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## Large-Area and High-Quality 2D Transition Metal Telluride

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Distinct from most semiconducting transition-metal sulfide and selenide compounds that are only stable in the 2H phase, tellurides exhibit much richer structural variations and electronic properties, from semiconducting in the 2H phase to semimetallic in the 1T' phase. Among all telluride compounds, MoTe<sub>2</sub> crystal is stable in both 2H and 1T' phase while WTe<sub>2</sub> only form 1T' phase in nature (Figure 1a). Therefore, transition metal ditellurides, represented by MoTe<sub>2</sub> and WTe<sub>2</sub>, have attracted tremendous interests recently due to the facile switching between different phases and their unique electronic properties. For instance, large and nonsaturating magnetoresistance in WTe<sub>2</sub> bulk crystal was reported by Ali et al.,<sup>[1]</sup> presumably

due to the perfect compensation of the electrons and holes even in an ultrahigh magnetic field. A laser-induced transition between 2H and 1T' phases in MoTe<sub>2</sub> thin film has been demonstrated to show an Ohmic homojunction. [2] Furthermore, high mobility up to 4000 and 10 000 cm² V $^{-1}$  s $^{-1}$  were reported in MoTe<sub>2</sub> and WTe<sub>2</sub>, respectively. [3,4] MoTe<sub>2</sub> and WTe<sub>2</sub> atomic layers are also predicted to be promising candidates of type-II Weyl semimetals, [5,6] even in their alloys 1T' (W, Mo)Te<sub>2</sub>. [7] More importantly, monolayer tellurides such as WTe<sub>2</sub> and MoTe<sub>2</sub> are predicted to be 2D topological insulators. [8,9] These make transition metal ditellurides an excellent platform for studying fundamental physical phenomena, such as superconductivity [10] and

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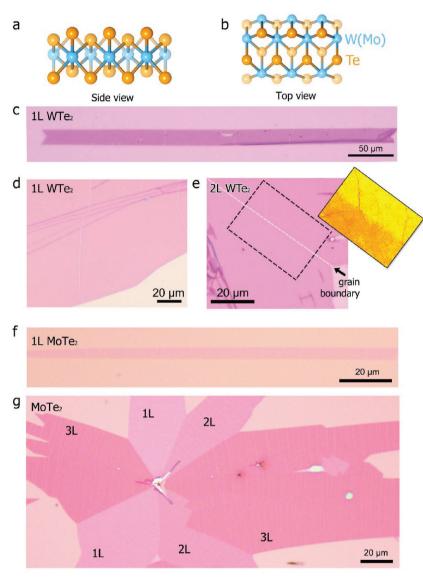
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**Figure 1.** Optical geometries of WTe $_2$  and MoTe $_2$  monolayers. a,b) Side and top views of the crystal structure of 1T' W(Mo)Te $_2$ , respectively. c) Optical image of a large single crystalline WTe $_2$  monolayer with a length of  $\approx$ 350  $\mu m$  and width of  $\approx$ 20  $\mu m$ . d) Optical image of a large WTe $_2$  monolayer film. e) Optical image of a large bilayer WTe $_2$  with grain boundary. The grain boundary is indicated by the arrow and the dashed line. Inset: false color image of the region indicated by the dashed rectangle, highlighting the location of the grain boundary. f) Optical image of a single crystalline MoTe $_2$  monolayer. With the length and width of  $\approx$ 150 and 8  $\mu m$ , respectively. g) Optical image of a MoTe $_2$  flake containing 1L, 2L, and 3L MoTe $_2$ . The number of layer can be easily identified by their contrast.

quantum spin Hall effect.<sup>[8]</sup> They are also promising candidates for various potential applications such as spintronics and high-efficiency thermoelectric devices.<sup>[11,12]</sup>

However, most of the novel physical phenomena in transition-metal ditellurides have been demonstrated in mechanically exfoliated layers, e.g., the magnetoresistance in few-layer WTe<sub>2</sub><sup>[4]</sup> and the superconductivity in bulk 1T'-MoTe<sub>2</sub><sup>[13]</sup> The exfoliation method is low-yield and time-consuming and only good for scientific research. Direct synthesis of few-layer and monolayer ditelluride would, therefore, be essential to large-scale applications. Among all the growth techniques, chemical

vapor deposition (CVD) has been demonstrated as a facile method in synthesizing monolayer crystals in large scale, including graphene,[14]  $MoS_2/WS_{2.}^{[15,16]}$ MoSe<sub>2</sub>/  $WSe_2^{[17,18]}$  and their heterojunctions, [19–21] by using different precursors under optimized reaction conditions. Although telluride compounds such as MoTe2 films were successfully synthesized by thermal flux and tellurization of molybdenum films,[3,22,23] controlled synthesis of high-quality telluride atomic layers, even down to monolayer, remains elusive under the existing CVD or physical vapor deposition conditions, mainly due to the lower environmental stability and activity of tellurium. For instance, the low chemical reaction activity of W and Mo with Te limits tellurization of W and Mo precursors (powder and oxides), although this method has been widely adopted for the preparation of sulfide and selenide monolayers. More specifically, the electronegativity difference between transitional metal (W or Mo) and Te is very small (≈0.4 or 0.3 eV), indicating a weak bonding between the metals and Te atoms which makes the stoichiometry of ditellurides difficult to be obtained. Furthermore, even though the stoichiometry of WTe<sub>2</sub> and MoTe2 is maintained, the as-synthesized product tends to decompose rapidly by emitting Te vapor at high reaction temperature (around 600 °C),[11] instead of evaporating into gas-phase telluride.

Here, we demonstrate a CVD strategy to directly synthesize WTe<sub>2</sub> and MoTe<sub>2</sub> few-layer and monolayers at large scale. The mixed compounds (weight ratio of the compound metal oxide:metal chlorides:Te is 1:1:1) and Te were used as the source of W (Mo) and Te, respectively. Such configuration of the precursors makes the reaction between Te and the metal sources react easily. The Te in the mixed compounds can decrease the melting point of the mixed compounds, while the other Te powder in the upstream was used as to keep the Te atmosphere in the whole reaction process. The corresponding chemical reaction and

more discussion about the role of the mixed compounds are shown in the Supporting Information. Raman spectroscopy was used to characterize the quality of the WTe<sub>2</sub> and MoTe<sub>2</sub> flakes to confirm their high quality. Two different stacking sequences of WTe<sub>2</sub> bilayers were observed by scanning transmission electron microscope (STEM). The as-grown WTe<sub>2</sub> atomic layers show nonsaturating magnetoresistance which resembles the features of the mechanical exfoliated ones. A novel semimetal-to-insulator transition is observed in the electrical measurements of WTe<sub>2</sub> few layers, whereas the few-layer MoTe<sub>2</sub> shows enhanced superconducting behavior.

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Side view and top view of the crystal structure of 1T' WTe2 and MoTe2 are shown in Figure 1a,b, respectively. A schematic diagram of the reaction system is shown in Figure S1(Supporting Information). The reaction temperature is between 750 and 850 °C (see experimental section). The thickness of the WTe2 and MoTe2 atomic layers can be controlled by the growth time. Single crystalline monolayer WTe2 can be consistently obtained in 5 min at 820 °C with specific flow amount of carrier gas (100 sccm Ar/15 sccm H2), while increasing the reaction time results in thick WTe2 flakes. Optical and atomic force microscopy (AFM) images of WTe2 flakes with different thicknesses are shown in Figures S3 and S4 (Supporting Information), respectively. Figure 1c shows a single-crystalline monolayer WTe<sub>2</sub> with a length of  $\approx$ 350  $\mu m$ and width of ≈20 µm. The sharp edges at the two ends of this rectangle suggest that the edge of WTe2 in 1T' phase is terminated by planes that are intersected by 50°. Figure 1d shows the optical image of a polycrystalline WTe2 monolayer film with domain size exceeding 100 µm. Figure 1e shows a bilayer WTe2 with a grain boundary at the intersection of two rectangular WTe2 domains (center of the image), as highlighted by the dashed line. For better visibility, the grain boundary with contrast enhancement and false color is shown in the inset of Figure 1e. Furthermore, large-area WTe2 films can be obtained by this method. The corresponding scanning electron microscopy (SEM) images of WTe2 film are shown in Figure S5 (Supporting Information). The size of the WTe2 film is up to 500 μm. Under similar conditions, ribbon-like monolayer 1T' MoTe<sub>2</sub> up to 150 μm (Figure 1f) and few layered 1T' MoTe<sub>2</sub> up to 200 µm (Figure 1g) are also obtained by controlling the Te source according to the phase diagram,[3] indicating that the large-area MoTe2 monolayer film can also be obtained. Unlike WTe2, different number of layers can be found in the same flake, as shown in Figure 1g, where monolayer (1L), bilayer (2L), and trilayer (3L) MoTe<sub>2</sub> are clearly distinguished by sharp contrast. AFM and SEM images of MoTe2 flakes are shown in Figures S6 and S7 (Supporting Information), respectively. It is well known that Te-based monolayers are unstable in ambient conditions. The stability test of monolayer MoTe2 and WTe2 are shown in Figure S8 (Supporting Information), which are consistent with the exfoliated ones.

Raman spectroscopy was employed to characterize the quality of the WTe2 and MoTe2 atomic layers. Figure 2a shows the Raman spectra of WTe2 films with different thicknesses ranging from monolayer to bulk. For few-layer WTe2 (<10 L), only four optical vibrational modes, namely  $B^{10}_{1}$ ,  $A^{3}_{2}$ ,  $A^{7}_{1}$ , and  $A^{9}_{1}$  were identified, compared with that reported in WTe<sub>2</sub> crystal and flakes.<sup>[24,25]</sup> Interestingly, the  $B^{10}_{1}$ mode was not reported in few-layer WTe2. Furthermore, the intensity of  $A_1^7$  peak becomes stronger than other modes as the layer number decreases, similar to the reported result that collected the Raman spectra along b axis of the mechanically exfoliated WTe2 atomic layer. [26] These results further confirm the high quality of our as-synthesized WTe2 atomic layers. The optical image and Raman intensity mapping (A<sup>9</sup><sub>1</sub>, ≈205 cm<sup>-1</sup>) of WTe<sub>2</sub> is shown in Figure 2b,c, respectively. The region for Raman mapping is highlighted in blue dashed square and the mapping size is around  $20 \times 20 \mu m$ . Raman mapping shows homogeneous intensity across the whole region, which indicates a low

defect concentration in the as-synthesized WTe2 monolayers. We also find that the grain boundary in WTe2 can be easily distinguished from the nearby regions in the Raman intensity mapping (Figure S9, Supporting Information). Monolayer and few-layer Raman spectra of MoTe<sub>2</sub> are shown in Figure 2d. The Raman peaks in monolayer were observed at 127, 161, 189, and 267 cm<sup>-1</sup>, corresponding to the Raman-active Ag modes of monolayer MoTe<sub>2</sub> in 1T' phase.<sup>[27]</sup> The spectrum agrees well with the previous reported result.[3,22,28] The Raman spectrum of 2H and 1T' MoTe2 synthesized by controlling the Te source are shown in Figure S10 (Supporting Information). Figure 2e shows a typical optical image of 1T' MoTe2 few layers. Raman intensity mapping was collected at the region highlighted by the blue dashed square. Due to the high contrast, monolayer and bilayer MoTe2 can be easily differentiated from Raman mapping (Figure 2f). In addition to Raman characterization, X-ray photoelectron spectroscopy was used to analyze the elemental distribution of WTe<sub>2</sub> and MoTe<sub>2</sub> atomic lavers. The ratio of transition metals (W or Mo) and Te was found to be very close to 1:2, in good agreement with the stoichiometry of WTe2 and MoTe<sub>2</sub>, as shown in Figure S11 (Supporting Information).

Atom-resolved STEM was applied to further investigate the atomic structure of the as-synthesized MoTe<sub>2</sub> and WTe<sub>2</sub> atomic layers. Figure 3a shows a high-resolution Z-contrast STEM image of a monolayer WTe2, revealing the 1T' phase which composes of quasi-1D tungsten-tellurium zigzag chains along the a-axis of the unit cell (highlighted by the dashed white rectangle) and connected by Te atoms in between, as indicated in the overlaid atomic structural model. The connected Te atoms can also be viewed as a Te chain parallel to the W-Te chains. The fast Fourier transformation (FFT) pattern shown in the inset further confirms the rectangular shape of the WTe2 unit cell. Figure 3b shows a line intensity profile along the b axis of the crystal, indicating two distinct positions for Te atoms bonding to the W atom in the distorted 1T phase, with a measured distance of 2.49 and 1.61 Å, respectively. The simulated STEM image using the overlaid atomic structural model achieves a good agreement with the experimental image, as shown in the inset of Figure 3a. We did not observe any intrinsic defects such as vacancies, dislocation, and grain boundaries in all the regions we examined, presumably due to the high reactivity of these defect sites which are easly oxidized. TEM characterization on thick flakes WTe2 is shown in Figure S12 (Supporting Information), further confirming the high quality of the sample.

The as-synthesized MoTe<sub>2</sub> atomic layer also maintains the 1T' phase similar to the WTe<sub>2</sub> in the monolayer form presented above, while structural difference can be observed in the few-layer flakes due to the different interlayer stacking structure, as shown in Figure S13 (Surpporting Information). The electron-energy-loss spectra (EELS) are also provided for direct comparison between the two materials in Figure S13 (Supporting Information). In light of the coexistence of different stacking orders in 2D materials, we also found that the CVD-grown 1T' WTe<sub>2</sub> maintains two different stacking sequences in the bilayer that are similar to other 2D materials. Figure 3c,d shows the atomic structures of two WTe<sub>2</sub> bilayer regions, which are distinguishable from each other. Specifically, Figure 3c shows a bilayer stacking where the second layer is mirror symmetric to



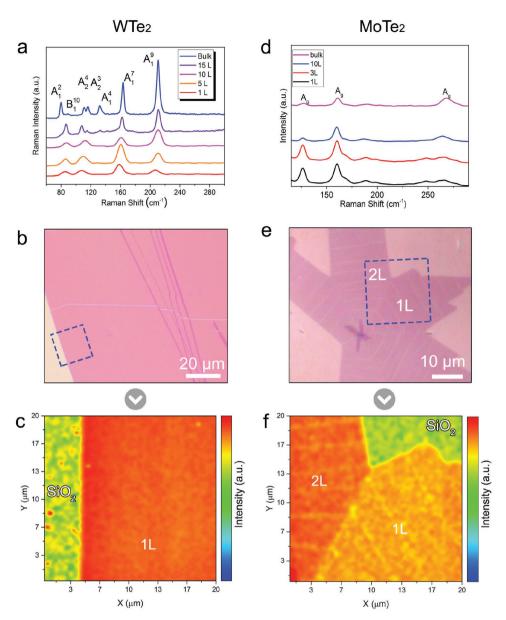
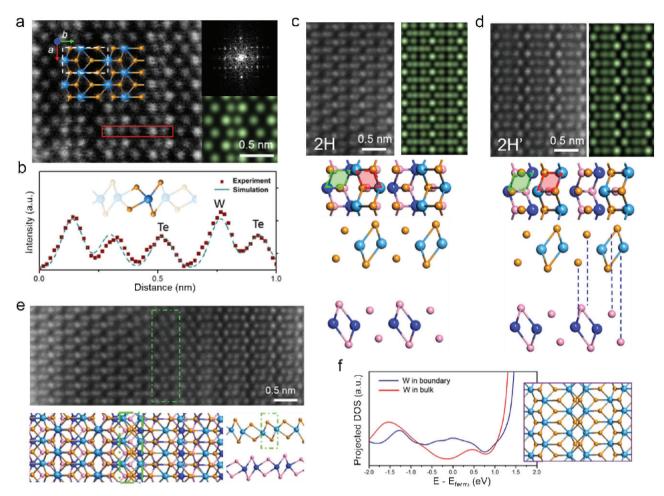


Figure 2. Raman spectra and mapping of WTe<sub>2</sub> and MoTe<sub>2</sub> monolayers. a) Thickness-dependent Raman spectra of WTe<sub>2</sub>. Seven characteristic vibration modes are identified in bulk WTe<sub>2</sub> while only 4 of them can be observed in few-layer WTe<sub>2</sub>. b) Optical image of a WTe<sub>2</sub> monolayer. c) Raman intensity mapping (from  $A^9_1$ ) from the region (highlighted by blue dashed square) in panel (b). The mapping size is  $20 \times 20 \,\mu\text{m}^2$ . The left side is SiO<sub>2</sub> while the right side shows a homogeneous WTe<sub>2</sub> film. d) Thickness-dependent Raman spectra of MoTe<sub>2</sub>. Four characteristic vibration modes are identified in bulk MoTe<sub>2</sub>. Raman intensity dramatically decreases for all Raman peaks in MoTe<sub>2</sub> thin flakes. e) Optical image of MoTe<sub>2</sub> containing monolayer and bilayer regions. f) Raman intensity mapping (A<sub>9</sub>@161 cm<sup>-1</sup>) from the region highlighted by blue dashed square in panel (e). The mapping size is  $20 \times 20 \,\mu\text{m}^2$ .

the first layer along the *b*-axis of the WTe<sub>2</sub> unit cell, as shown by the opposite orientations of the two layers in the top view of the structural model (the dashed diamonds). The Te chains of the second layer align with the W–Te zigzag chains of the first layer vertically, and vice versus, which can be visualized in the side view of the structural model. Such kind of stacking is similar to the 2H stacking in TMDs and AA' stacking in boron nitride (h-BN). Therefore, we called it 2H stacking following the convention. Figure 3d shows another stacking pattern of WTe<sub>2</sub>, where the second layer shifts half of a unit cell along the *b* axis away from the 2H stacking shown in Figure 3c, as indicated by

the structural model. We called such stacking as 2H' stacking. Both stackings form periodic stripe patterns, which consist of overlapped Te and W–Te chains from the two layers, in good agreement with the simulated STEM images (Figure S14, Supporting Information). Density functional theory (DFT) calculations on the 2H and 2H' stacking show very similar band structure, both of which maintain semimetallic properties, as shown in Figure S15 (Surpporting Information). Interlay shifting along b axis, [29] however, is rarely observed in WTe<sub>2</sub>, presumably due to the large interlayer interaction in materials composed by heavy elements. Figure 3e shows an atomically sharp stacking

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**Figure 3.** Atomic resolution STEM characterization of monolayer and bilayer WTe<sub>2</sub>. a) STEM Z-contrast image of a monolayer WTe<sub>2</sub>. The coordinate and structural model are overlaid on the image. Insets: FFT pattern and simulated STEM image of the monolayer WTe<sub>2</sub>. b) Line intensity profile file of the region highlighted by red rectangle in panel (a). c,d) STEM Z-contrast image of a bilayer WTe<sub>2</sub> with c) 2H stacking and d) 2H' stacking. The green and red dash diamonds indicate the orientation of the zigzag W-Te chains in the first and second layer, respectively. 2H and 2H' stacking is differed by half of a unit cell shifting along the *b*-aixs in the second layer. Images in green are simulated images. e) STEM Z-contrast image of an atomically sharp stacking boundary between the 2H (left) and 2H' (right) stacking. The structural model is optimized by DFT calculations. f) Projected DOS of the W atoms in the mirror twin boundary and in the bulk, showing more states near the Fermi level for W atoms in the boundary region. Inset: the structural model of the mirror twin boundary.

boundary between the 2H and 2H' stacking domains, as highlighted by the green dashed rectangle. A simulated STEM image based on the DFT-relaxed model between the two stackings is shown in Figure S15 (Surpporting Information), which resembles most feature of the stacking boundary observed experimentally. Due to the symmetry of the zigzag chains, the second layers in the two stacking form are also mirror symmetric to each other which forms a mirror twin boundary, similar to the graphene domain wall.<sup>[30]</sup> DFT calculations further indicate that the Te2 columns are misaligned in the relaxed structure of the mirror twin boundary (inset in Figure 3f), different from those observed in other TMDs.[31-33] A projected density of states (DOS) illustrates (Figure 3f) that the W atoms in the mirror twin boundary show more states at the Fermi level, suggesting that they are more metallic than the bulk counterpart, which may have important contributions in the electrical behavior to the monolayer and few-layer WTe2 and also implications on the quantum spin Hall effect in WTe2.

Layered CVD-grown ditelluride films provide an excellent platform to study the thickness-dependent electric transport. Here, the uniform and large-scaled WTe2 and MoTe2 atomic layers were selected to fabricate devices. The electrical properties of the as-synthesized WTe2 and MoTe2 flakes with different thickness were investigated by means of field- and temperaturedependent transport measurements, as shown in Figure 4. Figure 4a shows the resistivity of WTe2 as a function of temperature with thickness of 4 nm (5 layer) and 2 nm (2 layer). The corresponding optical and AFM images of the WTe2 devices are shown in Figure S16 (Surpporting Information). Above 50 K, the resistance of 4 nm WTe2 decreasing monotonically with decreasing temperature shows a metallic behavior, which is similar to the transport properties of the bulk WTe2. However, an upturn in resistance curve was observed with *T* further reduced. Such phenomenon may be due to the 2D electron-electron interactions at the reduced dimension or the degradation of atomically thin WTe2 in air.[34,35] Interestingly, the bilayer WTe2

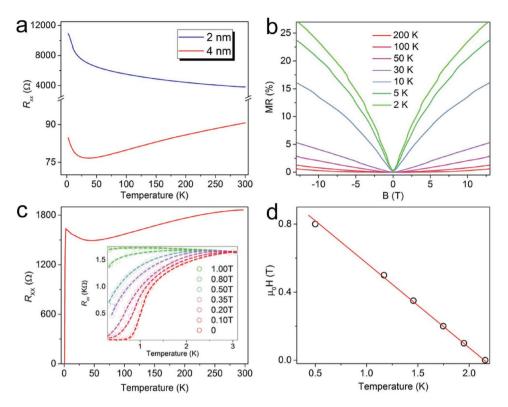


Figure 4. Transport in different thicknesses of WTe<sub>2</sub> and superconductivity in few layered MoTe<sub>2</sub>. a) Temperature-dependent resistance of WTe<sub>2</sub> flakes with a thickness of 4 and 2 nm under zero magnetic field, respectively. The corresponding field dependent magnetoresistance of 2 nm WTe<sub>2</sub> flake at different temperatures is shown in panel (b). WTe<sub>2</sub> shows a semimetal-to-insulator transition as the thickness decreased from 4 to 2 nm. c) Superconducting resistive transition of a few layered MoTe<sub>2</sub> at zero magnetic field. Inset: Superconductivity of the sample in different perpendicular magnetic fields. d) Temperature dependence of the upper critical field  $H_{c2}$ . The solid red line is the linear fit to  $H_{c2}$ .

(with thickness of 2 nm) displays solely insulating behavior under zero magnetic field, confirming that the semimetal-toinsulator transition originates from the effect of the reduced thickness. Such phenomenon has been observed in mechanically exfoliated WTe2 few layers, where the insulating state may be attributed to the Anderson localization in the 2D limit.<sup>[4]</sup> The temperature-dependent magnetoresistance (MR) calculated by MR =  $[\rho(H)-\rho(0)]/\rho(0)$  of bilayer WTe<sub>2</sub> as a function of the magnetic field is shown in Figure 4b. These results show that large and nonsaturating magnetoresistance is preserved in our CVD-grown WTe<sub>2</sub> even down to a bilayer sample, which further demonstrates their high quality. The MR reaches a maximum value of 28% at 2 K. For the thick WTe2 flakes (12 nm), the MR is about 2000% at 25 K in a field of 10 T, which is shown in Figure S17 (Surpporting Information). These values are close to the order of magnitude of the recent reports.<sup>[4]</sup>

Enhanced superconductivity is also observed in our assynthesized few-layer MoTe<sub>2</sub>. The optical image of the MoTe<sub>2</sub> device is shown in Figure S18 (Surpporting Information). Figure 4c shows the longitudinal resistance  $R_{xx}$  as a function of temperature T of few-layer MoTe<sub>2</sub> device in different perpendicular magnetic fields.  $R_{xx}$  decreases steadily from 300 to 40 K, indicating that the sample shows a metallic behavior which is consistent with previous studies on exfoliated samples.<sup>[3]</sup> With T further reduced, the sample gradually becomes superconducting below T = 2.5 K (the onset of transition) and reaches zero resistance at  $T_c = 0.5$  K. Surprisingly,

the superconductivity in thinner samples is strongly enhanced compared with  $T_c = 0.1$  K reported in its bulk counterpart.<sup>[13]</sup> The possible reason may be an enhancement of the effective electron-phonon coupling constant in thinner samples [36] The inset of Figure 4c displays the longitudinal resistance  $R_{xx}$  as a function of temperature in different perpendicular magnetic fields. We define the superconducting transition temperature  $T_c$  under different magnetic fields as the temperature at which the resistance drops to 10% of the normal state resistance  $R_N$ .  $T_c$  shifts systematically to lower temperatures with increasing magnetic fields B. Finally, the superconductivity was completely suppressed when  $B \ge 1$  T. We summarize the upper critical field  $H_{c2}$ - $T_c$  phase diagram in Figure 4d and find a linear relationship between  $H_{c2}$  and  $T_c$  near  $T_c$ . This is a characteristic of 2D superconductors and can be explained by the standard linearized Ginzburg-Landau (GL) theory

$$H_{c2}(T) = \frac{\phi_0}{2\pi\xi_{GL}(0)^2} \left(1 - \frac{T}{T_c}\right)$$
 (1)

where  $\xi_{GL}$  (0) is the zero-temperature GL in-plane coherence length and  $\phi_0$  is the magnetic flux quantum. By fitting the experimental data with the above formula, a coherence length of 38 nm was obtained, which is much larger than that in Mo<sub>2</sub>C.<sup>[37]</sup>

In summary, large-scale and atom-thin ditellurides including  $WTe_2$  and  $MoTe_2$  were synthesized. Complementary

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characterizations demonstrated the high-quality of as-grown samples. High-resolution STEM imaging resolved the atomic structure of WTe<sub>2</sub> and MoTe<sub>2</sub> and also observed the domain wall in bilayer WTe<sub>2</sub> where the stacking boundary are revealed between two distinct stacking sequences. Electric transport measurement also revealed the semimetal-to-insulator transition in WTe<sub>2</sub> and enhanced superconductivity in MoTe<sub>2</sub>. Our work will shed light on the synthesis of atom-thin telluride materials and boost the realization of quantum spin Hall devices.

## **Experimental Section**

Synthesis of MoTe<sub>2</sub> and WTe<sub>2</sub>: The WTe<sub>2</sub> (MoTe<sub>2</sub>) crystals were synthesized by CVD method using WO<sub>3</sub> (MoO<sub>3</sub>) and WCl<sub>6</sub> (MoCl<sub>5</sub>) (Sigma) as the W sources. The Te powder was used as the Te sources. The crystals were synthesized in quartz tube (1 in. diameter) with temperature from 700 to 850 °C. The system of the reaction is shown in Figure S1 (Supporting Information). Specifically, for the WTe2, the mixed gas of H<sub>2</sub>/Ar with 15 and 150 sccm was used as the carrier gas, the silicon boat containing 30 mg mixed powders with WO3:WCl6:Te = 1:1:1 (weight ratio) was put in the center of the tube. The  $SiO_2/Si$ substrate was placed downstream. Another silicon boat containing 0.5 g Te powder was put on the upstream. The temperature ramps up to the 820 °C in 17 min, and keeping at the reaction temperature for about 5 to 15 min. Then, the furnace cools down to room temperature gradually. For the MoTe<sub>2</sub>, the mixed gas of H<sub>2</sub>/Ar with 15 and 200 sccm was used as the carrier gas, the silicon boat containing 30 mg mixed powder with  $MoO_3:MoCl_5:Te = 1:1:1$  was put in the center of the tube. The  $SiO_2/$ Si substrate was placed downstream. Another silicon boat containing 0.5 g Te powders was put on the upstream. The temperature ramped up to the 780  $^{\circ}\text{C}$  in 16 min, and kept at the reaction temperature for about 5 to 15 min. Then, the furnace cooled down to room temperature gradually. Detailed description of the growth is given in the Supporting Information.

Raman Characterization: Raman measurements with an excitation laser of 532 nm were performed using a WITEC alpha 200R Confocal Raman system. Before Raman characterization, the system was calibrated with the Raman peak of Si at 520 cm<sup>-1</sup>. The laser powers are less than 1 mW to avoid overheating of the samples.

TEM and STEM Characterization: The STEM samples were prepared with a poly(methyl methacrylate) (PMMA) assisted method. A layer of PMMA of about 1 µm thick was spin-coated on the wafer with WTe<sub>2</sub> (MoTe2) samples deposited, and then baked at 180  $^{\circ}\text{C}$  for 3 min. Afterward, the wafer was immersed in NaOH solution (1 M) to etch the SiO<sub>2</sub> layer overnight. After lift-off, the PMMA/WTe<sub>2</sub> (MoTe<sub>2</sub>) film was transferred into DI water for several cycles to wash away the residual contaminants, and then it was fished by a TEM grid (Quantifoil Mo grid). The transferred specimen was dried naturally in ambient environment, and then dropped into acetone overnight to wash away the PMMA coating layers. The STEM imaging shown in the main text was performed on an aberration-corrected Nion UltraSTEM-100 operating at 100 kV. The convergence semiangle for the incident probe was 31 mrad. Z-contrast images were collected for a half-angle range of ≈86–200 mrad. STEM imaging and EELS analysis on MoTe2 shown in the Supporting Information were performed on a JEOL 2100F with a cold field-emission gun and an aberration corrector (the DELTA-corrector) operating at 60 kV. A Gatan GIF Quantum was used for recording the EELS spectra. The inner and outer collection angles for the STEM image ( $\beta$ 1 and  $\beta$ 2) were 62 and 129–140 mrad, respectively, with a convergence semiangle of 35 mrad. The beam current was about 15 pA for the ADF imaging and EELS chemical analyses. All imaging was performed at room temperature.

Devices Fabrication and Transport Measurement: The Hall bars were patterned on few layer MoTe<sub>2</sub> and WTe<sub>2</sub> using e-beam lithography.

The Ti/Au (5/50 nm) electrodes were deposited using the thermal evaporator, followed by the lift off process. For the measurements of WTe<sub>2</sub>, the transport measurement was performed in the Quantum Design PPMS system with temperature ranging from 300 to 2 K and magnetic field up to 14 T. For the superconductivity measurements of MoTe<sub>2</sub>, the transport experiment was carried out in a top-loading Helium-3 cryostat in a superconducting magnet. An AC probe current  $I_{\rm AC}=10$  nA at 30.9 Hz was applied from the source to the drain. Then, a lock-in amplifier monitored the longitudinal  $R_{\rm xx}$  through two additional electrical contacts.

## **Supporting Information**

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

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