To harness the intriguing properties of 2D van der Waals (vdW) ferromagnets (FMs) for versatile applications, the key challenge lies in the reliable material synthesis for scalable device production. Here, the epitaxial growth of single-crystalline $1\text{T}-\text{CrTe}_2$ thin films on 2-inch sapphire substrates are demonstrated. Benefiting from the uniform surface energy of the dangling bond-free $\text{Al}_2\text{O}_3$(0001) surface, the layer-by-layer vdW growth mode is observed right from the initial growth stage, which warrants precise control of the sample thickness beyond three monolayer and homogeneous surface morphology across the entire wafer. Moreover, the presence of the Coulomb interaction at the $\text{CrTe}_2/\text{Al}_2\text{O}_3$ interface plays an important role in tailoring the anomalous Hall response, and the structural optimization of the $\text{CrTe}_2$-based spin-orbit torque device leads to a substantial switching power reduction by 54%. The results may lay out a general framework for the design of energy-efficient spintronics based on configurable vdW FMs.
Figure 1. Epitaxial growth of wafer-scale 1T-CrTe₂ thin films. a) In situ RHEED pattern of the CrTe₂ film grown on the Al₂O₃ substrate. The spacing between the two first-order streaks (dashed white lines) is used to deduce the in-plane lattice constant. b) Cross-sectional high-resolution STEM image of the MBE-grown CrTe₂ sample. The atomic arrangement is consistent with the crystal structure of 1T-CrTe₂. The Cr and Te atoms are labeled as pink and yellow spheres, respectively. c) AFM images were taken at 36 different positions across the 2-inch 9 ML CrTe₂ wafer, and the statistical histogram of the measured root-mean-square roughness \( R_q \) confirms the homogeneous surface morphology. d) The XRD spectra of CrTe₂ films ranging from 9 to 33 ML. e) Identical rocking curves of the CrTe₂ (002) film peaks with the same FWHM value as the Al₂O₃ substrate highlight the very high crystalline quality of the CrTe₂ films. f) XRR data of the thickness-dependent CrTe₂ films with oscillatory Kiessig fringes. The spacing between neighboring interference peaks (labeled as arrows) is inversely correlated to the film thickness.

Electrochemical transfer techniques (i.e., the so-called top-down method), where a-few-layer flakes are exfoliated from their bulk crystals[1,2,5] Although it seems to be cost-effective and straightforward, this approach inevitably suffers from low productivity, small sample size, and limited thickness/orientation controllability.[1,5,6] To comply with the mass-production prerequisite, alternative bottom-up methods, such as the chemical vapor deposition (CVD)[7] the seeded growth,[8] and dual-coupling-guided growth,[9] are developed to fabricate large-size vdW FMs. However, since the majority of these methods are chemical reactions under equilibrium conditions, the grown 2D FMs would follow the Wulff construction which constantly leads to the edge-induced nucleation and the formation of self-limited triangular or hexagonal islands.[10–12] Even though dedicated surface treatments have recently been introduced to permit uniform nucleation,[8,9,11,12] the high growth rate would make the film thickness control challenging. As a result, the layer-dependent electronic/magnetic properties of vdW FMs and the opportunities to implement their heterostructure/superlattice in spintronics remain elusive.

In this Article, we report the wafer-scale growth of uniform CrTe₂ thin films and Bi₂Te₃/CrTe₂ heterostructures by molecular beam epitaxy (MBE). Owing to its low-energy physical deposition nature and non-equilibrium growth dynamics,[13–15] MBE ensures high homogeneity with atomic-scale surface roughness in the grown 2-inch samples, and the vdW layer-by-layer growth mode empowers us with the ability to finely tune the film thickness across the 2D-to-bulk regions. Together with the ab initio calculation of electronic band structure, we unveil that the quantum confinement and interfacial effects in the ultra-thin CrTe₂ film would effectively modify its Berry curvature and magnetic order strength, triggering the polarity change of the resulting anomalous Hall signal. Furthermore, by combining the ferromagnetic CrTe₂ with the topological insulators Bi₂Te₃, we realize the spin-orbit torque (SOT)-driven magnetization switching in the integrated vdW heterostructures with wafer-scale uniformity, a high endurance of more than \( 5 \times 10^4 \) read–write cycles, and the manageable thickness of the CrTe₂ layer offers an additional degree of freedom to tailor the performance of the SOT devices.

2. Results and Discussion

2.1. Wafer-Scale Epitaxial Growth of vdW FM CrTe₂ Thin Films

Experimentally, 2-inch Al₂O₃(0001) wafers were chosen as the substrate. The absence of dangling bonds at the substrate surface and the weak bonding energy between CrTe₂ and Al₂O₃ guarantee a uniform surface energy distribution, which in turns promotes the van der Waals epitaxial growth of CrTe₂.[16,17] Accordingly, a sharp reflection high-energy electron diffraction (RHEED) streaky pattern develops for the first layer, and the spacing between the two first-order reciprocal lattice rods in Figure 1a corresponds to an in-plane lattice constant of \( a = 3.84 \) Å, in agreement with expected 1T-CrTe₂ data.[18] In the meantime, the layer-by-layer vdW growth mode is evident from the RHEED oscillation, which allows for the control of film thickness down to 3 monolayer (ML), as elaborated in Section S1, Supporting Information. Furthermore, high-resolution transmission electron
microscopy (TEM) (Figure 1b) shows an intact, layered crystalline structure with a sharp interface, and within each constituent layer, the Cr (pink spheres) and Te (yellow spheres) atoms rigorously follow the Te–Cr–Te Z-shaped stacking configuration, validating the 1T phase of the as-grown CrTe₂ sample.[7]

Next, we conducted atomic force microscope (AFM) measurements to examine the conformity of the CrTe₂ film, and Figure 1c illustrates one dataset of the 9 ML sample with two noticeable features. First of all, in contrast to μm-size triangular or hexagonal terraces (i.e., as they would stem from the island nucleation process) observed in conventional vdW 2D materials,[19,20] the AFM mapping of the CrTe₂ film shows quite uniform nucleation and homogeneous atomic-scale surface morphology (i.e., the root-mean-square roughness $R_q$ is much less than the height of one CrTe₂ ML), which may take advantages of both the surface energy reduction (i.e., owing to the vdW gap at the CrTe₂/Al₂O₃ interface) and the long diffusion length of the adatoms.[11] Second, the histogram of $R_q$ collected from 36 different areas exhibits a normal distribution with a narrow standard deviation of $3\sigma = 0.28$ Å, hence manifesting the high uniformity across the whole 2-inch wafer (Section S2 Supporting Information).

In terms of film crystallinity, Figure 1d displays the X-ray diffraction (XRD) results of five MBE-grown CrTe₂ samples with thicknesses ($d$) ranging from 9 to 33 ML. It can be clearly seen that in addition to the substrate signal, all spectra display only a series of CrTe₂ (00l) diffraction peaks without any impurity phase within the instrument detection limit, and the extracted out-of-plane lattice parameter $c = 6.10$ Å is consistent with the 1T-CrTe₂ structure as well. Strikingly, Figure 1e reveals identical normalized CrTe₂ (002) Ω-scan curves with negligible mosaicity broadening (i.e., no misorientation of crystallites) compared with the Al₂O₃ (0006) reference line-shape, and the full width at half maximum (FWHM) of $0.037° \pm 0.001°$ is among the narrowest values reported for vdW films (i.e., it is noted that when the CrTe₂ film thickness is below 5 ML, the overall XRD curve is mainly dominated by the substrate signal, and this bulk-sensitive analytical method is not suitable to characterize ultra-thin CrTe₂ samples).[20–22] Besides, the pronounced Kiessig fringes observed in the X-ray reflection (XRR) data in Figure 1f not only quantify the film thickness, but also attest the low interlayer roughness of the CrTe₂ samples. Along with highly consistent RHEED and XRD/XRR results taken at different positions across the 2-inch samples (Section S2, Supporting Information), our comprehensive structural characterizations confirm the high quality and reproducibility of the wafer-scale 1T-CrTe₂ thin films, therefore offering a solid foundation for reliable device applications.

2.2. Tunable Anomalous Hall Response Driven by Temperature and Dimensionality

To determine the magnetic properties of the epitaxial 1T-CrTe₂ thin films, we fabricated mm-scale six-probe Hall bar devices on the 2-inch wafers (Figure 2a), and performed magneto-transport measurements at low temperatures. Figure 2b exemplifies the characteristic anomalous Hall effect (AHE) on a 14 ML CrTe₂ sample, where the nearly square-shaped $R_{xy}$ hysteresis loop and a butterfly-type double-split magneto-resistance (MR) curve highlight the well-established spontaneous magnetization at $T = 10$ K. Likewise, the strong perpendicular magnetic anisotropy (PMA) of the films is also verified by the element-specific X-ray magnetic circular dichroism (XMCD) hysteresis loops recorded at the Cr L₃ absorption edge, confirming that the magnetism originates from the Cr atoms (Section S3, Supporting Information). As visualized in Figure 2c, the XMCD loop exhibits the same behavior as the AHE one as a function of the applied magnetic field, which identifies that the ferromagnetism evolves together with the anisotropy of the orbital moment.[23]

Given that the reduced dimensionality can help to modify the electronic band structure and magnetic properties of vdW...
2D FM$_t$\cite{2,5,24} we subsequently conducted systematic magnetoelectric transport measurements on three CrTe$_2$ samples with $d = 5$, 9, and 14 MLs. Remarkably, the AHE results show a distinct thickness-dependent trait. As evidenced in Figure 2d, the coercive field $H_c$ decreases from 1.17 T (14 ML) to 1.15 T (9 ML) and 0.98 T (5 ML) at $T = 1.5$ K, (i.e., this $H_c$-$d$ relation holds true at temperatures ranging from 1.5 K to 120 K) and this evolution trend suggests the weakening of the magnetism with reduced dimensionality. More importantly, the AHE hysteresis loop reverses its polarity from negative to positive when the thickness is reduced to 5 ML at $T = 1.5$ K. Similarly, such a negative-to-positive transition behavior of the AHE polarity is also observed in the bulk-type 14 and 9 ML CrTe$_2$ samples during the warming-up process, yet the corresponding transition temperature $T^*$ is reduced from 115 to 70 K (i.e., in contrast, the AHE sign remains to be positive in the 2D-limit 5 ML thin film regardless of the base temperature).

To understand this dimensionality-dependent anomalous Hall results, we applied density-functional theory (DFT) calculations to elucidate the underlying physics.\cite{25–28} Generally, the AHE response of CrTe$_2$ has both intrinsic and extrinsic contributions. Given the close interplay between the Berry curvature and electronic band structure, we investigated how the intrinsic Berry phase affects the intrinsic AHE polarity of CrTe$_2$ with the reduction of the sample thickness. In particular, based on the electronic structures of the monolayer (Figure 3a) and bulk CrTe$_2$ (Figure 3b), we further took into account the Coulomb interaction from the correlated Cr 3d orbitals by using the DFT + $U$ calculation with the on-site Coulomb potential $U$ being a fitting parameter.\cite{29,30} As outlined in Figure 3c, the sign of the intrinsic anomalous Hall conductivity $\sigma_{xy}$ of the bulk CrTe$_2$ stays negative for $0 < U < 5$ eV, whereas it becomes positive when $U > 2$ eV for the thinner layer CrTe$_2$ case (Section S4, Supporting Information), qualitatively consistent with the experimental data in Figure 2. Here, we need to point out that only the $\sigma_{xy}$ variation trend as a function of $U$ makes sense (i.e., instead of the calculated result with a specific value of $U$) because a reliable $U$ value cannot be obtained from neither experimental nor theoretical estimations. Consequently, the above results suggest that the intrinsic Berry curvature can cause the AHE sign change in CrTe$_2$ with reduced sample thickness, and this conclusion also agrees with the fact that effective Coulomb interaction increases in low-dimensional system due to the suppression in electron kinetic energy. On the other hand, since the extrinsic contributions from disorder or impurity scatterings are hard to quantify with ab initio methods, other possible mechanisms which may cause the change of the AHE polarity need further investigation. For instance, transferring the CrTe$_2$ films to another type of substrate may help to justify the influence of the interfacial Coulomb interaction on the PMA and AHE response.

Likewise, we can attribute the temperature-dependence of the anomalous Hall resistance in the thick CrTe$_2$ samples (i.e., $d \geq 9$ ML) to the modulation of Berry curvature associated with the magnetization change.\cite{28,31} Specifically, the diminished magnetic moment at elevated temperatures may bring about a redistribution of electronic states and switch the AHE polarity below a critical value of $M_C$ (i.e., above a characteristic transition temperature $T^*$),\cite{31} given that the Berry curvature is highly sensitive to the occupied density of states near the Fermi level. Following the same scenario, as the overall magnetization is gradually suppressed in the thinner films, it is expected that the 9 ML CrTe$_2$ sample would reach $M_C$ at a lower transition temperature $T^*$ compared to the 15 ML counterpart. In the ultra-thin film case (i.e., $d = 5$ ML) where the saturated magnetic moment is always smaller than $M_C$, its temperature-invariant positive AHE response is therefore in line with that of the bulk-type CrTe$_2$ system in the high-temperature region.

2.3. Energy-Efficient SOT Switching in Bi$_2$Te$_3$/CrTe$_2$ Heterostructures

Spin-orbit-torque (SOT) has been utilized for next-generation magnetic random-access memory (MRAM) technology, owing to its low power consumption and logic-programmable capability.\cite{32,33} Considering that the SOT-driven magnetization switching is determined by the charge-to-spin conversion efficiency, it has been discovered that topological quantum materials (e.g., topological insulators, Weyl/Dirac semimetals), in which the intrinsic spin-orbit coupling (SOC) is strong enough to cause band inversion, can enable sufficient current-induced spin polarization along the surface/edge states through the spin-momentum locking mechanism.\cite{14,15} Apart from the spin current channel, the magnetic strength in the adjacent FM layer is another key factor to affect the switching current level. In this regard, the tunable magnetism identified in our low-dimensional
CrTe₂ samples, assuming that they can be incorporated into a SOT structure, could afford more flexibility for optimizing the device performance.

Accordingly, to fully explore the potential of CrTe₂ for SOT-MRAM applications, we adopted the same vdW growth procedure to prepare Bi₂Te₃/CrTe₂ heterostructures by MBE (i.e., in order to make a fair comparison, the top Bi₂Te₃ layer thickness is fixed to 18 nm in all samples investigated in this work). As shown in Figure 4a, the ultra-smooth CrTe₂ surface and the in situ integration process in the ultra-high vacuum environment result in the formation of a sharp hetero-interface, and the energy dispersive X-ray (EDX) spectroscopy images ascertain the uniform element distribution without observable inter-layer diffusion (Section S5, Supporting Information). After sample growth, standard μm-sized cross-bar device arrays were fabricated on the 2-inch Bi₂Te₃/CrTe₂ wafers (Figure 4b), and the relevant CrTe₂-thickness-dependent current-driven magnetization switching results are presented in Figure 4c. Guided by the schematic diagram in Figure 4a, the presence of a constant in-plane field \( B_x = +0.09 \, \text{T} \) sets up the initial magnetization state of the sample in the (+x, +z) quadrant, and the applied DC current \( I_{\text{DC}} \) along the ±x-direction dictates the effective spin-orbit field \( B_{SO} = \lambda_{SO} \cdot I_{\text{TI}} \cdot \sigma \times M \) (where \( \lambda_{SO} \) is the coefficient characterizing the SOC strength, \( I_{\text{TI}} \) is the charge current component conducting through the Bi₂Te₃ channel, and \( \sigma = \sigma_x \cdot \hat{y} \) is the electron spin accumulated at the Bi₂Te₃/CrTe₂ interface). Consequently, as the DC current is successively scanned from +25 to −25 mA, the measured Hall resistances \( R_{xy} \) (blue circles in Figure 4c) all retain at constant values until the positive-to-negative transitions occur in the large negative \( I_{\text{DC}} \) domains (i.e., the parallel (+B_x, +I_{\text{DC}}) configuration stabilizes the magnetic moment \( M_z \) along the +z-axis, whereas the negative DC bias could trigger the magnetization switching as long as the reversed \( B_{SO} \) overcomes the intrinsic magnetic anisotropy field \( B_K \)). On the other hand, the application of an opposite \( B_x = -0.09 \, \text{T} \) changes the initial condition of \( M \), and the observation of the clockwise \( R_{xy} \) hysteresis loops (red triangles in Figure 4c) are in accordance with the same damping-like SOT chirality, hence confirming the deterministic switching scenario.

Most intriguingly, the \( (B_x, I_{\text{DC}}) \)-dependent anomalous Hall data in Figure 4c discloses a dramatic reduction of the threshold switching current level \( I_{SW} \) when the top CrTe₂ layer thickness \( d \) decreases from 21 ML (17.5 mA) to 5 ML (7.5 mA) at \( T = 120 \, \text{K} \) (i.e., the AHE polarity remains positive for all Bi₂Te₃ (18 nm)/CrTe₂ (d) thin films at \( T = 120 \, \text{K} \), as shown in Section S6, Supporting Information). Equivalently, it means that
the dynamic power dissipation $P_{SW}$ can be reduced by up to 54% with appropriate bilayer structural engineering (Figure 4d). To appreciate such a positive $I_{SW}$-$d$ correlation, it is recalled that the reduced dimensionality of the CrTe$_2$ film would not only weaken its perpendicular magnetic anisotropy (e.g., the ferromagnetism becomes so weak that the SOT-driven magnetization switching of the $d = 3$ ML device can only be observed when $T \leq 100$ K, as shown in Section S7, Supporting Information), but also make the thinner FM layer more insulating so that a larger portion of charge current is regulated inside the Bi$_2$Te$_3$ channel. Governed by the SOT operational principle that the modulation of the CrTe$_2$ FM order mainly relies on the competition/balancing between the torques exerted by the effective fields, it can therefore be concluded that a lower DC current can meet the magnetic requirements by the SOT operational principle that the modulation of the CrTe$_2$ FM order mainly relies on the competition/balancing between the torques exerted by the effective fields, ($\gamma$ is the gyromagnetic ratio, and $r_k$ is the torque exerted by PMA) thanks to the attenuated $B$ and higher $I_{TM}/I_{DC}$ ratio in the Bi$_2$Te$_3$/CrTe$_2$ (5 ML)-based SOT device. By further subtracting the shunting current via the parallel two-channel model (see Section S8, Supporting Information), the critical current density available for the charge-to-spin conversion is found to stay at $J_{PL} = 2.64 \times 10^7$ A cm$^{-2}$ as long as the bottom CrTe$_2$ layer exceeds 9 ML, while it drops to $1.93 \times 10^6$ A cm$^{-2}$ for the $d = 5$ ML case, again highlighting the unique advantage of vdW FMs.

In conclusion, we have achieved the epitaxial growth of uniform 1T-CrTe$_2$ films on a wafer-scale by MBE, and established a vdW integration strategy that allows for the in situ construction of Bi$_2$Te$_3$/CrTe$_2$ heterostructures. Endorsed by the coexistence of the intrinsic PMA, atomically-sharp interfaces, and strong SOC, we showcase the compelling SOT device performance of this simple bilayer stack with lower switching power and low device-to-device variation ($<2.5\%$) of the SOT crossbar array across the 2-inch wafer, our Bi$_2$Te$_3$/CrTe$_2$ heterostructures can serve as a compelling material system for high-density MRAM. In addition, the design rule inaugurated in our work may expedite the search for new 2D vdW FMs-based heterostructures, which would further enable wafer-scale vdW material synthesis and feasible spintronic device applications.

3. Conclusions

In conclusion, we have achieved the epitaxial growth of uniform 1T-CrTe$_2$ films on a wafer-scale by MBE, and established a vdW integration strategy that allows for the in situ construction of Bi$_2$Te$_3$/CrTe$_2$ heterostructures. Endorsed by the coexistence of the intrinsic PMA, atomically-sharp interfaces, and strong SOC, we showcase the compelling SOT device performance of this simple bilayer stack with lower switching power and low device-to-device variation ($<2.5\%$) of the SOT crossbar array across the 2-inch wafer, our Bi$_2$Te$_3$/CrTe$_2$ heterostructures can serve as a compelling material system for high-density MRAM. In addition, the design rule inaugurated in our work may expedite the search for new 2D vdW FMs-based heterostructures, which would further enable wafer-scale vdW material synthesis and feasible spintronic device applications.

4. Experimental Section

Sample Fabrication and Structural Characterizations: Wafer-scale CrTe$_2$ thin films and Bi$_2$Te$_3$/CrTe$_2$ heterostructures were grown on 2-inch Al$_2$O$_3$(0001) substrates by MBE in a vacuum of $1 \times 10^{-10}$ mbar. Prior to the sample growth, the Al$_2$O$_3$ substrate was pre-annealed at 600 °C in order to remove any absorbed contamination. During the MBE growth, the growth temperature for CrTe$_2$ and Bi$_2$Te$_3$ was kept at 200 °C, and the sample manipulator was constantly rotated to ensure uniformity; high-purity Cr and Bi atoms were evaporated from standard Knudsen cells, while the Te atoms were evaporated by a thermal cracker cell. A beam flux monitor was used to measure the flux ratio, and RHEED was used to monitor the real-time growth process. After sample growth, slices of CrTe$_2$ with different crystal orientations were milled out using the FIB technique (TESCAN LYRA3 FIB-SEM, TESCAN, Czech Republic), and a probe aberration-corrected scanning transmission electron microscopy (Cs-STEM, Themos Z3000, FEI, USA) was employed to resolve the crystal structure. The STEM was equipped with energy-dispersive X-ray analysis (EDX, Bruker Super-X, Bruker, USA), allowing for the determination of the Cr-to-Te stoichiometric ratio. Additionally, X-ray diffraction and reflectivity were used to confirm the lattice constant and to calibrate the thickness.

Device Fabrication and Transport Measurements: The CrTe$_2$ thin films were etched into a six-probe Hall bar geometry with typical channel size of $2 \times 1$ mm$^2$. The electrodes were made by welding small pieces of indium onto the contact areas. The magneto-transport measurements were performed in a He$^4$ refrigerator (Oxford Teslatron PT system). Multiple lock-in amplifiers and Keithley source meters (with an excitation AC current amplitude of $I = 1 \mu$A) were applied to the sample, and the temperature, magnetic field, and lock-in frequency served as the experimental variables. For the SOT-driven magnetization switching measurements, the cross-bar patterns of the CrTe$_2$/Bi$_2$Te$_3$ heterostructures were prepared by a standard photo-lithography process, and the Ti (15 nm)/Au (150 nm) electrodes were defined by e-beam evaporation. After device fabrication, a 2-ms writing current pulse was applied by Keithley 6221 while another 2-ms reading current pulse was applied thereafter to measure the $R_{xy}$ value by Keithley 2182.

X-Ray Absorption Spectroscopy and XMCD Measurements: Element-specific X-ray absorption spectroscopy (XAS) and XMCD measurements were performed on beamline 106 at the Diamond Light Source (Oxfordshire, UK), allowing for the probing of the magnetic ground state of the CrTe$_2$ layers. The base temperature was 2 K and a magnetic field of $\sim 6$ T was applied along the surface normal direction (out-of-plane). The spectroscopic data was acquired in total-electron-yield detection mode which has an 1/e sampling depth of $\sim$5 nm. The XAS spectra were recorded at the Cr L$_{3,2}$ edges between 560 and 595 eV. The XMCD results were obtained by taking the difference between two XAS spectra with the helicity vector of the circularly polarized X-rays parallel and antiparallel to the magnetic field, respectively.

Density Functional Theory Calculations: The ab initio calculations were performed within the framework of DFT as implemented in the Vienna ab initio simulation package (VASP), with the exchange-correlation functional considered in the generalized gradient approximation of the Perdew–Burke–Ernzerhof method. A plane-wave basis set was used with a kinetic energy cutoff of 500 eV. The $k$-meshes of $11 \times 11 \times 1$ for the monolayers and slabs (including 4, 5, 7, and 8 ML) and $11 \times 11 \times 7$ for the bulk CrTe$_2$ were applied. To account for the correlation effect of the transition metal element Cr and fit the experimental results, the GGA+U functional with varied $U$ values was adopted for the d-orbitals of Cr rather than SCAN and HSE functionals. The spin-orbital coupling was considered self-consistently in this work. The intrinsic anomalous Hall conductivity were evaluated based on the maximally localized Wannier functions (details see Section S9, Supporting Information) as obtained through the VASP/WANNIER90 interfaces in a non-self-consistent calculation, and calculated on $k$-meshes of $1001 \times 1001 \times 1$ for the monolayer, $601 \times 601 \times 1$ for the slabs, and $301 \times 301 \times 301$ for the bulk by the WannierTools package.
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