Spin mapping of intralayer antiferromagnetism and spin–flop transition in monolayer CrTe₂

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Abstract: Intrinsic antiferromagnetism in van der Waals (vdW) monolayer (ML) crystals enriches the understanding regarding two-dimensional (2D) magnetic orders and holds special virtues over ferromagnetism in spintronic applications. However, the studies on intrinsic antiferromagnetism are sparse, owing to the
lack of net magnetisation. In this study, by combining spin-polarised scanning tunnelling microscopy and first-principles calculations, we investigate the magnetism of vdW ML CrTe₂, which has been successfully grown through molecular beam epitaxy. Surprisingly, we observe a stable antiferromagnetic (AFM) order at the atomic scale in the ML crystal, whose bulk is a strong ferromagnet, and correlate its imaged zigzag spin texture with the atomic lattice structure. The AFM order exhibits an intriguing noncollinear spin–flop transition under magnetic fields, consistent with its calculated moderate magnetic anisotropy. The findings of this study demonstrate the intricacy of 2D vdW magnetic materials and pave the way for their in-depth studies.

MAIN TEXT

The exploration of the long-range magnetic order down to the single-layer limit is not only of fundamental importance for examining its existence at finite temperatures but also driven by the technological requirements of miniaturised device circuits. According to the Mermin–Wagner theorem [1], the long-range magnetic order cannot exist in one and two dimensions at finite temperatures owing to the low energy cost of fluctuations, which benefit the total energy of the system via entropy increase. However, in the presence of magnetic anisotropy, the fluctuation effect can be effectively suppressed, and the long-range magnetic order revives. The recent discovery of intrinsic ferromagnetism at ultrathin van der Waals (vdW) films of CrI₃ [2] and Cr₂Ge₂Te₆ [3] have spurred considerable interest in seeking magnetism in single layers and their manipulations [4-9].
Antiferromagnetic (AFM) orders have gained increasing attention in spintronic applications owing to their advantages over their ferromagnetic counterpart in achieving ultrafast dynamics, large magnetoresistance transport, and immunity to magnetic field disturbance associated with absent stray fields [10-14]. However, despite the fruitful research on two-dimensional (2D) ferromagnetism, AFM orders at the 2D limit are significantly less explored because of the vanishing net magnetisation, causing difficulty in detection. Pioneering studies on Raman spectroscopy have identified AFM orders in a vdW monolayer (ML) FePS₃ via spin–phonon interactions [15,16]. As AFM orders possess various possible configurations, their specific spin textures are urged to be determined with real-space spin-sensitive probes. Spin-polarised scanning tunnelling microscopy (SPSTM), performed by recording spin-dependent electron tunnelling currents, can directly identify the magnetic order with atomic-scale spin resolution [17], enabling considerable progress in the studies on magnetism in vdW materials [18-21]. Particularly, a double-stripe AFM spin structure, coexisting with superconductivity, was observed on a Fe₁₊yTe/Bi₂Te₃ heterojunction via SPSTM [21]. However, such superconductivity is believed to be incurred from prominent interfacial interactions, thus casting doubt on whether the AFM order in Fe₁₊yTe is intrinsically stable.

Here, we report the successful growth of CrTe₂ films, a layered vdW crystal, on a SiC-supported bilayer graphene substrate and achieve a spin-resolved imaging of the ML CrTe₂ with SPSTM. Although CrTe₂ has strong itinerant ferromagnetism in bulk [22], its ML surprisingly reveals an AFM spin texture at the atomic scale. The AFM
order is of zigzag type with the magnetic easy axis along the Cr–Te bonding direction. With a magnetic field applied perpendicular to the basal plane, the ML CrTe₂ exhibits a spin–flop-type magnetic transition, which, to our knowledge, is the first report of such a real-space observation. Our work paves the way for in-depth studies on the fundamental physics of 2D antiferromagnetism and sets a foundation for its application in AFM spintronics.

Because of the multivalent nature of the Cr cation, chromium tellurides have multiple polymorphs [23], posing a challenge to grow single-phase compounds. With the judicious tuning of the flux ratio of Cr and Te and the substrate temperature, we successfully grew uniform CrTe₂ films. CrTe₂ has a trigonal layered structure belonging to space group P3m1 [24]. Each vdW layer of CrTe₂ constitutes chromium cations centring at tellurium octahedrons, forming a 1T structure (Fig. 1(a)). Fig. 1(b) shows a typical scanning tunnelling microscopy (STM) topographic image of the as-grown thin films, which are dominated with an ML and a second layer, with a fraction of a third layer. The second-layer film manifests an apparent height of 0.63 ± 0.16 nm, which is insensitive to the bias and is consistent with the theoretical ML height (0.6168 nm). However, the apparent height of the first-layer film varies between 1.05 nm and 0.88 nm at different biases (Supplementary Fig. S1), reflecting a large vdW gap at the interface and the distinct electronic structure between the graphene and CrTe₂.

To disentangle the electronic effect on the structural determination, the CrTe₂ film is characterised with cross-sectional scanning transmission electron microscopy
A high-angle annular dark-field (HAADF) STEM image (Fig. 1(c)) confirms the 1T structure of the bilayer CrTe$_2$ with Te capping. Owing to its heavier mass, Te has brighter contrast than Cr does. The associated integrated differential phase contrast (iDPC) image (Fig. 1(d)) resolves the fine details of the film, clearly showing the Cr cations, interlayer vdW gaps, and supporting graphene/SiC substrate. The heights of the second- and first-layer CrTe$_2$ are estimated as 0.6 nm and 0.8 nm, respectively, conforming to the STM measurement. The elemental composition of the film is 1:2 for the Cr:Te ratio according to the standard-based quantification analysis of the electron energy loss spectrum (EELS) line scan across the interface (Fig. 1(e)). Moreover, the energy-dispersive spectroscopy (EDS) mapping (Supplementary Fig. S2) further verifies the Cr and Te elemental distributions in each layer.

An atomic-resolution STM image of the ML film (Fig. 1(f)) and its associated fast Fourier transform (FFT) image (Fig. 1(g)) reveal an isosceles lattice with in-plane constants measured as $a_1 = 3.2$ Å, $a_2 = 3.6$ Å, and $a_3 = 3.6$ Å, which obviously deviates from an expected regular one of the bulk surfaces. The isosceles lattice is further confirmed by the STM imaging of a domain boundary, which expresses a mirror symmetry between the two neighbouring domains (Supplementary Fig. S3). An evident 2×1 periodicity is exhibited in the atomic-resolution image and the FFT image of the ML CrTe$_2$, which is closely related to its AFM spin texture, as elucidated later. Six additional Bragg peaks are present around the central zone (Fig. 1(g), yellow rectangle), which stem from a 6×6 Moiré pattern at the graphene/SiC(0001) interface. On the second-layer film, the 2×1 structure exhibits mixing domains of three
equivalent orientations (Fig. 1(h)).

For bulk CrTe$_2$ crystals, a ferromagnetic ground state with a Curie temperature of 310 K was previously identified [22], rendering it as a promising candidate for exploring high-Curie-temperature ML ferromagnets. A recent study found that the Curie temperature of a ~10-ML CrTe$_2$ exhibits ferromagnetism at room temperature [24]. However, theoretical studies predict that the magnetism of vdw MLs varies under a few external perturbations, including strain [25-27] and doping or interlayer coupling [28,29], leaving their magnetic orders as an open issue.

SPSTM measurement is applied to the ML CrTe$_2$ with a Cr-coated W tip, where the tip magnetisation is cant. Because of its small stray field, the AFM Cr tip barely perturbs the magnetisation of the sample and is resistant against the magnetic field. An SPSTM image at −0.1 V (Fig. 2(a)) shows a clear zigzag-like pattern that is distinct from the 2×1 structure shown in Fig. 1(f). Its magnetic unit cell (Fig. 2(a), white rectangle) is twice the original lattice in both directions. As is seen from its FFT image (Fig. 2(b)), the diffraction spots (white circles) of the zigzag pattern, in comparison with those in Fig. 1(g), are coexisting with those of the 2×1 structure (yellow circles) and are orthogonal to one another. The orthogonality is substantiated from the image of the same area at −60 mV (Supplementary Fig. S4), where the 2×1 structure becomes more visible. Because the zigzag pattern is only resolvable with the magnetic tip, it is ascribed as a spin contrast.

Such ascription is further confirmed by applying magnetic fields perpendicular to the basal plane of the films, i.e., z direction. Fig. 2(c and d) depicts two SPSTM
images of the same area taken at 1 T and −1 T, respectively. There is a defect acting as a marker (green circle). Intriguingly, the zigzag pattern concertedly reverses its contrast relative to the defect under the opposite field, thereby unambiguously proving its origin from the spin contrast. The spin contrast becomes more enhanced in its difference image (Fig. 2(e)) between Figs. 2(c) and (d) and vanishes in their sum image (Fig. 2(f)), resembling the spin-averaged image of Fig. 1(f). Such a spin-contrast reversal can also be clearly seen from their line profiles (Fig. 2(g)) extracted along the same locations of Figs. 2(c–f). The spin contrast keeps its registration with the defect marker in either field orientations up to 5 T (Supplementary Fig. S5) and memorises the history of the field orientation upon its removal (Supplementary Fig. S6). The threshold field is small (<0.2 T) to induce the spin-contrast reversal and is also highly reproducible measured with different Cr tips. Hence, the spin-contrast reversal is from the CrTe₂ because the magnetisation of Cr tips conventionally persists more than 2 T and varies for different tips. Moreover, the zigzag spin contrast of different domains of ML CrTe₂ is not identical (Supplementary Fig. S7), suggesting that the tip magnetisation is canted with finite in-plane spin sensitivity, as is also substantiated from the spin mapping with an Fe-coated tip (Supplementary Fig. S8).

We perform density functional theory (DFT) calculations to unveil the origin of the spin contrast and its field-induced switching. A striped AABB-type AFM state is more favoured among all considered magnetic configurations (Supplementary Fig. S9) in the fully relaxed freestanding ML CrTe₂ (Supplementary Table S1). This phase
reduces the three-fold structural symmetry and leads to a 0.1 Å shrink for the lattice constant along the \( y \) direction, i.e., from 3.70 Å to 3.60 Å (Supplementary Table S1), which is rather smaller than the experimentally observed 0.4 Å shrink. Many studies suggest that the substrate could apply an appreciably strong in-plane strain to the 2D layers and modify their in-plane magnetic order [21, 30-32]. Here, we adopt a \( 4 \times 2\sqrt{3} \) CrTe\(_2\) / \( 5 \times 3\sqrt{3} \) bilayer graphene with a rectangular supercell (Supplementary Fig. S10) to model the substrate effect. The superlattice adopts the lattice constant of the intact graphene, which gives rise to a lattice constant of 3.2/3.5 Å for CrTe\(_2\), which is well consistent with the experimental values.

In this superlattice, a zigzag order (Fig. 3(a)), 35.1 meV/Cr more stable than the striped AFM order, becomes the most favoured magnetic configuration. The stabilisation of the zigzag order may originate from either the interfacial charge transfer from the substrate or the in-plane strain applied by the substrate. Our calculation indicates that the in-plane strain plays a significant role in the stability switching. A free-standing ML CrTe\(_2\) compressively constrained in the superlattice constants favours the zigzag order (Supplementary Table S1), whereas charge doping up to 0.2 \( e \) or \( h \) per Te to the intact freestanding ML does not change the order of stability between two magnetic configurations (Supplementary Fig. S11). We thus focus on the zigzag order in the following discussion.

Spin densities are also mapped on the atomic structure of the ML CrTe\(_2\) (Fig. 3(a and b)). Each Te atom is spin-polarised by its adjacent Cr atoms. Consequently, its three \( p \) orbitals are categorised into two groups, namely, \( p_{x/y} \) and \( p_z \) orbitals, with
opposite spin polarisations (in green and red, respectively), consistent with its small net magnetic moment of \(-0.06 \, \mu_B\) (Supplementary Table S1). The \(p_z\) orbital has a dumbbell-like shape, pointing along a Cr–Te bond. Moreover, the \(p_{x,y}\) orbital appears with a bagel-like shape, with its basal plane orthogonal to the \(p_z\) dumbbell axis. The simulated SPSTM images based on the zigzag spin configuration (Fig. 3(b)) are well consistent with those of the experimental observation (Fig. 2(a), inset). Fig. 3(c) shows a zoomed-in view of the circled region in Fig. 3(a). Here, in addition to the \(x, y,\) and \(z\) axes, we define three additional directions, i.e., \(O_0, O_{i1},\) and \(O_{i2},\) for the magnetic anisotropy energy (MAE) calculations. Our calculations indicate that the \(O_0\) direction is the easy axis for magnetisation (Supplementary Table S2), consistent with the experimentally observed finite out-of-plane magnetic anisotropy. Magnetisation along the \(y\) direction is the second stable direction with a small MAE of 0.29 meV/Cr. However, 4.08 meV/Cr is needed to rotate the magnetisation direction to \(z,\) making it less achievable under a small magnetic field.

We proposed a noncollinear spin–flop model to explain the field-induced spin-contrast reversal, which usually occurs in antiferromagnets with small magnetic anisotropy. The magnetic moments of the Cr cation in CrTe\(_2\) are, as schematically depicted Fig. 3(d), in a collinear AFM state under the zero field. With a magnetic field applied along the \(+z\) direction (Fig. 3(e)), the magnetic moments pointing in the upper-left direction (marked with red arrows) slightly rotate clockwise approaching to the \(+z\) axis. However, the moments oriented along the lower-right direction (green arrows) significantly lie down to the \(y\) axis, ascribed to the rather small MAE of 0.29
meV/Cr between the easy and $y$ axes. If the field direction flips, then the tilting direction of each moment reverses accordingly, e.g., approaching the $-z$ axis for the green moments and nearly lying down for the red moments (Fig. 3(f)). Such a field-induced noncollinear rotation of local magnetic moments causes a spin-contrast reversal of the SPSTM images acquired under opposite magnetic fields as the tip magnetisation is resistant to the field.

As shown in Fig. 4(a), the density of states (DOS) of the ML CrTe$_2$ with the magnetic moments pointing to the $O_0$ direction (easy axis) appreciably differs from that to the $y$ direction (secondary easy axis), especially for the states residing from $-0.35$ to $0.25$ eV. A $\sim 10$-meV upward shift is identified if the moments collinearly rotate from the easy to the secondary easy axes, which is, in principle, detectable by scanning tunnelling spectroscopy (STS). However, as illustrated in Fig. 3(c–e), the moments rotate in a noncollinear manner, which causes a mixing between the two spin channels and/or among different orbitals [30,31]. This noncollinear picture suggests that the major portion of orbitals that contribute to the tunnelling current, most likely, switches between the Te $p_z$ and $p_{x/y}$ orbitals under the $+z$ and $-z$ magnetic fields. We thus examine the DOSs projected on the $p_z$ and $p_{x/y}$ orbitals, as shown in Fig. 4(b), to examine if they show apparent differences. A pronounced peak (P$_1$) sits at $-0.29$ eV, which is dominated by the $p_z$ orbital. Two additional peaks (P$_2$ and P$_3$) reside at $-0.44$ and $-0.49$ eV, respectively, the major compositions of which are $p_{x/y}$ and $p_z$ orbitals. The magnetic field-induced rotation of the Cr moment described earlier expects the variation of the $p_{x/y}$ and $p_z$ orbitals and their associated peaks in the
contribution of the tunnelling conductance. Hence, a field-induced shift of \(~150\) meV, corresponding to the energy spacing between \(P_1\) and \(P_2\), is expected to be detectable in the spin-resolved spectra. Visualised wavefunction norms of the state \(P_1\), depicted in Fig. 4(c and d), show largely reduced \(p_z\) orbitals and lightly lying-down \(\text{Te-}p_{x/y}\) orbital if the moment rotates from the \(O_o\) to \(y\) directions. This finding further suggests the feasibility of the spin–flop picture that leads to different SP-STS spectra.

We examine such an effect by taking spectra with the same micro Cr tip as that in Fig. 2 under various magnetic fields. The spin-resolved \(dI/dV\) spectra (Fig. 4(e)) feature two characteristic peaks at approximately \(-0.5\) and \(0.88\) V. Although the peak at \(0.88\) V stays unchanged, the peak at \(-0.5\) V shifts progressively toward lower energy with increasing field along the \(z\) orientations, by up to \(~120\) meV at \(5\) T. This value is similar with that of the calculated results. The field-induced peak shift is reproducible with a different Cr tip (Supplementary Fig. S12). Furthermore, the energy of the different \(\text{Te-}p\) orbitals expects a spatial variation that correlates with the zigzag spin pattern, as is also unequivocally observed in the spin-resolved spectra (Supplementary Fig. S13). Therefore, such a spectral peak shift indeed supports the picture of the magnetic field-induced spin-mixing and reorientation of \(\text{Te-}p\) orbitals.

In conclusion, we observed a stable AFM order in ML CrTe₂ with SPSTM, whose spin contrast is from magnetically proximitized \(\text{Te-}p\) orbitals with an easy axis pointing in the Cr–Te bonding direction. A spin–flop-type magnetic transition occurs under magnetic fields, driving the ML CrTe₂ into a noncollinear spin texture. The unambiguous identification of the ML antiferromagnetism and the unprecedentedly
observed atomic-scale spin–flop transition add a new dimension for understanding the intralayer magnetic order at the 2D limit. The ML CrTe₂ not only provides a system for in-depth studies on fundamental physics in 2D antiferromagnetism, such as noncollinear spin-related magneto-transport phenomena [32], spin excitation [33], and layer-dependent magnetic order transition, but also envisions to facilitate the development of AFM spintronics in miniaturised device applications.

METHODS

**Molecular-beam epitaxy (MBE) growth.** The graphene substrate was obtained via graphitisation of the SiC(0001) substrate with cycles of vacuum flashing treatment, the details of which are provided in [34]. The graphene obtained with such a treatment is dominated with bilayers. The 1T-CrTe₂ films were grown on the graphene-covered SiC(0001) substrate via co-evaporation of high-purity Cr (purity, 99.995%) and Te (purity, 99.999%) atoms with a flux of ~1:30 according to MBE. The substrate was kept at 573±10 K to facilitate the chemical formation of 1T-CrTe₂, where the substrate temperature was monitored with an infrared spectrometer with an emissivity of 0.9. We found that the substrate temperature is crucial for growing single-phase CrTe₂ films. For ex-situ STEM characterisation, the CrTe₂ films were protected against degradation with Te capping layers of ~200 nm thickness.

**STM measurements.** The measurements were performed in a custom-made Unisoku STM system [35] mainly at 4.2 K unless described exclusively. The spin-averaged STM data were measured with an electrochemically etched W wire, which had been characterised on a Ag(111) surface prior to the measurements. The SPSTM data were
taken with Cr or Fe tips. The Cr tip was prepared by coating ~50 layers of Cr (purity: 99.995%) on a W tip, which had been flashed to ~2000 K to remove oxides, followed by annealing at ~500 K for 10 min. The Fe tip was prepared by coating ~30 layers of Fe on a W tip, following similar flash and annealing procedures as the Cr tip. The tunnelling spectra were obtained through a lock-in detection of the tunnelling current with a modulation voltage of 983 Hz. The topographic images were processed by WSxM.

**STEM imaging.** The cross-section STEM specimens were prepared using a routine focused ion beam. STEM imaging, EDS and EELS analysis on the MBE-grown CrTe₂ were performed on a FEI Titan Themis with an extreme field emission gun and a DCOR aberration corrector operating at 300 kV. The inner and outer collection angles for the STEM images (β₁ and β₂) were 48 and 200 mrad, respectively. The convergence semi-angle of the probe was 25 mrad. The beam current was approximately 100 pA for the HAADF imaging, EDS and EELS chemical analyses. All imaging procedures were performed at room temperature.

**DFT calculations.** The calculations were performed using the generalised gradient approximation and projector augmented-wave method [36] as implemented in the Vienna *ab-initio* simulation package [37]. A uniform Monkhorst–Pack $k$ mesh of $15 \times 15 \times 1$ was adopted for integration over the Brillouin zone. An orthorhombic $2 \times 2 \sqrt{3}$ supercell was used to show the zigzag AFM order in the ML CrTe₂, with a $k$ mesh of $10 \times 6 \times 1$. A kinetic energy cutoff of 700 eV for the plane-wave basis set was used for the structural relaxation and electronic structure calculations. A sufficiently
large vacuum layer over 20 Å along the out-of-plane direction was adopted to eliminate the interaction among layers. Dispersion correction was performed at the vdW-DF level [38], with the optB86b functional for the exchange potential [39], which was proven to be accurate in describing the structural properties of layered materials [40-42] and was adopted for structural-related calculations in this study. All atoms were allowed to relax until the residual force per atom was less than 0.01 eV/Å.

To compare energy among different magnetic configurations, we used the PBE functional [43] with consideration of the spin–orbit coupling, based on the vdW-DF-optimised atomic structures. The on-site Coulomb interaction to the Cr $d$ orbitals had $U$ and $J$ values of 4.4 eV and 0.6 eV, respectively, as revealed by a linear response method [44]. These values are comparable to those adopted in modelling CrI$_3$ [45,46], CrS$_2$ [28], and CrSe$_2$ [29].

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**REFERENCES**

1. Mermin, N.D., and Wagner, H. Absence of ferromagnetism or antiferromagnetism in one- or two-dimensional isotropic Heisenberg models. *Phys. Rev. Lett.* **17**, 1133 (1966).

2. Huang, B., Clark, G., Navarro-Moratalla, E. *et al*. Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit. *Nature* **546**, 270-273 (2017).

3. Gong, C., Li L. *et al*. Discovery of intrinsic ferromagnetism in two-dimensional van der Waals crystals. *Nature* **546**, 265-269(2017).

4. Gong, C. & Zhang, X. Two-dimensional magnetic crystals and emergent heterostructure devices. *Science* **363**, 706 (2019).
5. Burch, K. S., Mandrus, D. & Park, J.-G. Magnetism in two-dimensional van der Waals materials. *Nature* **563**, 47–52 (2018).

6. Sethulakshmi, N. *et al.* Magnetism in two-dimensional materials beyond graphene. *Mater. Today* **27**, 107–122 (2019).

7. Deng, Y. J. *et al.* Gate-tunable room-temperature ferromagnetism in two-dimensional Fe₃GeTe₂. *Nature* **563**, 94–99 (2018).

8. Bonilla, M. *et al.* Strong room-temperature ferromagnetism in VSe₂ monolayers on van der Waals substrates. *Nat. Nanotechnol.* **13**, 289–293 (2018).

9. O’hara, D. J. *et al.* Room temperature intrinsic ferromagnetism in epitaxial manganese selenide films in the monolayer limit. *Nano Lett.* **18**, 3125–3131 (2018).

10. Baltz V., Manchon A., Tsoi M. *et al.* Antiferromagnetic spintronics. *Rev. Mod. Phys.* **90**, 015005(2018).

11. Jungwirth T., Marti X., Wadley P. *et al.* Antiferromagnetic spintronics. *Nat. Nanotechnol.* **11**, 231-241(2016).

12. Cheng R., Daniels, M.W. *et al.* Ultrafast switching of antiferromagnets via spin-transfer torque. *Phys. Rev. B* **91**, 064423(2015).

13. Wienholdt S., Hinzke D., Nowak U. THz Switching of Antiferromagnets and Ferrimagnets. *Phys. Rev. Lett.* **108**, 247207(2012).
14. Núñez A. S. *et al.* Theory of spin torques and giant magnetoresistance in antiferromagnetic metals. *Phys. Rev. B* **73**, 214426 (2006).

15. Wang X., Xing Zhi, Ke Zhao D. *et al.* Raman spectroscopy of atomically thin two-dimensional magnetic iron phosphorus trisulfide (FePS3) crystals. *2D Materials* **3**, 31009 (2016).

16. Lee, J.U. *et al.* Ising-Type Magnetic Ordering in Atomically Thin FePS3. *Nano Lett.* **16**, 7433 (2016).

17. Wiesendanger, R. Spin mapping at the nanoscale and atomic scale. *Rev. Mod. Phys.* **81**, 1495 (2009).

18. Enayat, M. *et al.* Real-space imaging of the atomic-scale magnetic structure of Fe1+yTe. *Science* **345**, 653–656 (2014).

19. Chen, W. *et al.* Direct observation of van der Waals stacking–dependent interlayer magnetism. *Science* **366**, 983–987 (2019).

20. Hänke, T. *et al.* Reorientation of the diagonal double-stripe spin structure at Fe1+yTe bulk and thin-film surfaces. *Nat. Commun.* **8**, 13939 (2017).

21. Manna, S., Kamlapure, A., Cornils, L. *et al.* Interfacial superconductivity in a bi-collinear antiferromagnetically ordered FeTe monolayer on a topological insulator. *Nat Commun.* **8**, 14074 (2017).

22. Freitas, Daniele C., Ruben W. *et al.* Ferromagnetism in layered metastable 1T-CrTe2. *Journal of Physics: Condens. Matter* **27**, 176002 (2015).
23. Ipser, H., Komarek, K.L., Klepp, K.O. Transition metal-chalcogen systems viii: The Cr-Te phase diagram. *Journal of the Less Common Metals* **92**, 265-282 (1983).

24. Sun X., Li W., Wang X. *et al.* Room temperature 2D ferromagnetism in few-layered 1T-CrTe$_2$. arXiv:190909797 (2019).

25. Ma, Y. *et al.* Evidence of the Existence of Magnetism in Pristine VX$_2$ Monolayers (X = S, Se) and Their Strain-Induced Tunable Magnetic Properties. *ACS Nano* **6**, 1695–1701 (2012).

26. Lv H., Lu Wen Qin, Shao Ding F. *et al.* Strain-controlled switch between ferromagnetism and antiferromagnetism in 1T-CrX$_2$ (X=Se,Te) monolayers. *Phys. Rev. B* **92**, 214419 (2015).

27. Sivadas, N. *et al.* Magnetic ground state of semiconducting transition-metal trichalcogenide monolayers. *Phys. Rev. B* **91**, 235425 (2015).

28. Wang C., Zhou X., Pan Y. *et al.* Layer and doping tunable ferromagnetic order in two-dimensional CrS$_2$ layers. *Phys. Rev. B* **97**, 245409 (2018).

29. Wang, C. *et al.* Bethe-Slater-curve-like behavior and interlayer spin-exchange coupling mechanisms in two-dimensional magnetic bilayers. *Phys. Rev. B* **102**, 020402 (2020).

30. Bode, M. *et al.* Magnetization-direction dependent local electronic structure probed by scanning tunneling spectroscopy. *Phys. Rev. Lett.* **89**, 237205 (2002).
31. Perini, M. et al. Electrical Detection of Domain Walls and Skyrmions in Co Films Using Noncollinear Magnetoresistance. Phys. Rev. Lett. **123**, 237205 (2019).

32. Chen, H., Niu, Q. and MacDonald, A. H. Anomalous Hall Effect Arising from Noncollinear Antiferromagnetism. Phys. Rev. Lett. **112**, 017205 (2014).

33. Kang, S. et al. Coherent many-body exciton in van der Waals antiferromagnet NiPS$_3$. Nature **583**, 785–789(2020)

34. Yang X. et al. Possible Phason-ploarons in purely one-dimensional charge order of Mo$_6$Se$_6$ nanowires. Phys. Rev. X **10**, 031061 (2020).

35. Peng, L. et al. Observation of topological states residing at step edges of WTe$_2$. Nat. Commun. **8**, 659 (2017).

36. Kresse, G. & Joubert, D. From ultrasoft pseudopotentials to the projector augmented-wave method. Phys. Rev. B **59**, 1758-1775 (1999).

37. Kresse, G. & Furthmüller, J. Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set. Comput. Mater. Sci. **6**, 15-50 (1996).

38. Dion, M., Rydberg, H., Schröder, E., Langreth, D. C. & Lundqvist, B. I. Van der Waals Density Functional for General Geometries. Phys. Rev. Lett. **92**, 246401 (2004).

39. Klimeš, J., Bowler, D. R. & Michaelides, A. Van der Waals density functionals applied to solids. Phys. Rev. B **83**, 195131 (2011).
40. Hong, J. et al. Exploring atomic defects in molybdenum disulphide monolayers. *Nat. Commun.* **6**, 6293 (2015).

41. Qiao, J. et al. Few-layer Tellurium: one-dimensional-like layered elementary semiconductor with striking physical properties. *Sci. Bull.* **63**, 159-168 (2018).

42. Zhao, Y. et al. High-Electron-Mobility and Air-Stable 2D Layered PtSe$_2$ FETs. *Adv. Mater.* **29**, 1604230 (2017).

43. Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized Gradient Approximation Made Simple. *Phys. Rev. Lett.* **77**, 3865-3868 (1996).

44. Cococcioni, M. & de Gironcoli, S. Linear response approach to the calculation of the effective interaction parameters in the LDA+U method. *Phys. Rev. B* **71**, 035105 (2005).

45. Xu, C., Feng, J., Xiang, H. & Bellaiche, L. Interplay between Kitaev interaction and single ion anisotropy in ferromagnetic CrI$_3$ and CrGeTe$_3$ monolayers. *NPJ Comput. Mater.* **4**, 57 (2018).

46. Jiang, P. et al. Stacking tunable interlayer magnetism in bilayer CrI$_3$. *Phys. Rev. B* **99**, 144401 (2019).
**FIG. 1 Morphology of 1T-CrTe$_2$ films.** (a) Top view (up) and side view (down) of the crystal structure of 1T-CrTe$_2$. The orange (brown) balls represent the top (bottom) Te atoms, and the dodger-blue balls represent the Cr atoms. (b) Pseudo-3D topographic STM image ($V_b = 2$ V, $I_t = 10$ pA) of CrTe$_2$ films. The thicknesses of the films are marked. (c,d) Atomic resolution of the HAADF-STEM (c) and iDPC images (d) of the side view of 2 ML CrTe$_2$. (e) EELS quantification analysis along the red line in (c), demonstrating that the chemical stoichiometry of Cr and Te is 1:2 at the
green-shaded region. (f,g) Atomic resolution of the STM image ($V_b = 0.1 \text{ V}, I_t = 85 \text{ pA}$) of 1 ML CrTe$_2$ (f) and its FFT image (g). Inset of (f) shows a zoomed-in view of the ML CrTe$_2$. The periodicity of the $2\times1$ structure is marked. The yellow circles in (g) mark the $2\times1$ superstructure of the Te lattice. (h) Atomic resolution of the STM image ($V_b = -25 \text{ mV}, I_t = 10 \text{ pA}$) of 2 ML CrTe$_2$.

FIG. 2 Spin mapping of the zigzag AFM spin texture in 1 ML 1T-CrTe$_2$. (a, c, and d) SPSTM images ($V_b = -0.1 \text{ V}, I_t = 100 \text{ pA}$) of 1 ML CrTe$_2$ taken with a Cr tip under
different magnetic fields. Inset of (a) is a DFT-simulated SPSTM image of the ML CrTe$_2$ in the zigzag-type AFM order. (b) FFT image of (a). (e) Difference image obtained by subtracting (d) to (c). (f) Sum image of (d) and (c). (g) Line profiles extracted along the white lines in (c–f). The green lines in (a, c–e) mark the direction of the zigzag spin rows.

FIG. 3 Calculated AFM order in 1ML CrTe$_2$. (a) Perspective view of the CrTe$_2$ ML in the zigzag order. Green and red arrows represent the magnetisation directions of Cr atoms. Spin density contours of the selected Te atoms are plotted with an isosurface value of 0.001 e/Bohr$^3$, where the red (green) contours denote the spin-up (spin-down).
(b) Top view of the CrTe$_2$ ML in the zigzag order, superimposed on the DFT-simulated SPSTM image (top) and the spin density contours of the Te atoms (bottom). (c) Magnetisation axes considered in the MAE calculation. $x$, $y$ and $z$ axes correspond to the directions of the lattice vectors. A Cr-Te plane is marked with a partially transparent orange rectangular. $O_0$ and $O_{i1}$ axes represent the two types of Cr-Te bonding directions. $O_{i2}$ axis is along the mirror direction between two $O_{i1}$ Cr–Te bonds. (d–f) Schematics of the SPSTM measurement configurations under different magnetic field directions. The ML CrTe$_2$ is viewed along the black arrow in (a). The tip magnetisation direction is represented with blue arrows.

**FIG. 4 Electronic characteristics coupled with magnetism.** (a) DOS of the ML CrTe$_2$ where the magnetic moment of each Cr cation is oriented along the $O_0$ (the easy axis) and $y$ (secondary easy axis) directions. (b) DOSs projected on the Te $p_z$ and $p_{xy}$ orbitals in the ML CrTe$_2$ with Cr magnetic moments along the $O_0$ axis. Characteristic
peaks P₁, P₂ and P₃ are marked. (c,d) Wavefunction norms of the P₁ peak with the Cr magnetic moments along the O₂ (c) and y (d) directions. The isosurface value of the contours is 0.0002 e/Bohr³. Red and green lines indicate the positions of Te cations and the upper boundary of the contours, respectively. (e) Tunnelling spectra (Vₕ = −1 V, Iᵣ = 100 pA, V_mod = 14.14 mV rms) of the same location indicated in the green cross of Fig. 2(d) obtained with the same micro Cr tip at various magnetic fields. The black curves are fitted to the peaks around −0.5 eV with a Lorentz shape and a polynomial background. (f) Statistics of the fitted peak energies of (e) against the magnetic field, where the error bars are from the fitting.