Room Temperature Ferromagnetism of Monolayer Chromium Telluride with Perpendicular Magnetic Anisotropy

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The realization of long-range magnetic ordering in 2D systems can potentially revolutionize next-generation information technology. Here, the successful fabrication of crystalline Cr$_3$Te$_4$ monolayers with room temperature (RT) ferromagnetism is reported. Using molecular beam epitaxy, the growth of 2D Cr$_3$Te$_4$ films with monolayer thickness is demonstrated at low substrate temperatures (≈100 °C), compatible with Si complementary metal oxide semiconductor technology. X-ray magnetic circular dichroism measurements reveal a Curie temperature ($T_C$) of $\approx$344 K for the Cr$_3$Te$_4$ monolayer with an out-of-plane magnetic easy axis, which decreases to $\approx$240 K for the thicker film ($\approx$7 nm) with an in-plane easy axis. The enhancement of ferromagnetic coupling and the magnetic anisotropy transition is ascribed to interfacial effects, in particular the orbital overlap at the monolayer Cr$_3$Te$_4$/graphite interface, supported by density-functional theory calculations. This work sheds light on the low-temperature scalable growth of 2D nonlayered materials with RT ferromagnetism for new magnetic and spintronic devices.

1. Introduction

The family of 2D crystals has been expanding rapidly over the past decade, exhibiting a wide variety of novel electronic and optical properties, and more recently magnetism.[1–3] This is because long-range magnetism is not predicted to manifest in 2D systems due to the enhanced thermal fluctuations, as suggested by the Mermin–Wagner theorem.[4] In 2016, pioneer works on FePS$_3$ have revealed the persistence of antiferromagnetic ordering down to the monolayer limit, with the Néel temperature around 100 K.[5,6] In 2017, breakthroughs in 2D ferromagnetism were achieved in atomically thin layers of CrI$_3$[7] and Cr$_2$Ge$_2$Te$_6$[8] (with the Curie temperature $T_C$ below 100 K), showing that magnetic anisotropy can stabilize the long-range magnetic ordering with an excitation gap opening. Subsequently, other 2D materials with exotic magnetic properties, e.g., Fe$_3$GeTe$_2$,[9] Fe$_4$GeTe$_2$,[10] Fe$_5$GeTe$_2$,[11] VSe$_2$,[12–16] and MnSe$_2$,[17] have been reported. In particular, room temperature (RT) intrinsic and extrinsic ferromagnetism has been demonstrated in monolayer VSe$_2$[12–16] and MnSe$_2$.[17] Alternatively, extrinsic RT ferromagnetic ordering has been introduced into the otherwise nonmagnetic 2D semiconductors by doping, e.g., monolayer (Co, Cr)-doped MoS$_2$,[18] V-doped WS$_2$,[19,20] V-doped WS$_2$,[21] and so on. Among them, the highest reported $T_C$ are 500 K for (Co, Cr)-doped MoS$_2$,[18] and 360 K for V-doped WSe$_2$. However, challenges remain in
identifying true monolayer 2D magnets with RT ferromagnetic ordering, and large-area low temperature controllable growth compatible with Si complementary metal oxide semiconductor (CMOS) technology. Their novel properties and emergent phenomena in the 2D limit require validation for applications in next-generation memory and information technology, spintronics, and quantum computing.

Chromium chalcogenides (Cr\(_{m}X_n\), X = S, Se, and Te), an emergent class of nonlayered materials with robust magnetic ordering, have attracted increasing research interest. For example, epitaxial Cr\(_2\)S\(_3\),\(^{[22]}\) CrSe,\(^{[23]}\) CrTe\(_2\),\(^{[24]}\) CrTe,\(^{[25]}\) and Cr\(_2\)Te\(_3\)\(^{[26–28]}\) thin films with thicknesses down to several nanometers have been grown by chemical vapor deposition and/or molecular beam epitaxy (MBE), showing high Curie temperature \(T_C\) (e.g., \(\approx305\) K for 8.7 nm CrTe\(_2\) thin film\(^{[24]}\)), and promising electrical transport properties (e.g., unconventional anomalous Hall effect\(^{[25]}\)) for spintronic devices. The successful synthesis of 2D Cr\(_3\)Te\(_4\)\(^{[29]}\) and Cr\(_3\)Te\(_6\)\(^{[30]}\) with monolayer thickness have also been recently reported, but characterization of their magnetic properties is lacking.

Herein, we report the RT ferromagnetism of 2D Cr\(_3\)Te\(_4\) in the monolayer form. Using the MBE method, we successfully grew crystalline Cr\(_3\)Te\(_4\) monolayers on graphite at low substrate temperatures, i.e., \(\approx100\) °C. The growth mode transits from 2D to 3D at elevated substrate temperatures above 300 °C. X-ray magnetic circular dichroism (XMCD) measurements reveal that the magnetic properties of 2D Cr\(_3\)Te\(_4\) are modulated by the film thickness. Thicker Cr\(_3\)Te\(_4\) films (\(\approx7\) nm) have a \(T_C\) lower than RT along its in-plane easy axis (\(\approx240\) K), while monolayer Cr\(_3\)Te\(_4\) displays RT ferromagnetism with \(T_C\) up to \(\approx344\) K, and perpendicular magnetic anisotropy. The RT ferromagnetism in monolayer Cr\(_3\)Te\(_4\) is confirmed by magnetic force microscopy (MFM). Density-functional theory (DFT) calculations suggest that the transition of the magnetic easy axis, and the enhancement of ferromagnetic coupling in the Cr\(_3\)Te\(_4\) monolayer are ascribed to interfacial effects at the monolayer Cr\(_3\)Te\(_4\)/graphite interface. The low temperature growth process, and the realization of RT magnetism in 2D Cr\(_3\)Te\(_4\) will facilitate the practical applications of 2D magnets in spintronic devices and high-density memory devices, compatible with Si CMOS technology.

2. Results and Discussion

Bulk chromium tellurides (Cr\(_m\)Te\(_n\)) are a group of materials with similar structures, which can be taken as derivates of the parent CrTe. CrTe has a nonlayered structure consisting of octahedral CrTe\(_2\) layers (an analogy to 1T MoS\(_2\) monolayer) connected by intercalated Cr atoms bonding with neighboring Te atoms in trigonal prism. With different amounts of Cr vacancies in the intercalated Cr layers, a range of Cr\(_m\)Te\(_n\) with varied stoichiometric ratios such as CrTe, Cr\(_2\)Te\(_2\), Cr\(_3\)Te\(_4\), and Cr\(_5\)Te\(_8\) can be formed and behave as ferromagnetic, antiferromagnetic, and ferrimagnetic materials.\(^{[31,32]}\) In particular, bulk Cr\(_3\)Te\(_4\) as shown in Figure 1a,b has a Te-Cr-Te-Cr quadruple nonlayered structure, with 50% Cr vacancies in the intercalated layer (space group \(P2/m\)). Bulk Cr\(_3\)Te\(_4\) is ferromagnetic with an averaged magnetic moment predicted to be 3.32 \(\mu_B\) per Cr atom by our DFT calculations, which is close to the previously reported values.\(^{[33]}\) It is noted that bulk Cr\(_3\)Te\(_4\) is not subject to a \(T'\)-like phase transition as indicated by the negligible distance difference between neighboring Cr-Cr atoms (Table 1). When the thickness decreases to the monolayer limit (five atom layers, i.e., Te-Cr-Te-Cr-Te), the composition ratio changes to Cr:Te = 1:2 (Figure 1c,d). Remarkably, monolayer Cr\(_3\)Te\(_4\) shows a \(T'\)-like phase transition, leading to periodic lattice distortion, i.e., a 0.32 Å difference between neighboring Cr-Cr bonds (Figure 1d). The averaged magnetic moment of Cr in ML Cr\(_3\)Te\(_4\) is 3.15 \(\mu_B\) per atom, close to that of bulk. Interestingly, the magnetic anisotropy changes from an in-plane easy magnetic axis of around 0.17 meV Cr\(^{-1}\) in bulk Cr\(_3\)Te\(_4\) to an out-of-plane easy axis of around –0.13 meV Cr\(^{-1}\) in freestanding monolayer
Cr$_3$Te$_4$, the latter of which can be further enhanced to $-2.17$ meV Cr$^{-1}$ by the graphene substrate. The unexpected switching of the easy axis of magnetization between the bulk and monolayer Cr$_3$Te$_4$ could be a result of the thickness effect. In the monolayer limit, Cr$_3$Te$_4$ has different local coordination environments and a stronger substrate effect, and hence different interatomic exchange, resulting in preferential perpendicular magnetic anisotropy.

To realize the 2D growth of the nonlayered Cr$_3$Te$_4$ crystal, MBE was selected as precise growth at the monolayer level is achievable by controlling source fluxes and substrate temperature (see the Experimental Section). The morphology of the MBE-grown Cr$_3$Te$_4$ ultrathin films on graphite varies significantly with the substrate temperature by keeping the evaporation rate constant. Figure 2a–d shows large-scale topographic images ($500 \times 500$ nm$^2$) of Cr$_3$Te$_4$ thin films (before the substrates were fully covered) grown at different substrate temperatures, 400, 300, 200, and 100 $^\circ$C, respectively. The as-grown Cr$_3$Te$_4$ flakes are typically regular hexagonal shape at high temperature (e.g., 400 and 300 $^\circ$C), and irregular dendritic shape at low temperature (e.g., 100 $^\circ$C). The thickness of the Cr$_3$Te$_4$ flakes decreases gradually with the substrate temperature, as revealed by the lateral profiles in Figure 2e–h. Typically, the thickness of the Cr$_3$Te$_4$ flakes is $\approx 4.5$ nm at 400 $^\circ$C (Figure 2e), 2.1–3.0 nm at 300 $^\circ$C (Figure 2f), 0.9–2.1 nm at 200 $^\circ$C (Figure 2g), and 0.9–1.5 nm at 100 $^\circ$C (Figure 2h). For the samples grown at 100 and 200 $^\circ$C, the height of the first layer is only 0.93 $\pm$ 0.02 nm, indicating the successful growth of 2D Cr$_3$Te$_4$ with monolayer thickness. This height consists of a Cr$_3$Te$_4$ monolayer (a Te-Cr-Te-Cr-Te quintuple layer) in addition with an interfacial separation between the Cr$_3$Te$_4$ film and the graphite substrate. The subsequent layers increase by an increment of $\approx 0.6$ or $\approx 0.3$ nm, which corresponds to a Te-Cr-Te-Cr quadruple layer (Figure 1) and a Te-Cr bi-atomic layer, respectively. The less observed $\approx 0.3$ nm increment is unique for such nonlayered materials.

Figure 2. Cr$_3$Te$_4$ thin films grown at various substrate temperature. a–d) Large-scale STM images ($500 \times 500$ nm$^2$) reveal the temperature-dependent morphology of Cr$_3$Te$_4$ films grown at: a) 400 $^\circ$C ($V_{tip} = 1.0$ V, $I_{tip} = 20$ pA), b) 300 $^\circ$C ($V_{tip} = 1.0$ V, $I_{tip} = 20$ pA), c) 200 $^\circ$C ($V_{tip} = 1.5$ V, $I_{tip} = 20$ pA), and d) 100 $^\circ$C ($V_{tip} = 2.0$ V, $I_{tip} = 10$ pA). e–h) Line profiles correspond to the yellow line in panels (a)–(d), respectively, revealing the different thicknesses of the Cr$_3$Te$_4$ flakes. The averaged thicknesses of the Cr$_3$Te$_4$ films are 4.45, 2.35, 1.43, and 1.0 nm, for (a)–(d).
are correspondingly given in Figure S1 in the Supporting Information. Since the Cr₃Te₄ crystal is nonlayered and possesses isotropic covalent bonding in all three dimensions, preferential growth in 3D island mode occurs under thermodynamic equilibrium conditions, e.g., at higher substrate temperatures. The interfacial interactions between the Cr₃Te₄ film and graphite substrate could play a critical role in promoting anisotropic growth, facilitating 2D growth mode at lower temperatures.

Further studies were carried out to investigate 2D Cr₃Te₄ films grown at 100 °C under optimized conditions. Figure 3a,b shows the topological images of Cr₃Te₄ monolayer and thicker film (~7 nm), respectively. Figure 3c represents a typical scanning tunneling microscope (STM) image with atomic resolution recorded at monolayer region. A quasi-hexagonal lattice structure with unidirectional 1D modulation is observed, similar to that commonly observed in 1T'-TMDs.[34,35] The unit cell is highlighted by a red rectangle, with a = 6.9 ± 0.1 Å and b = 4.0 ± 0.1 Å, agreeing well with the calculated lattice constant of Cr₃Te₄ (Table 1). The 1D modulation can be easily captured in the corresponding fast Fourier transform (FFT) image given in Figure 3e, attributing to the peaks highlighted by the white circles. This 1D superstructural modulation is still visible in the noncontact atomic force microscopy (nc-AFM) image shown in Figure 3d, indicating that it is probably due to lattice distortion. Variable temperature STM measurements reveal that the 1D modulation remains up to 206 K (Figure S3 in Supporting Information), indicating that it might not arise from a charge-density wave. The same atomic structure is observed in bi- and multilayer films, confirming the uniformity of the MBE-grown Cr₃Te₄ samples. Figure 3f shows the simulated STM image of monolayer Cr₃Te₄ on a single-layer graphene substrate (approximation of bulk graphite), based on the model presented in Figure 1c,d, consistent with the experimental STM image in Figure 3c. The 1D modulation, observed both experimentally and theoretically, is ascribed to the surface fluctuation as well as the bond distortion arising from periodic Cr vacancies in the Cr₃Te₄ monolayer (Figure S3 in Supporting Information).

Photoemission spectroscopy (PES) measurements based on synchrotron radiation were performed to determine the...
The XMCD signal at NI is stronger than that at GI, indicating the signal is most prominent at 78 K, and gradually decreases as the temperature increases, up to around 250 K at NI, and 300 K at GI. More XMCD data on ~1.5 nm Cr$_3$Te$_4$ grown on graphite and MoS$_2$ substrates for comparison are shown in Figures S9 and S10 in the Supporting Information, respectively.

Further analyses of the XMCD spectra based on sum rules analysis were carried out to estimate the magnetic moments of the Cr element. Figure 4e,f shows the summary of the magnetism of the Cr$_3$Te$_4$ monolayer and thicker films as a function of temperature. In the thicker film (Figure 4e), the total magnetic moment ($m = m_s + m_L$) in both NI and GI directions are similar from 78 to 300 K. $m_s$ is of ~1.23 $\mu_B$ per atom in the thicker Cr$_3$Te$_4$ film at 78 K, and almost vanishes at 250 K.

Fitting the experimental data by $M(T) \propto \left(1 - \frac{T}{T_c}\right)$, we yield $T_c$ of 248 ± 5 K in the GI direction, and 235 ± 5 K in the NI direction, both slightly below RT. The thicker film demonstrates less magnetic anisotropy, and its easy axis is parallel to the sample surface. The macroscopic magnetism of the thicker film was further investigated by SQUID measurements (Figure S11, Supporting Information). On the other hand, the Cr$_3$Te$_4$ monolayer shows pronounced perpendicular magnetic anisotropy, where the total magnetic moment at NI is much larger than that at GI. At 78 K, the estimated value is ~0.79 $\mu_B$ per Cr atom at NI, but only ~0.16 $\mu_B$ per atom at GI; at 300 K, the value decreases to ~0.09 $\mu_B$ at NI and ~0.05 $\mu_B$ at GI. Hence, the easy axis for spontaneous magnetization is normal to the sample surface of the monolayer Cr$_3$Te$_4$. From the fitting $M-T$ curves in Figure 4f, $T_c$ is around 344 ± 6 K for the Cr$_3$Te$_4$ monolayer. The observed enhancement of ferromagnetic coupling and the magnetic anisotropy transition in monolayer Cr$_3$Te$_4$ is consistent with our DFT prediction, but the measured magnetic moments are smaller than the predicted values. The estimated magnetic moment from the XMCD data, due to the residuals from the capping layers (e.g., the small Te$_{2}$O$_{3}$ components) or defects, might be different from the theoretically predicted value based on the idealized atomic structure. In addition, the saturation effect is hard to be eliminated as the lack of accurate data for X-ray absorption depth and mean electron escape depth. All these factors may contribute to the discrepancy.

The existence of ferromagnetism above RT in the Cr$_3$Te$_4$ monolayer is further confirmed by MFM. The MFM measurements were employed in ambient condition at RT to obtain a spatially resolved magnetic image of monolayer Cr$_3$Te$_4$. The AFM images and the corresponding MFM images of monolayer Cr$_3$Te$_4$ are shown in Figure 5. From the topographic AFM images, Cr$_3$Te$_4$ islands with sizes of tens to a hundred of nanometers can be seen in Figure 5a,b. The corresponding MFM images (Figure 5c,d) recorded at the same area shows clear magnetic phase contrast and display the same Cr$_3$Te$_4$ topography. In Figure 5d, the dotted line highlights the contour of the monolayer Cr$_3$Te$_4$ with obvious MFM signals. The homogeneity of the MFM phase signal within the Cr$_3$Te$_4$ region indicates its out-of-plane ferromagnetism at RT, where the spins of the monolayer Cr$_3$Te$_4$ are aligned perpendicular to the substrate and parallel to the MFM tip, maximizing the detected signal. The MFM signals quench away in a couple of
hours in ambient condition, indicating the relative air instability of the Cr$_3$Te$_4$ films (Figure S8, Supporting Information). Combining the XMCD and MFM data, we conclude that monolayer Cr$_3$Te$_4$ is ferromagnetic at RT with out-of-plane easy axis. Further DFT calculations were performed to understand the underlying physics of the thickness-dependent magnetic properties. Figure 6a shows that the spin density is dominantly around the Cr atoms without contribution from graphene substrate. Although the interface spacing between Cr$_3$Te$_4$ and graphene is as large as 3.56 Å within the range of van der Waals bonding, the interaction between them is nontrivial. Significant charge redistribution is observed in Figure 6b. The charge density is depleted on the proximity of graphene, and the excess charge density is accumulated on the bottom of Cr$_3$Te$_4$ monolayer, indicating that the charge transfers from graphene into Cr$_3$Te$_4$. Further Bader analysis suggests about 0.1 e$^-$ transfers from graphene into the monolayer Cr$_3$Te$_4$. This is ascribed to interfacial electrostatic interaction. Figure 6c shows the in-plane averaged electrostatic potential across the Cr$_3$Te$_4$/graphene interface, from which we can obtain the WF of Cr$_3$Te$_4$ and graphene, respectively. The calculated WF is 4.3 eV for graphene, and 5.3 eV for Cr$_3$Te$_4$ monolayer, consistent with our experimentally measured WF data. The WF difference between them leads to the charge transfer when forming Cr$_3$Te$_4$/graphene heterointerface.

From the project density of state (PDOS) shown in Figure 6d, we can also see that the Dirac cone of graphene is shifted to higher energy, indicating a hole-doping in graphene due to charge transfer to Cr$_3$Te$_4$. Besides, the PDOSs of Cr atoms near the interface and C atoms show a similar profile around the Fermi level, which clearly suggests orbital overlap between them, especially between d$_{xz}$, d$_{yz}$, and d$_{z^2}$ orbitals of Cr and p$_z$ orbital of graphene as mediated by the Te p$_z$ orbital in between. The p$_z$ orbital of graphene may pin the direction of the magnetic moments on Cr atoms, leading to an enhanced out-of-plane magnetic easy axis. This was further supported by the atomic and orbital resolved MAE. As shown in Table S3 in the Supporting Information, the main contribution of MAE is from the Te atom. This is because Te has much stronger spin–orbital coupling (SOC) than Cr as the SOC strength is linear to the atomic number $Z$ in the power of 4 ($Z_{Cr} = 24$ and $Z_{Te} = 52$). Overall, the MAE of both Cr and Te increases from bulk,
free-standing monolayer to Cr$_3$Te$_4$/graphene heterostructure. Interestingly, the MAE of Te is negative in bulk form, indicative of an in-plane orientation. These results explain the overall MAE of the three cases presented in the main article.

Considering the dominant role of Te in the overall MAE, only the orbital-resolved MAEs of Te atoms are shown in Table S4 in the Supporting Information. In bulk Cr$_3$Te$_4$, the coupling between p$_x$ and p$_y$ contributes most to the MAE and is negative. Such coupling also dominates in ML Cr$_3$Te$_4$ but becomes positive. Interestingly, the inclusion of graphene enhances the coupling with p$_z$ orbital, but decreases in-plane orbital (p$_x$ and p$_y$) coupling, which leads to an enhanced MAE of the Cr$_3$Te$_4$/graphene heterostructure. To further confirm the interfacial effects, we increase the interfacial distance between Cr$_3$Te$_4$ and graphene to >10 Å, and find that it has similar MAE with the free-standing Cr$_3$Te$_4$ with a small enhancement of 0.07 meV Cr$^{-1}$. All these results support the claim that the enhanced out-of-plane MAE of CrTe on the graphene is due to the interfacial coupling. Such interface coupling is robust against the thickness effects of both Cr$_3$Te$_4$ and graphene. Increasing the thickness of either chromium telluride monolayer (to nine atomic layers) or graphene (to bilayer) leads to slight change of MAE to −1.38 and −2.02 meV Cr$^{-1}$, respectively. Increasing the thickness of graphene has negligible effects on the MAE due to the large distance (>7 Å) between the bottom graphene and Cr$_3$Te$_4$ ML and the weak van der Waals interaction between graphene layers. However, the increase of Cr$_3$Te$_4$ thickness decreases the MAE but retains the out-of-plane anisotropy. This is in line with the trend that the bulk Cr$_3$Te$_4$ tends to have an in-plane magnetic anisotropy (positive MAE value).

We note that the Cr atoms show two different magnetic moments of 3.20 and 3.10 $\mu_B$, respectively, indicating the different oxidization states of the Cr atoms. The double exchange mechanism could be applied in such a situation. The parallel alignment (ferromagnetic coupling) of the magnetic moments on Cr ions allows the hopping of electrons between the Cr ions with different oxidation state, which reduces the kinetic energy.$^{[41]}$

Based on our experiments and DFT calculations, we suggest that the 2D growth mode at low substrate temperature (100 °C), the transition of out-of-plane magnetization easy axis, and the high Curie temperature for the Cr$_3$Te$_4$ monolayer on graphite, are due to the interfacial interaction-induced orbital overlap.

We further perform Monte Carlo (MC) simulations with the Metropolis algorithm$^{[42,43]}$ to estimate the Curie temperature.

Figure 5. AFM and MFM images of monolayer Cr$_3$Te$_4$. a) Large-scale morphologic image (5 × 5 μm$^2$) shows the irregular dendritic shape of monolayer Cr$_3$Te$_4$ grown at 100 °C, and c) the zoom-in (1.5 × 1.5 μm$^2$) of the area highlighted by a black square in panel (a). b,d) The MFM images corresponding to (a) and (c), respectively, revealing the RT ferromagnetism in the sample.
of the Cr$_3$Te$_4$ monolayer. Since the monolayer Cr$_3$Te$_4$ has an out-of-plane easy magnetic axis and the magnetic anisotropy is large (2.17 meV f.u.$^{-1}$), we can apply the Ising model for Monte Carlo simulations (more details of the method are given in the Supporting Information). As shown in Figure 4g, both the temperature-dependent magnetization and magnetic susceptibility suggest a $T_C$ of 300 K for Cr$_3$Te$_4$ monolayer on graphene, in good agreement with our experimental results. For comparison, Monte Carlo simulations were also performed for a reported perfect 7-atomic Cr$_3$Te$_4$, yielding a $T_C$ of 520 K, in line with a previous value (see Figure S16, Supporting Information).

3. Experimental Section

Cr$_3$Te$_4$ Growth: The Cr$_3$Te$_4$ films were grown on commercial highly oriented pyrolytic graphite (HOPG) in an MBE chamber attached to the Omicron LT-STM system with a base pressure of $1 \times 10^{-9}$ mbar. The HOPG substrates were freshly exfoliated and then annealed at 500 °C for more than 1 h before MBE growth. Elemental Cr and Te were used as effusion source materials. During the deposition, the Te:Cr ratio was kept high ($\approx 10:1$) to maintain Te overpressure since it was more volatile. The graphite substrate was held at elevated temperature (e.g., 100 to 400 °C) to provide sufficient adatom diffusion energy for surface atom mobility to reach near thermodynamic equilibrium. LT-STM measurements were done in situ. For ex situ X-ray photoelectron spectroscopy (XPS)/XAS measurements, nonmagnetic Te capping layers were deposited onto the Cr$_3$Te$_4$ surface to prevent contamination in ambient environment during the transport. The Te capping layers were removed by annealing at 250 °C for 30 min as confirmed by the XPS and STM measurements (Supporting Information).

LT-STM and nc-AFM Measurements: LT-STM and nc-AFM studies were performed at 77 K in an Omicron LT-STM system interfaced to a Nanonis controller equipped with STM/qPlus sensor. Electrochemically etched tungsten tips were used, where a bias voltage was applied on it, and the sample holder was grounded. The STM images were acquired in constant-height mode with an oscillation amplitude of 1 Å was used to record the
frequency shift ($\Delta f$) of the qPlus resonator (sensor frequency $f_s = 24$ kHz, $Q = 8000$). A lock-in technique was used to measure the $\Delta f/\Delta V$ spectra, with a modulation of 625 Hz and 30 mV.

**PES and XAS Measurements**: PES, XAS, and XMCD measurements were carried out at the SINS beam line of the Singapore Synchrotron Light Source (SLS). The incident photon energy was 700 eV and the energy resolution was 0.2 eV for the core level spectra. The XAS and XMCD at V L2,3 edge at RT were obtained by recording the sample current (total yield mode) as a function of photon energy. Elliptically polarized light with a degree of circular polarization (DCP) = 80 and an energy resolution of 0.3 eV was employed for the XMCD measurements. To measure the in-plane and out-of-plane spin and orbital magnetic moments, the light was incident at a grazing or normal angle, respectively, from the sample surface, with its propagation direction along the sample in-plane or out-of-plane magnetization direction. An external magnetic field of $\pm 1$ T was applied to magnetize the sample along the in-plane or out-of-plane direction. The XMCD was done by changing the direction of applied magnetic field while keeping the helicity of the light.

**MFM Measurements**: MFM measurements of the Cr$_3$Te$_4$ samples were conducted by Bruker Dimension ICON system under ambient condition. A magnetic MESP-HM-V2 probe (antimony doped Si with magnetic CoCr coating) with high magnetic moment of $3e^{-13}$ emu and nominal coercivity of 400 Oe was used. The MFM phase signal was recorded in a lift-while-tapping mode with the sample-probe distance of 20 nm and high drive frequency of 0.5 kHz to achieve good signal-to-noise ratio. The MFM phase image was collected at 256 × 256 pixels in around 8 min for one single scan.

**DFT and Monte Carlo Simulations**: The first-principles calculations were performed based on the projector-augmented-wave method as implemented in the Vienna ab initio simulation package (VASP). The exchange-correlation interaction was treated with the Perdew–Burke–Ernzerhof form of the generalized gradient approximation. The plane-wave cutoff energy was set to 500 eV. The lattice parameters and atomic positions were fully relaxed until the energy and force on each atom were converged to the order of $10^{-4}$ eV/Å. The Hubbard U of 3.2 eV was adopted to deal with the strongly correlated effect between Cr 3d electrons. The plane-wave cutoff energy was set to 500 eV. The lattice parameters and atomic positions were fully relaxed until the energy and force on each atom were converged to $10^{-4}$ eV/Å and $10^{-3}$ eV Å$^{-1}$, respectively. The interface of Cr$_3$Te$_4$ monolayer/graphene was modeled by placing (3 × 3) monolayer Cr$_3$Te$_4$ supercell on (3 × 3) graphene supercell, in which about 3.7% tensile strain was applied on Cr$_3$Te$_4$ monolayer. In this model, graphene monolayer was used to represent graphite substrate, which did not affect the main results due to the weak interlayer interaction between graphene layers. For the interface calculations, Van der Waals correction was considered using the method of Grimme (DFT-D3). The STM images were simulated using the Tersoff–Hamann method. The Bader charge analysis was used to estimate the number of valence charges. For the comparison of magnetic anisotropy energy, the spin–orbit coupling effect was included in the calculations with a higher accuracy of $10^{-8}$ eV. In all calculations, the dipole correction was applied. For the comparison of magnetic anisotropy energy, the spin–orbit coupling effect was included in the calculations with a higher accuracy of $10^{-8}$ eV. In all calculations, the dipole correction was applied. The Monte Carlo simulations were performed based on Ising spin Hamiltonians using 60 × 60 × 1 supercells with periodic boundary conditions. For each temperature, 5 × 10$^7$ steps were used to reach thermal equilibrium. The first 2.5 × 10$^7$ steps were discarded, and the last 2.5 × 10$^7$ steps were used to calculate the temperature-dependent physical quantities.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

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

**Keywords**

2D magnets, monolayer chromium telluride, room temperature ferromagnetism, scanning tunneling microscopy, X-ray magnetic circular dichroism

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