Direct evidence of ferromagnetism in a quantum anomalous Hall system

Wenbo Wang1, Yunbo Ou2, Chang Liu3, Yayu Wang2,3, Ke He2,3, Qi-Kun Xue2,3 and Weida Wu1*

Quantum anomalous Hall (QAH) systems are of great fundamental interest and potential application because of their dissipationless conduction without the need for an external magnetic field. The QAH effect has been realized in magnetically doped topological insulator thin films. However, full quantization requires extremely low temperature (T < 50 mK) in the earliest works, although it has been significantly improved by modulation doping or co-doping of magnetic elements. Improved ferromagnetism has been shown in these thin films, yet direct evidence of long-range ferromagnetic order is lacking. Herein, we present direct visualization of long-range ferromagnetic order in thin films of Cr and V co-doped (Bi, Sb)2Te3 using low-temperature magnetic force microscopy with in situ transport. The magnetization reversal process reveals typical ferromagnetic domain behaviour — that is, domain nucleation and possibly domain wall propagation — in contrast to much weaker magnetic signals observed in the endmembers, possibly due to superparamagnetic behaviour. The observed long-range ferromagnetic order resolves one of the major challenges in QAH systems, and paves the way towards high-temperature dissipationless conduction by exploring magnetic topological insulators.

Dissipationless conduction is technologically appealing because of a wide range of potential applications. There are two ways to achieve dissipationless conduction in condensed matter systems — through superconductivity and through topological chiral edge states. For this reason, high-temperature superconductivity has been extensively investigated for decades. Dissipationless conduction due to chiral edge states is realized in the Quantum Hall effect (QHE), which requires low temperature and an external magnetic field. The closely related phenomenon, the quantum anomalous Hall effect (QAHE), however, does not require an external magnetic field. The realization of the QAHE requires the breaking of time-reversal symmetry and a topologically nontrivial band structure with the Fermi level inside the bandgap. There have been a number of theoretical proposals, and eventually it was realized experimentally in magnetically doped three-dimensional topological insulator thin films. Here the ferromagnetism, induced by doping magnetic elements, breaks time-reversal symmetry, and opens a mass gap at the Dirac point of the topological surface states. By tuning the Fermi level inside the mass gap, the magnetic topological insulator thin film is equivalent to two copies of half-integer QHE systems. Quantized Hall conduction was first observed at ultralow temperature (−30 mK) in Cr-doped Bi2Sb2−xTex (BST) thin films synthesized by molecular beam epitaxy (MBE). This observation was soon confirmed by other groups. Later, a robust QAHE with higher precision in quantization was observed in a V-doped BST thin film, which is a harder ferromagnet with a greater coercive field (Hc) and a higher Curie temperature (Tc) with the same doping level. Nevertheless, ultralow temperature (T < 50 mK) is needed to achieve full quantization. Therefore, it is imperative to understand the origin of the requirement for ultralow temperature for full quantization.

Magnetic inhomogeneity has been proposed to be one of the main factors that limit the QAHE temperature. Disordered ferromagnetic or superparamagnetic behaviour and electronic inhomogeneity have been reported in Cr-doped BST thin films. Since the mass gap is proportional to the strength of the exchange interaction, the reduced QAHE temperature is probably limited by the regions with the weakest exchange. Although modulation doping of Cr was shown to improve the quantization temperature in peraly-layer thin films, it is unclear whether it reduces magnetic inhomogeneity. On the other hand, recent angle-resolved photoemission spectroscopy (ARPES) studies of V-doped BST thin films suggest that the valance band maximum (VBM) is above the Dirac point. Therefore, an ultralow temperature is needed to localize the bulk states near the Dirac point along with sufficient disorder. These mechanisms for lowering the quantization temperature indicate that Cr and V co-doping could be a viable way to reduce magnetic inhomogeneity while enhancing the mobility gap of localized states. Empirically, alloy doping is commonly known as an effective route to improve ferromagnetic order in a diluted magnetic semiconductor.

Indeed, an increased QAHE temperature was observed in Cr- and V-doped BST thin films. At the optimal Cr/V ratio, full quantization was achieved at 300 mK, an order of magnitude higher than the endmembers with single dopants. The Hall hysteresis loop is more square-like, suggesting a sharper magnetization reversal (that is, reduced magnetic inhomogeneity). Furthermore, the temperature dependence of the anomalous Hall resistance is more field-like. These observations indicate improved ferromagnetism in Cr/V co-doped topological insulator thin films. However, direct microscopic evidence of long-range ferromagnetic ordering is still lacking. Note that the intrinsic anomalous Hall effect is determined by the Berry phase of the occupied bands, which is independent of the magnitude of the magnetization. In this letter, we report a systematic study of Cr/V co-doped BST thin films using magnetic force microscopy (MFM). Our MFM results reveal clear ferromagnetic domain behaviour of the magnetization reversal process in the optimally doped BST thin films, confirming long-range ferromagnetic ordering in this QAHE system, presumably via the van Vleck mechanism. Furthermore, the ferromagnetism of co-doped thin films is robust against a significant change in the bulk charge carrier density, although the exchange interaction is enhanced by

1Department of Physics and Astronomy, Rutgers University, Piscataway, NJ, USA. 2State Key Laboratory of Low Dimensional Quantum Physics, Department of Physics, Tsinghua University, Beijing, China. 3Collaborative Innovation Center of Quantum Matter, Beijing, China. 4e-mail: wdwu@physics.rutgers.edu
Figure 1 | Schematic of the in situ transport set-up and the Cr concentration ($y$) dependence of $\sigma_y$ and $H_y$/FWHM$^{\text{a,b}}$. a, Schematic of the Hall bar device for MFM and in situ transport measurements. The 5 quintuple layers (QLs) Cr/V co-doped BST thin film was grown on a STO(111) substrate using MBE, followed by deposition of a 15 nm layer of Au film. Both the Au film and the magnetic tip were grounded to eliminate any electrostatic interactions between them. A back-gate voltage $V_g$ was applied to the bottom electrode to tune the charge carrier density. The Hall resistance $\rho_{xy}$ and longitudinal resistance $\rho_{xx}$ were obtained by measuring $V_{yx}$ and $V_{xx}$, respectively. Cr concentration ($y$) dependence of the zero magnetic field Hall conductance $\sigma_y$ (blue) and the ratio of coercivity ($H_c$) to the full-width at half-maximum (FWHM) of the magnetoresistance (MR) at 1.5 K.

The direct evidence of long-range ferromagnetic order eases concerns about the fragility of the QAHE due to magnetic inhomogeneity, alleviating the need for ultralow temperature to achieve full quantization. Our results should encourage further exploration of the QAHE and related phenomena in magnetically doped topological materials for dissipationless conduction at elevated temperature.

Figure 1a shows a schematic of the Hall bar device of the magnetic topological insulator thin films fabricated for MFM and in situ transport measurements. Three (Cr$_{V_{y=0}}$, Bi$_{y=0.16}$, Sb$_{y=0.4}$)$_{10}$Te$_3$ films ($y=0$, 0.16, 1, respectively, and $x \approx 0.4$) are fabricated into the Hall bar devices. All MFM data presented here were taken at 5 K. As shown in Fig. 1b, the $y=0.16$ film is the optimized sample ($T_c \approx 28$ K) with the best ferromagnetic behaviour (sharpest reversal) and the highest Hall conductance (at 1.5 K). In addition, the temperature dependence of the Hall resistance is more field-like, indicating robust ferromagnetism$^{16}$. The longitudinal resistance starts to decrease immediately below $T_c$, indicating that the sample enters the QAHE regime as soon as the long-range ferromagnetic order forms (see Supplementary Fig. 1). The end-member ($y=0$ or 1), however, enters the QAHE regime at a much lower temperature. A gate voltage ($V_g$) was applied to the back of the SrTiO$_3$(111) (STO) substrate to tune the Fermi level. At 1.5 K, the Hall resistance reaches $0.95 h/e^2$ (where $h$ is Planck’s constant and $e$ is the electronic charge) at $V_g^0$ (Supplementary Fig. 1). Such a quantization level was achieved only below 50 mK in single Cr- or V-doped thin films$^{15,16,24}$.

Figure 2a,h shows MFM images and in situ transport data ($\rho_{xx}$ and $\rho_{yy}$) of the optimally doped film ($y=0.16$) at $V_g^0 \approx 10$ V at various magnetic fields. The $\rho_{yy}(H)$ loop shows a saturation $\approx 0.5 h/e^2$ with a coercive field $H_c \approx 0.26$ T. The magnetization reversal process from downward (red) to upward (blue) magnetized states is illustrated in the MFM images. The downward saturated state has very weak magnetic contrast with a small positive field ($+0.05$ T), indicating the single-domain state persists at a small reversed field, showing compelling evidence of robust ferromagnetism. The observed stable single-domain state, with little relaxation, is in sharp contrast to the superparamagnetic behaviour previously reported in Cr-doped BST films, where significant relaxation was already observed at small magnetic fields$^{5}$. (See Supplementary Fig. 1 for transport results.)

At 0.15 T, up domains start to nucleate, represented by light blue regions. As the field increases further, the up domains expand and the down domains shrink. At the coercive field $H_c$ where $\rho_{xx}=0$ and $\rho_{yy}$ peaks at $\approx 0.8 h/e^2$, equally populated up and down domains were observed, confirming the zero-magnetization state ($M=0$). For $H > 0.35$ T, no red regions are visible in the MFM images, indicating the system is in a saturated (single domain) state. The MFM observation of ferromagnetic domain behaviour is in excellent agreement with the in situ transport data, suggesting the local observation is representative of the global (bulk) properties. Note that the Hall data at 1.5 K and 5 K show similar square-like hysteresis loops, indicating no qualitative difference between these two temperatures. Furthermore, a higher coercive field and sharper reversal observed at the lower temperature suggest better ferromagnetism. Therefore, the observed ferromagnetic behaviour is expected to persist at the lower temperature, where full quantization was observed on the same sample$^{16}$.

The ferromagnetic domain behaviour can be further illustrated using the difference between the MFM images of adjacent fields, as shown in Fig. 2a. The locations where the changes in MFM signal are above the noise level ($\pm 2 \text{ mHz}$) are defined as the newly reversed regions. They are marked in different colours for each field value, which are shown in the right column of Fig. 2a. The unchanged areas are marked in white. In Fig. 2b–g, these differential images are stacked together to show the spatial correlations of magnetization reversal events so as to differentiate isolated domain nucleation from domain expansion (see Supplementary Fig. 5 for the complete data set). For example, as shown in Fig. 2c, some of the newly reversed regions (yellow) at 0.2 T have no correlation with previously reversed regions (red). These regions, labelled by dashed squares, are isolated nucleation sites. The other yellow regions, labelled by solid circles, have some overlap with the red regions, probably due to tilted domain walls. This behaviour is consistent with domain growth via either domain wall propagation or domain-wall-induced nucleation. The domain behaviour is distinctly different from the superparamagnetic behaviour of random switching events in a previous report$^{17}$.

In addition to direct visualization of ferromagnetic domain behaviour, MFM data can also be used to extract the hysteresis loop of the normalized magnetization ($M_s/M_s$) vs. field ($H$) curve to estimate the population of up and down domains$^{18}$. As shown in Fig. 2h, the $M_s/M_s(H)$ curve agrees quantitatively with the $\rho_{yy}(H)$ loop. The agreement between local (domain population) and global ($\rho_{xx}$) measurements demonstrates that our MFM results are representative of the bulk magnetic properties. Consistently, the domain contrast, estimated by the root-mean-square value of the MFM signal ($\delta f_{\text{rms}}$), peaks at $H_c$ when up
and down domains are equally populated. The observed domain behaviour provides unambiguous evidence of long-range ferromagnetic order in the optimally Cr/V co-doped BST thin films. In contrast, MFM measurements on single-doped BST films do not reveal clear ferromagnetic behaviour (see Supplementary Figs. 3 and 2 for MFM results of endmembers). Therefore, our MFM data provide direct evidence that long-range ferromagnetic order is essential for the enhancement of the QAHE18–20.

Long-range ferromagnetic order is one of the key ingredients of the QAHE. Yu et al. proposed a Van Vleck mechanism in magnetically doped topological insulators, where the exchange interaction between local moments is mediated by band electrons with significant Van Vleck susceptibility35–38. Therefore, the Van Vleck mechanism is independent of the bulk carrier density. However, other studies indicate that an RKKY-type exchange plays a significant role when bulk or surface carriers are present39–41. To shed light on the exchange mechanisms, we investigate the bulk carrier dependence of the ferromagnetism by applying a gate voltage. Similar to the neutral point $V_g^0$ case, both electron-doped (300 V) and hole-doped ($-300$ V) states show typical ferromagnetic domain behaviour, confirming that long-range ferromagnetic order is robust against tuning the Fermi level near the neutral point (see Supplementary Figs. 6 and 7), supporting the presence of a Van Vleck mechanism. Figure 3a–c shows the MFM images at $H_g$ and the nucleation maps at the three gate voltages. Comparing the three multi-domain states, hole-doping results in a larger domain size, fewer nucleation sites, and stronger domain contrast, whereas electron-doping results in an opposite trend. Consistently, $H_g$ is enhanced (suppressed) by hole- (electron-) doping, as shown in the $\rho_{sv}(H)$ loops in Fig. 3d. On the other hand, both hole- and electron-doping away from the neutral point suppress the AHE. Note that the domain contrast of the multi-domain state is proportional to the saturated magnetization45. The enhanced domain contrast at $H_g$ indicates an increase in saturated magnetization with hole-doping. Compared to the local magnetic moment density, the gate-induced charge carrier density is negligible (see Supplementary Information section D for an estimation), so the enhanced magnetization at zero temperature $M_s(0)$ is independent of the gate voltage. Therefore, the enhancement of the magnetization at 5 K indicates a decrease in the reduced temperature $T/T_C$ (that is, an increase of $T_C$, due to enhanced exchange coupling in the hole-doped films). This is also consistent with a higher $H_g$ and fewer nucleation sites in the hole-doped state, suggesting the presence of the RKKY mechanism. Therefore, the gating dependence of the MFM results indicates that both the Van Vleck and the RKKY mechanisms are present in magnetically doped topological insulators. Interestingly, some nucleation sites (labelled by black circles) are independent of the gate voltage, so they are probably caused by neutral defects or imperfections that are insensitive to charge carriers. On the other hand, there are nucleation sites that do depend on the gate voltage, which indicates that they might be related to charged defects.

The $V_g$-dependences of $\rho_{sv}$ and $H_g$ are summarized in Fig. 4a. The $\rho_{sv}(V_g)$ curve was measured at zero magnetic field with slowly ramping of $V_g$ from $-300$ V to $+300$ V after the film was saturated at high magnetic field. The $\rho_{sv}(V_g)$ curve shows a peak at $V_g^0 \approx 10$ V, the charge neutral point. This result agrees well with the saturation state resistance from hysteresis loop measurements (red stars), indicating the single-domain state at zero magnetic field is robust against tuning the bulk carrier density (that is, robust ferromagnetism). In contrast to the monotonic gate dependence of the coercive field $H_c$, $\rho_{sv}$ shows a non-monotonic behaviour, consistent with the proximity of the Dirac point to the VBM in BST, as shown in
Fig. 3 | Gate dependence of ferromagnetic behaviour. a-c, MFM images around the coercive field (top) and nucleation-site (bottom) maps, at $V_g = 300\,\text{V}$, $10\,\text{V}$ and $-300\,\text{V}$, respectively. Larger domain size and stronger domain contrast were observed for $V_g = -300\,\text{V}$ (hole-doping). Black circles label some of the common nucleation sites at three different $V_g$ values. d, Hall resistance (top) and MFM domain contrast ($\delta$) as a function of magnetic field at three different $V_g$ values. The values of $H_c$ deduced from the two panels are consistent with each other, as $H_c \approx 0.21\,\text{T}$ ($V_g = 300\,\text{V}$), $H_c \approx 0.26\,\text{T}$ ($V_g = 10\,\text{V}$), $H_c \approx 0.33\,\text{T}$ ($V_g = -300\,\text{V}$).

Fig. 4 | Gate dependence of $\rho_{yx}$ and $H_c$ and schematic band structure. a, $\rho_{yx}$ and $H_c$ versus $V_g$. Red stars are the zero-field $\rho_{yx}$ from hysteresis loops at $300\,\text{V}$, $10\,\text{V}$ and $300\,\text{V}$. b, Schematic picture of the band structure of the Cr/V co-doped BST film. The Dirac point of the surface state is close to the VBM.

Fig. 4b (refs 10,16,19). The hole-doping will not only enhance the exchange interaction via an RKKY-type exchange mechanism, but will also significantly enhance dissipation, thus reducing $\rho_{yx}$. On the other hand, electron-doping pushes the Fermi level up above the VBM, but presumably below the conduction band minimum (CBM). Therefore, electron-doping probably induces surface state carriers with only slightly enhanced dissipation. Previous ARPES studies of V-doped BST film suggest that the Dirac point is below the VBM, so the mass gap overlaps with bulk states27. In contrast, transport studies indicate the Dirac point is above the VBM for Cr-doped BST films10,16. Therefore, it is possible that Cr-doping in co-doping samples slightly lowers the VBM so that it is easier to localize the bulk states inside the mass gap (see Supplementary Fig. 9). Alternatively, Cr/V co-doping might significantly enhance the scattering of bulk carriers, resulting in enhanced localization of the in-gap bulk states. Further studies on the impact of dopants on magnetic exchange and transport are needed to understand the mechanism behind the enhanced QAHE temperature in co-doped samples. The observed robust ferromagnetism resolves one of the major concerns in magnetically doped topological insulators, and opens a door to the exploration of high-temperature dissipationless conduction with magnetic topological materials.

Methods
Methods, including statements of data availability and any associated accession codes and references, are available at https://doi.org/10.1038/s41567-018-0149-1.

Received: 28 August 2017; Accepted: 20 April 2018; Published online: 28 May 2018

References
1. Haldane, F. D. M. Model for a quantum Hall effect without Landau levels: condensed-matter realization of the ‘parity anomaly’. Phys. Rev. Lett. 61, 206601 (2003).
2. Onoda, M. & Nagaosa, N. Quantized anomalous Hall effect in two-dimensional ferromagnets: Quantum Hall effect in metals. Phys. Rev. Lett. 90, 055502 (2003).
3. Liu, C. X., Qi, X. L., Dai, X., Fang, Z. & Zhang, S. C. Quantum anomalous Hall effect in Hg$_{1-x}$Mn$_x$Te quantum wells. Phys. Rev. Lett. 101, 146802 (2008).
4. Qi, X. L., Hughes, T. L. & Zhang, S. C. Topological field theory of time-reversal invariant insulators. Phys. Rev. B 78, 195424 (2008).
5. Yu, R. et al. Quantized anomalous Hall effect in magnetic topological insulators. Science 329, 61–64 (2010).
6. Qiao, Z. H. et al. Quantum anomalous Hall effect in graphene from Rashba and exchange effects. Phys. Rev. B 82, 161414 (2010).
7. Nomura, K. & Nagaosa, N. Surface-quantized anomalous Hall current and the magnetoelectric effect in magnetically disordered topological insulators. Phys. Rev. Lett. 106, 166802 (2011).
8. Zhang, H., Lazo, C., Bluegel, S., Heinze, S. & Mokrousov, Y. Electrically tunable quantum anomalous Hall effect in graphene decorated by 5d transition-metal adatoms. Phys. Rev. Lett. 108, 056802 (2012).
9. Ezawa, M. Valley-polarized metals and quantum anomalous Hall effect in silicene. Phys. Rev. Lett. 109, 055502 (2012).
10. Chang, C.-Z. et al. Experimental observation of the quantum anomalous Hall effect in a magnetic topological insulator. *Science* **340**, 167–170 (2013).

11. Checkelsky, J. G. et al. Trajectory of the anomalous Hall effect towards the quantized state in a ferromagnetic topological insulator. *Nat. Phys.* **10**, 731–736 (2014).

12. Kou, X. et al. Scale-invariant quantum anomalous Hall effect in magnetic topological insulators beyond the two-dimensional limit. *Phys. Rev. Lett.* **113**, 137201 (2014).

13. Kou, X. et al. Metal-to-insulator switching in topological insulators. *Nat. Commun.* **6**, 8474 (2015).

14. Feng, Y. et al. Observation of the zero Hall plateau in a quantum anomalous Hall state. *Phys. Rev. Lett.* **107**, 182401 (2015).

15. Ou, Y. et al. Enhancing the quantum anomalous Hall effect by magnetic doping in topological insulators. *Adv. Mater.* **30**, 1703062 (2018).

16. Lachman, E. O. et al. Visualization of superparamagnetic dynamics in magnetic topological insulators. *Sci. Adv.* **1**, e1500740 (2015).

17. Grauer, S. et al. Coincidence of superparamagnetism and perfect quantization in the quantum anomalous Hall state. *Phys. Rev. B* **92**, 201304 (2015).

18. Lee, I. et al. Imaging Dirac-mass disorder from magnetic dopant atoms in the ferromagnetic topological insulator Cr,(Bi,SB)2−xTe3. *Proc. Natl Acad. Sci. USA* **112**, 1316–1321 (2015).

19. Bednorz, J. G. & Muller, K. A. Possible high Tc superconductivity in the Ba−La−Cu−O system. *Z. Phys. B* **64**, 189–193 (1986).

20. Wu, M. K. et al. Superconductivity at 93 K in a new mixed-phase Y−Ba−Cu−O compound system at ambient pressure. *Phys. Rev. Lett.* **58**, 908–910 (1987).

21. Maeda, H., Tanaka, Y., Fukutomii, M. & Asano, T. A new high-Tc oxide superconductor without a rare earth element. *Ipn J. Appl. Phys.* **27**, L209–L210 (1988).

22. Schilling, A., Cantoni, M., Guo, J. D. & Ott, H. R. Superconductivity above 130 K in the Hg−Ba−Ca−Cu−O system. *Nature* **363**, 56–58 (1993).

23. Chang, C.-Z. et al. High-precision realization of robust quantum anomalous Hall state in a hard ferromagnetic topological insulator. *Nat. Mater.* **14**, 473–477 (2015).

24. Grauer, S. et al. Scaling of the quantum anomalous Hall effect as an indicator of axion electrodynamics. *Phys. Rev. Lett.* **118**, 246801 (2017).

25. Chang, C.-Z. et al. Chemical-potential-dependent gap opening at the Dirac surface states of Bi2Se3 induced by aggregated substitutional Cr atoms. *Phys. Rev. Lett.* **112**, 056801 (2014).

26. Li, W. et al. Origin of the low critical observing temperature of the quantum anomalous Hall effect in V-doped (Bi,SB)2−xTe3 film. *Sci. Rep.* **6**, 32732 (2016).

27. Anderson, P. W. Absence of diffusion in certain random lattices. *Phys. Rev.* **109**, 1492–1505 (1958).

28. Andriotis, A. N. & Menon, M. Defect-induced magnetism: Codoping and a prescription for enhanced magnetism. *Phys. Rev. B* **87**, 155309 (2013).

29. Qi, S. F. et al. High-temperature quantum anomalous Hall effect in n−p codoped topological insulators. *Phys. Rev. Lett.* **117**, 056804 (2016).

30. Nagaosa, N., Sinova, J., Onoda, S., MacDonald, A. H. & Ong, N. P. Anomalous Hall effect. *Rev. Mod. Phys.* **82**, 1539–1592 (2010).

31. Ruderman, M. A. & Kittel, C. Indirect exchange coupling of nuclear magnetic moments by conduction electrons. *Phys. Rev.* **96**, 99–102 (1954).

32. Kou, X. F. et al. Interplay between different magnetisms in Cr-doped topological insulators. *ACS Nano* **7**, 9205–9212 (2013).

33. Wang, W. & Wu, W. Visualizing ferromagnetic domain behavior of magnetic topological insulator thin films. *npj Quant. Mater.* **1**, 16023 (2016).

34. Li, M. et al. Experimental verification of the Van Vleck nature of long-range ferromagnetic order in the vanadium-doped three-dimensional topological insulator SnTe. *Phys. Rev. Lett.* **114**, 146802 (2015).

35. Li, H. et al. Carriers dependence of the magnetic properties in magnetic topological insulator Sn1−xBi0.9−xTe3. *Appl. Phys. Lett.* **101**, 072406 (2012).

36. Checkelsky, J. G., Ye, J., Onose, Y., Iwasa, Y. & Tokura, Y. Dirac-fermion-mediated ferromagnetism in a topological insulator. *Nat. Phys.* **8**, 729–733 (2012).

37. Sesli, P. et al. Signatures of Dirac fermion-mediated magnetic order. *Nat. Commun.* **5**, 5349 (2014).

38. Chang, C.-Z. et al. Zero-field dissipationless chiral edge transport and the nature of dissipation in the quantum anomalous Hall state. *Phys. Rev. Lett.* **115**, 057206 (2015).

39. Wang, W. et al. Visualizing weak ferromagnetic domains in multiferroic hexagonal ferrite thin film. *Phys. Rev. B* **95**, 134443 (2017).

**Acknowledgements**

We thank C. Chang for helpful discussions and P. Sass for proofreading the manuscript. This work at Rutgers is supported by the Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, US Department of Energy under Award numbers DE-SC0008147 and DE-SC0018153. The work at Tsinghua University is supported by the National Natural Science Foundation of China and the Ministry of Science and Technology of China.

**Author contributions**

W.Wu, K.H. and Y.W. conceived the project. W.Wu and W.Wa designed the MFM experiments. W.Wa and H.D. performed the experiments. W.Wu and H.D. analysed the data. Y.O. synthesized the MBE films under the supervision of K.H. and W.Wa, and carried out the transport measurements. W.Wu and K.H. wrote the manuscript with inputs from all authors.

**Competition interests**

The authors declare no competing interests.

**Additional information**

Supplementary information is available for this paper at https://doi.org/10.1038/s41567-018-0149-1.

Reprints and permissions information is available at www.nature.com/reprints.

Correspondence and requests for materials should be addressed to W.W.
Methods

Sample preparation. Epitaxial thin film Cr/V co-doped BST films capped with 2 nm Al were grown on a heat-treated SrTiO$_3$(111) (STO) substrate by co-evaporation in a MBE system. The nominal thickness of the film is 5 QLs. The film was scratched by hand into a Hall bar shape connected with a large square-like area for MFM measurements. An approximately 15 nm Au film layer was deposited on the square area to eliminate electrostatic interactions between the sample and the magnetic tip.

MFM measurement and in situ transport measurement. The MFM experiments were carried out in an in-house-built cryogenic atomic force microscope (AFM) using commercial piezo-resistive cantilevers (spring constant $k \approx 3 \text{N m}^{-1}$, resonant frequency $f_0 \approx 42 \text{kHz}$). This AFM is interfaced with a Nanonis SPM Controller (SPECS) and a commercial phase-lock loop (SPECS) $^{34,41}$. The tips for the MFM were prepared by depositing a nominally 100 nm Co film onto bare tips using $e$-beam evaporation. MFM images were taken in a constant-height mode with the scanning plane ~40 nm above the sample surface. To avoid the relaxation effect (domain wall creeping) near $H_c$ and to minimize the stray field effect of the MFM tips, all MFM images were taken at a low magnetic field of approximately 0.05 T after the magnetic field was ramped to the desired values. The MFM signal, the change of cantilever resonant frequency, is proportional to the out-of-plane stray field gradient$^{42}$. Electrostatic interaction was minimized by nulling the tip–surface contact potential difference. Blue (red) regions in the MFM images represent up (down) ferromagnetic domains, where magnetizations are parallel (anti-parallel) with respect to the positive external field.

The magnetic topological insulator films were fabricated into Hall bar devices. The Hall resistance and longitudinal resistance were measured by standard lock-in techniques with an alternating current of 5 $\mu$A modulated at 314 Hz.

Data availability. The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

References
41. Wang, W. et al. Visualizing ferromagnetic domains in magnetic topological insulators. APL Mater. 3, 083301 (2015).
42. Rugar, D. et al. Magnetic force microscopy: General principles and application to longitudinal recording media. J. Appl. Phys. 68, 1169–1183 (1990).