interlayer interaction." These two sentences are not aligned to each other, and the authors must revise them. I guess, it may be more plausible to argue that the origin of decrease of H_C during decreasing thickness is due to the weakening of magnetic anisotropy or others.*

Reply: Thank you for the suggestion. We have revised the description to “As the thickness increases from 1 u.c. to 9 u.c., the corresponding H_C increases significantly from 230 Oe to 2360 Oe at 300 K, which could be ascribed to the enhancement of magnetic anisotropy energy with increasing thickness.”

Comment 2: The authors must provide the AFM measurement data of samples with various thickness. The authors have confused the unit cell and monolayer and I think it is better to clarify this issue by providing AFM images.

Reply: That’s a good point. We have performed the AFM measurements of the epitaxial Fe_3GaTe_2 at various growth time, as shown in **Figure R1**. Initially, oriented nucleation seeds formed on the surface of the F-Mica substrate. These nuclei expended laterally and gradually evolved into single-crystal domains. The coalescence of these domains led to a contiguous single-crystal film. The height of the atomically flat domains was measured at approximately 1.65 nm, which corresponds to the thickness of one unit cell

of Fe_3GaTe_2 . This indicates that the film grew unit cell by unit cell, rather than monolayer by monolayer.

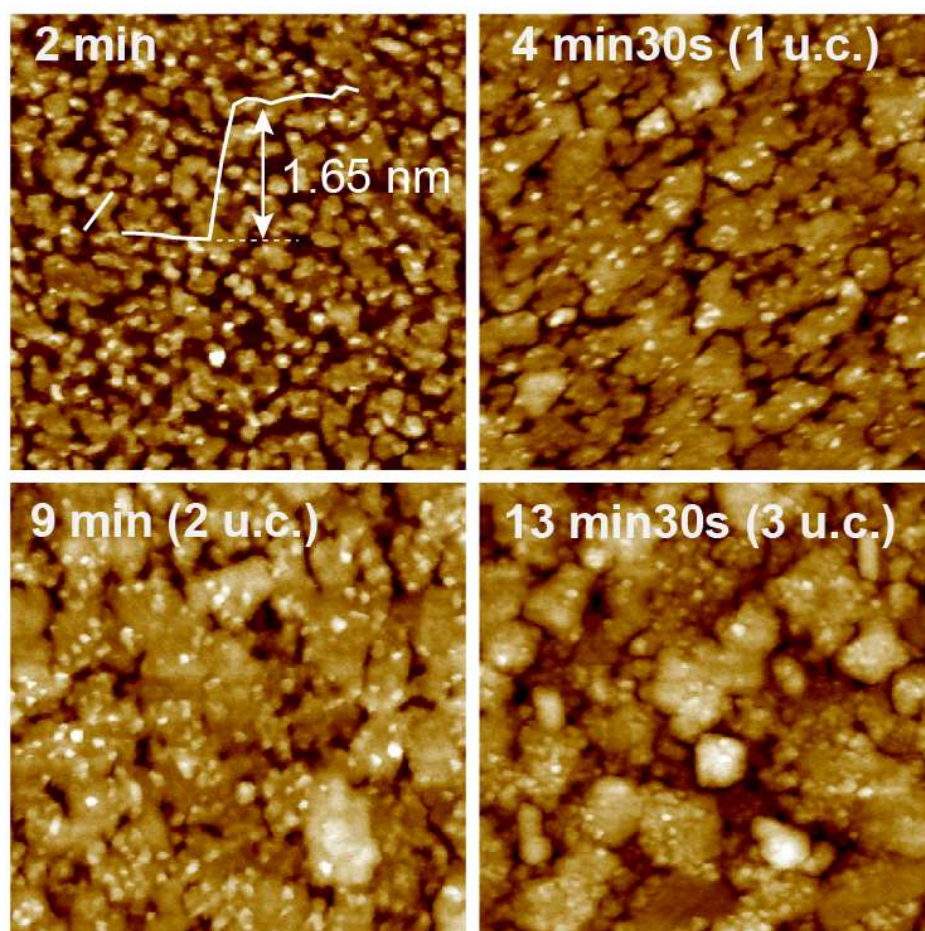


Figure R1 AFM images of the epitaxial Fe_3GaTe_2 at various growth times.

Minor comments

1. The electrical measurement data is shown in supplementary figure 8, not 6.
2. The resistivity of Fe_3GaTe_2 with various devices are shown in supplementary figure 10, not 9.
3. The author name of ref. 15 is miswritten.
4. The color legend for figure 4e is not consistent with its graph. Please change the color.
5. The caption for Supplementary figure 14 is not fully written. Also, the color legend for supplementary figure 14.c is different with its graph.

Reply: Thanks for the gentle reminder. We have carefully corrected these mistakes.

Reviewer #2

Comment 1: *The authors use the dM/dT plot to determine the T_C from M - T curves, which is good. However, if the T_C of 1 u.c. FGT is about 350 K (Figure 3d), why does the clear hysteresis loop still maintain at 400 K (Figure 3e)?*

Reply: That's a good question. One possible explanation for this observation is the presence of magnetic anisotropy or coercivity that persists slightly above the T_C [*Phys. Rev. B* **103**, 125418, (2021)]. In certain materials, particularly those with strong spin-orbit coupling or magnetic anisotropy, remnants of magnetic ordering could remain above the T_C . Specifically, although the bulk magnetic order has diminished, localized regions or domains would still exhibit magnetic hysteresis due to the anisotropic effect. Additionally, while the determination of T_C from the dM/dT plot indicates the point where long-range magnetic order transitions to paramagnetic behavior, short-range interactions or domain effects might still cause hysteresis loops above this temperature. Therefore, while T_C represents the temperature at which long-range magnetic order is lost, it is possible for magnetic hysteresis to be observed at temperatures slightly above T_C .

Comment 2: *If the authors acknowledge that the significantly higher Curie temperature of epitaxial FGT comes from strain effects, the title should not be referred to as "intrinsic ferromagnetism". It is not correct to claim intrinsic since there is strong extrinsic effect. The relevant statements in the main text should also be revised..*

Reply: Thanks for the suggestion. We have revised the description in the manuscript.

Comment 3: *The proof for the existence of strain effect and the effect to induce the high T_C is not solid. More experimental evidence for the existence of strain effect should be given.*

Reply: That's a good point. The in-plane lattice constants of the epitaxial FGT films on F-Mica were calculated to be $a = b = 4.101$ Å, which exceeds that of bulk value of 3.986 Å, demonstrating that the epitaxial FGT films are subjected tensile strain. We further carried out HAADF-STEM measurement from FGT/F-mica interface to FGT surface, as shown in **Figure R2**. The STEM image of the FGT confirms that the in-plane lattice constants remain constant from the interface to the surface and are consistently larger than the bulk value, revealing that epitaxial film retains tensile

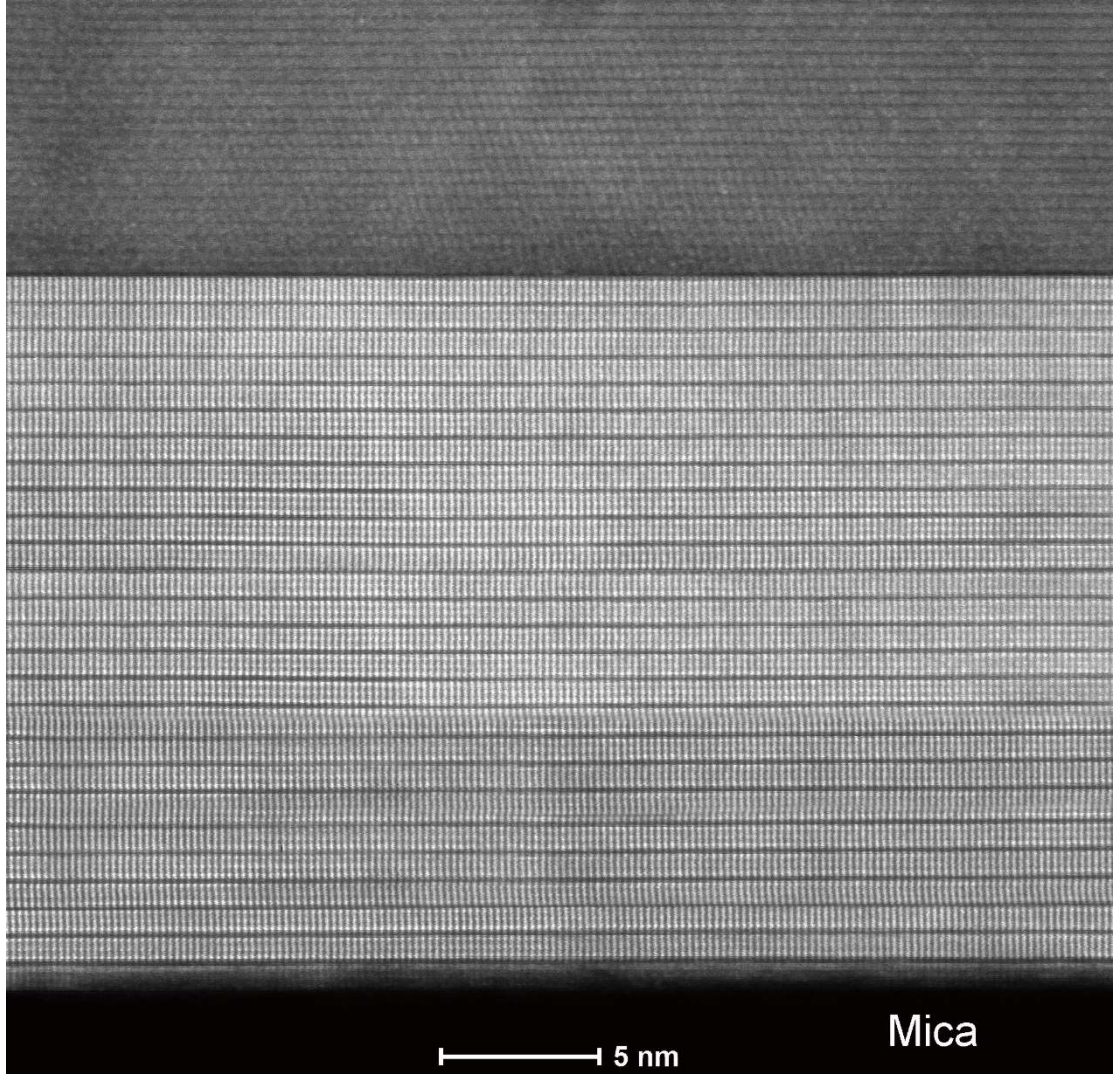


Figure R2 HAADF-STEM image confirms that the in-plane lattice constants remain constant from the interface to the surface and are consistently larger than the bulk value, revealing that epitaxial film retains tensile strain without relaxation.

strain without relaxation.

To elucidate significant enhancement of Curie temperature due to strain effect, we have grown a 1 u.c. FGT film on F-mica using a 1 u.c. PtTe₂ buffer layer. The PtTe₂ exhibits a hexagonal lattice symmetry with lattice constants $a = b = 4.026 \text{ \AA}$ and $c = 5.221 \text{ \AA}$ [*Nat. Commun.* **15**, 2605 (2024)]. The lattice mismatch between FGT and PtTe₂ is approximately 0.1 %, which is half that of FGT/F-mica. **Figure R3** presents the M - H loops at various temperatures, alongside the temperature-dependent magnetization derivative (dM/dT) for both FGT/PtTe₂/F-mica and FGT/F-mica. As shown in **Figure R3(b)** and **(d)**, the T_C of the FGT/PtTe₂/F-mica is approximately 330K, which is lower than that ($T_C = 345 \text{ K}$) of FGT on F-mica. These results reveal that the strain effect

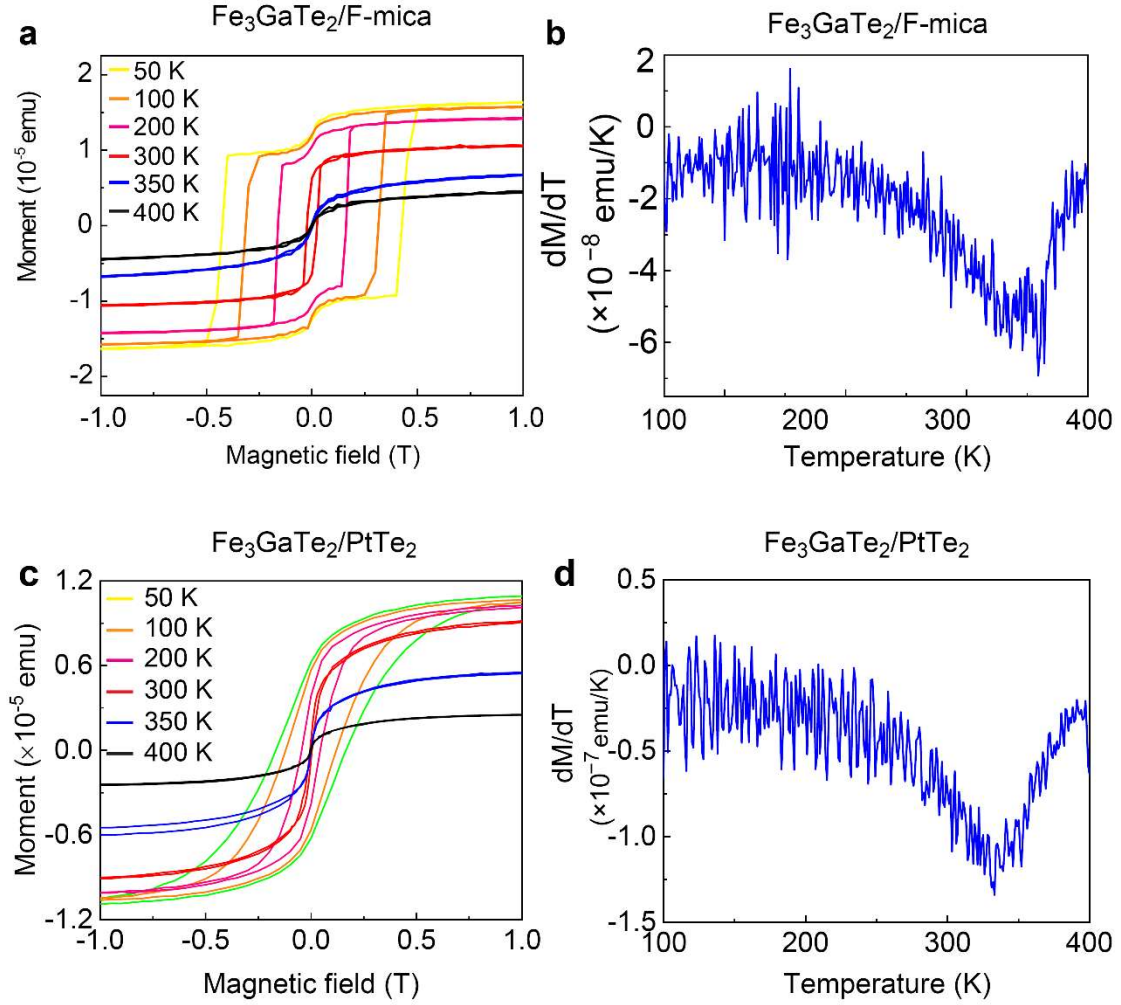


Figure R3. M - H loops at various temperatures and dM/dT for FGT/F-mica and FGT/PtTe₂/F-mica.

would significantly enhance the ferromagnetism of the epitaxial FGT.

To theoretically investigate the enhancement of the T_C in the epitaxial FGT on F-mica substrate, we performed density functional theory (DFT) calculations with Vienna Ab-initio Simulation Package (VASP) software and the Monte Carlo (MC) simulation implemented in the VAMPIRE software package, as shown in [Figure R4](#). For the 1 u.c. FGT without strain, the T_C is calculated to be 303 K, which is consistent with reported value of approximately 295 K for exfoliated FGT of same thickness (*npj 2D Materials and Applications* **8**, 22, 2024). Notably, with application of 0.1% tensile strain, the T_C dramatically increases to 345 K, and further increases to 396 K under 0.4% strain. The theoretical results indicate that the strain effect from the substrate would significantly enhance the T_C of FGT.

To investigate the mechanisms underlying the enhancement of the T_C , we employed DFT calculations to analyze the magnetic exchange couplings in the FGT

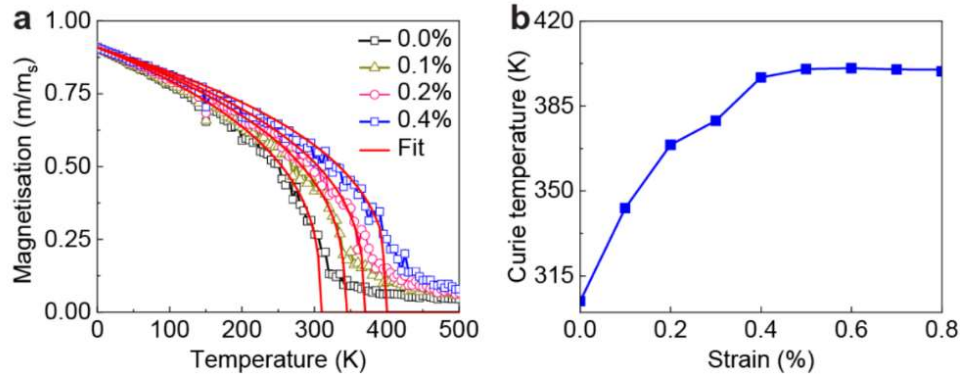


Figure R4 Temperature-dependent magnetization calculated by Classical Heisenberg model Monte Carlo simulations. With increasing strain, the T_C is dramatically enhanced.

under varying strains. The interactions are categorized into three groups, namely (i) interplane interactions, which include J_{12} (interactions between atoms Fe₁-Fe₂) and J_{25} (Fe₂-Fe₅), (ii) in-plane exchanges, comprising J_{22} (Fe₂-Fe₂) and J_{55} (Fe₅-Fe₅), (iii) interlayer coupling between adjacent layers, which involve J_{14} (Fe₁-Fe₄) and J_{23} (Fe₂-Fe₃). Exchange interactions occur between the Fe species located at various Fe-Fe distances, extending up to 16 Å, beyond which they saturate to zero. We have considered the evolution of the comparatively large interactions (e.g., J_{12} , J_{25} , J_{22} , and J_{14}) as a function of distance, between an atom and its atomic neighbor. The ferromagnetic ground state of the FGT arises from robust ferromagnetic exchange couplings and a substantial number of ferromagnetic neighbors, as shown in [Figure R5 \(a\)-\(e\)](#). Under tensile strain, the interlayer exchange coupling parameters significantly increase, particularly J_{12} , which enhances by approximately 50% at 0.6% strain. This trend aligns with the increase in T_C predicted by Monte Carlo simulations. Consequently, the strain-enhanced ferromagnetism in FGT could be primarily attributed to the alterations of magnetic exchange coupling. Additionally, magnetic anisotropy energy (MAE) is also crucial for stabilizing ferromagnetism by suppressing thermal fluctuations that would otherwise disrupt long-range ferromagnetic order¹¹. [Supplementary Figure 15 \(g\)](#) illustrates the variation of the MAE under tensile strain, exhibiting significantly increase of the MAE with strain. Consequently, we conclude that the enhancement in the T_C of the epitaxial FGT results from strain-induced changes in both MAE and magnetic exchange coupling.

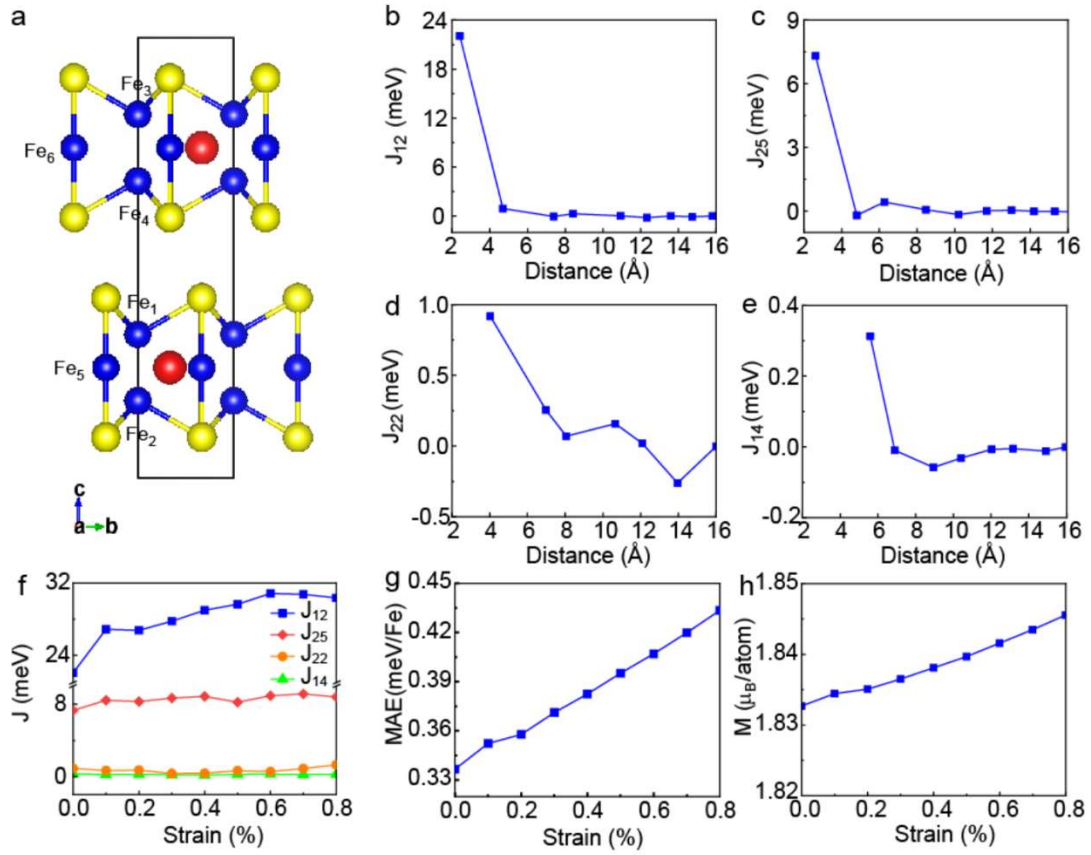


Figure R5 (a) Lateral view of a unit cell of FGT. Exchange interactions (b) J_{12} , (c) J_{25} , (d) J_{22} and (e) J_{14} for bulk FGT along with their evolution with distance to a maximum 16 $\text{\AA}$. Evolution of (f) exchange coupling parameters, (g) MAE, and (h) average magnetic moment with increase of tensile strain.

Reviewer #3

Comment 1: First of all, I would like to thank the authors for attempting the Arrott plot analysis, which has corrected the T_C for the 1 u.c. sample from 380K to 345K. It would be beneficial to apply this method to all samples to ensure consistency and accuracy of the T_C values..

Reply: We appreciate the suggestion. We have further measured the Arrott Plots of the epitaxial FGT films with varying thickness. The T_C of the epitaxial FGT increase dramatically with thickness. When the thickness exceeds 6 u.c., the T_C surpasses 400 K, exceeding the measurement limit of our PPMS system.

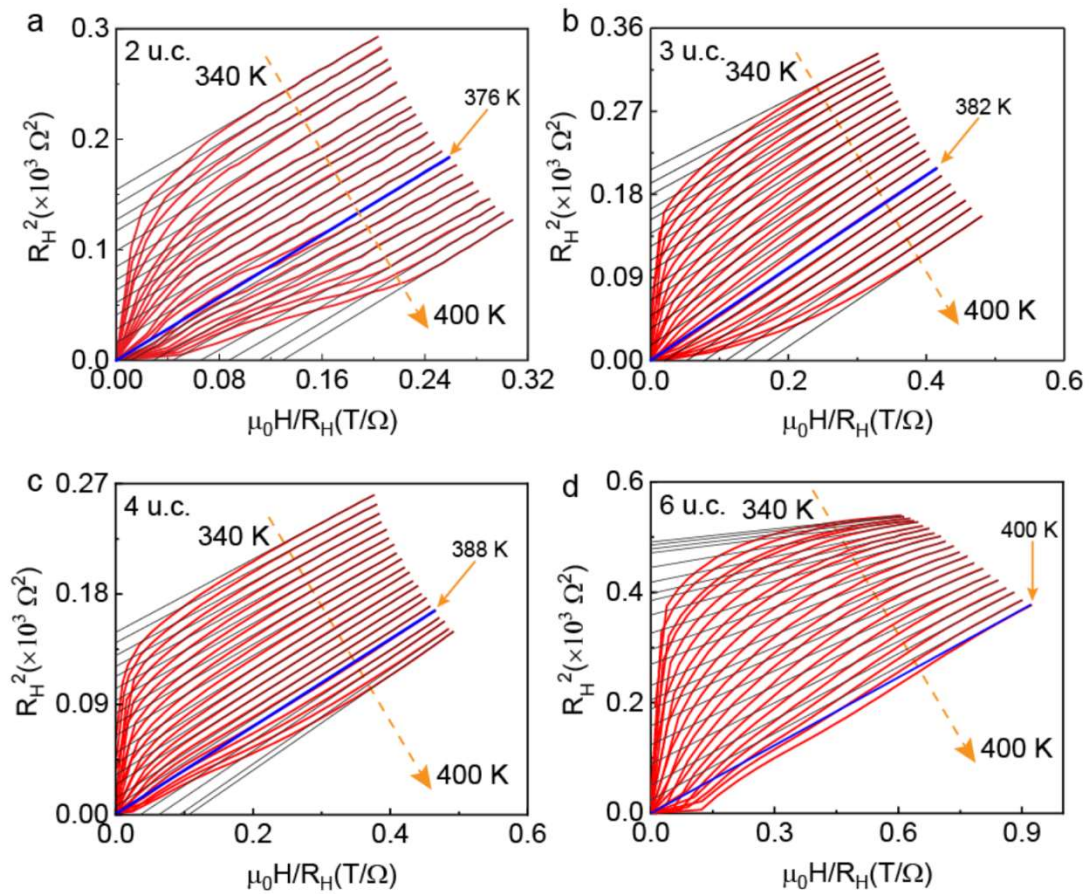


Figure R6 Arrott Plots of the epitaxial FGT films with varying thickness.

Table R1. Summary of thickness-dependence of T_C extrapolated by the Arrott plots.

Thickness	1 u.c.	2 u.c.	3 u.c.	4 u.c.	6 u.c.
T_C (K)	345	376	382	388	400

Comment 2: *The observed increase in T_C from 1 u.c. to 9 u.c., and the peak T_C at 9 u.c., remains unclear. This aspect is a key highlight in the abstract and warrants a thorough explanation. Given that strain typically decreases with increasing thickness, it is crucial to examine the relationship between T_C , the number of u.c., and the strain percentage in detail for all 9 thickness. A table summarizing these parameters for all thicknesses would be necessary to elucidate this relationship.*

Reply: Thank you for the insightful point. To better understand stress state of the epitaxial FGT on the F-Mica substrate, we performed HAADF-STEM measurement of the FGT from FGT/F-mica interface to FGT surface, as shown in **Figure R7**. The STEM image of the FGT confirms that the in-plane lattice constants remain unchanged from FGT/Mica interface to FGT surface. The in-plane lattice constant of the epitaxial FGT films on F-Mica is calculated to be $a = b = 4.101 \text{ \AA}$, larger than bulk value of $3.986 \text{ \AA}$, demonstrating that the epitaxial FGT films are under tensile stress without relaxation. Therefore, we suggest that the increase in T_C of FGT from 1 u.c. to 9 u.c. is likely not due to the strain evolution with thickness.

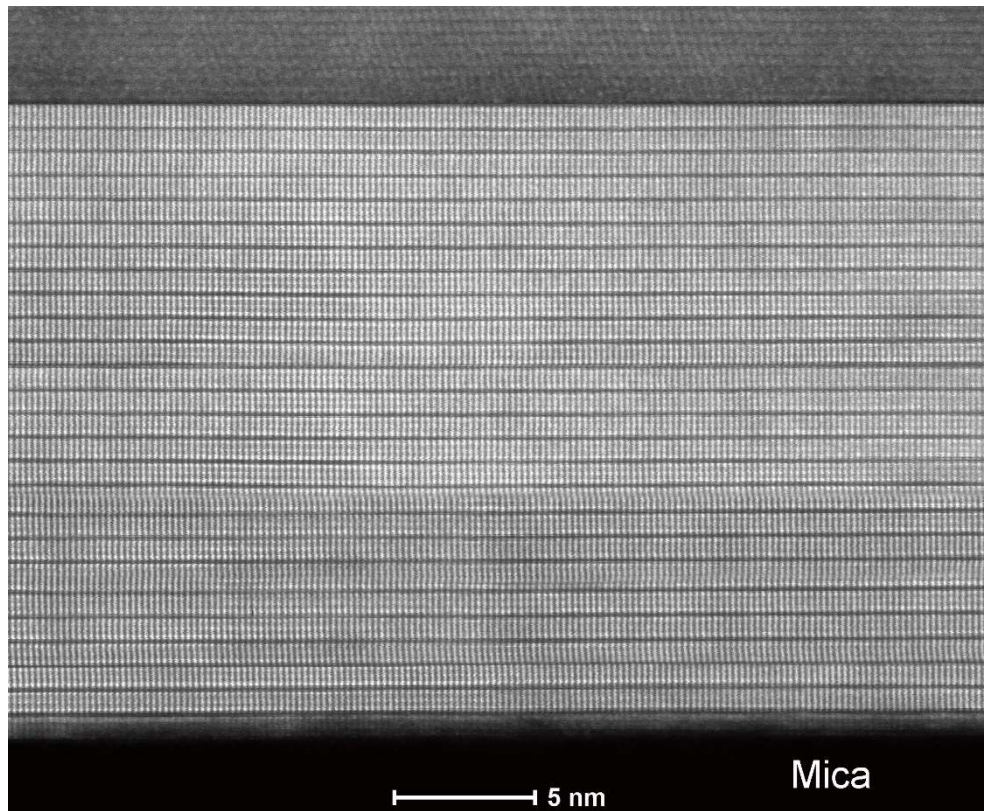


Figure R7 HAADF-STEM image of the FGT confirms that the lattice constants keep invariable from FGT/F-Mica interface to FGT surface and are larger than that of bulk, revealing that epitaxial film is under tensile stress without relaxation.

Magnetic anisotropy energy (MAE) plays a crucial role in sustaining ferromagnetism, significantly influencing the T_C by suppressing thermal fluctuation that destroy long-range order [Nature, **546**, 265 (2017)]. **Figure R8** shows the MAE of the epitaxial FGT films at 300 K, demonstrating dramatic increase with thickness. Therefore, we suggest that the enhancement of T_C in the epitaxial FGT with increasing thickness can be ascribed to the substantial increase of anisotropy energy.

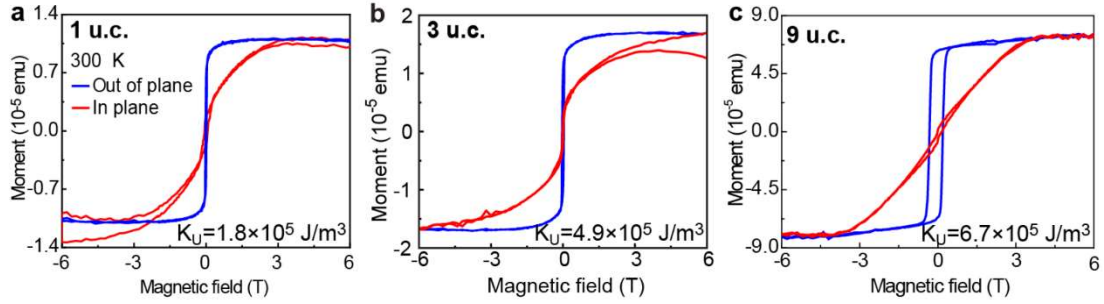


Figure R8 Magnetic anisotropy energy of the epitaxial FGT films with different thickness at 300 K, which increases with the thickness.

Comment 3: Furthermore, it is important to explore and discuss any additional factors strain that could contribute to the observed enhancement in T_C . Such a discussion would provide a more comprehensive understanding of the underlying mechanisms at play.

Reply: That's a good suggestion. To theoretically understand the T_C enhancement of the epitaxial FGT on F-mica substrate, we performed density functional theory (DFT) calculations with Vienna Ab-initio Simulation Package (VASP) software and the Monte Carlo (MC) simulation implemented in the VAMPIRE software package, as shown in **Figure R9**. For the 1 u.c. FGT without strain, the T_C is calculated to be 303 K, which is consistent with reported value (approximately 295 K) of exfoliated FGT with same thickness [npj 2D Materials and Applications **8**, 22 (2024)]. However, with application of 0.1% strain, the T_C dramatically increases to 345 K, and further increases to 396 K under 0.4%. The theoretical results reveal that the strain effect significantly enhance the T_C .

To investigate the origin of the enhanced T_C , magnetic exchange couplings in FGT under different strains were determined using DFT calculations. The interactions were categorized into three groups, namely (i) interplane interactions, including J_{12}

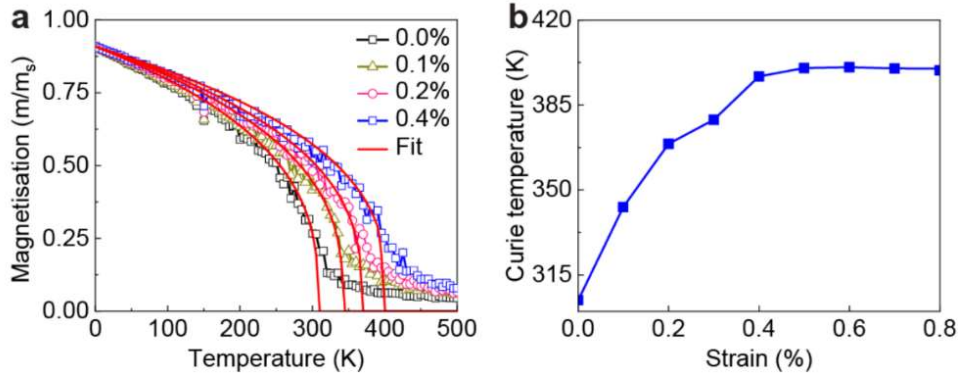


Figure R9 Temperature-dependent magnetization calculated by Classical Heisenberg model Monte Carlo simulations. With the strain increasing, the T_C is dramatically enhanced.

(interactions between atoms $\text{Fe}_1\text{-Fe}_2$) and J_{25} ($\text{Fe}_2\text{-Fe}_5$); (ii) in-plane exchanges, comprising J_{22} ($\text{Fe}_2\text{-Fe}_2$) and J_{55} ($\text{Fe}_5\text{-Fe}_5$); and (iii) coupling between adjacent layers, such as J_{14} ($\text{Fe}_1\text{-Fe}_4$) and J_{23} ($\text{Fe}_2\text{-Fe}_3$). These interactions occur between Fe atoms at varying Fe-Fe distances. The analysis considered the evolution of the comparatively large interactions (e.g., J_{12} , J_{25} , J_{22} , and J_{14}) as a function of atomic distance up to a maximum of 16 Å, where interactions decay to zero. The ferromagnetic ground state of FGT is driven by strong exchange coupling and a large number of ferromagnetic neighbors, as shown in **Figure R10 (a)-(e)**. Under tensile strain, the intralayer exchange coupling parameters increase significantly, with J_{12} enhanced by $\sim 50\%$ at 0.6% strain, consistent with the trend of the T_C increase predicted by the Monte Carlo simulation, as shown in **Figure R10 (f)**. This suggests that the strain-enhanced ferromagnetism in FGT primarily arises from modifications in magnetic exchange coupling.

Additionally, magnetic anisotropy energy (MAE) is also crucial for stabilizing ferromagnetism by suppressing thermal fluctuations that would otherwise disrupt long-range ferromagnetic order¹¹. **Figure R10** illustrates the variation of the MAE under tensile strain, exhibiting significantly increase of the MAE with strain. Consequently, we conclude that the enhancement in the T_C of the epitaxial FGT results from strain-induced changes in both MAE and magnetic exchange coupling.

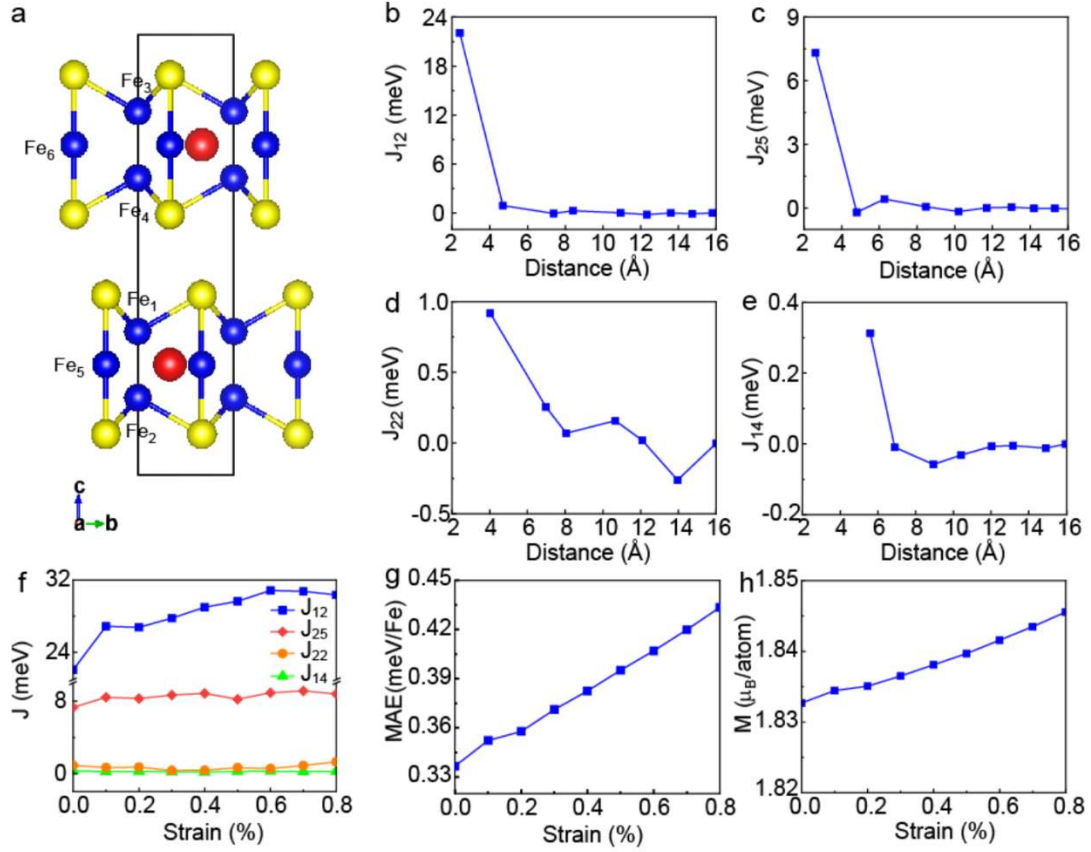


Figure R10 (a) Lateral view of a unit cell of FGT. Exchange interactions (b) J_{12} , (c) J_{25} , (d) J_{22} and (e) J_{14} for bulk FGT along with their evolution with distance to a maximum 16 Å. Evolution of (f) exchange coupling parameters, (g) MAE, and (h) average magnetic moment with increase of tensile strain.