Obtaining high-quality, large-size 2D vdW single crystals is an important prerequisite for realizing the potential of highly integrated 2D devices. Here, Yang et al. report a chloride-mediated CVT approach to achieve ultrahigh-yield and large-scale fast growth of centimeter-sized transition-metal telluride single crystals with extremely high quality.
Ultrahigh yield and large-scale fast growth of large-size high-quality van der Waals transition-metal telluride single crystals

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SUMMARY

Stable, rapid, and large-scale production of high-quality, large-size two-dimensional (2D) van der Waals single crystals is an important prerequisite for realizing the potential of highly integrated 2D devices. Here, we report a chloride-mediated chemical vapor transport (CVT) approach to achieve large-scale production of MoTe2 and WTe2 single crystals with lateral sizes up to 2 cm and extremely high quality. The largest magnetoresistance and carrier mobility can reach 170% and 1,390.09 cm2 V-1 s-1 for 2H-MoTe2 crystals and 506,017% and 11,952.10 cm2 V-1 s-1 for Td-WTe2 crystals, respectively, and all are among the best reported ones. Compared with the conventional CVT method, which has a long growth cycle and much reactive reagent surplus, the growth speed of the current method is about 73 to 9,144 times faster, with ultrahigh yields of nearly 100%. The high crystallinity guarantees the crystals can be easily exfoliated to large-scale 2D crystals down to monolayers for assembling 2D high-performance devices.

INTRODUCTION

Two-dimensional (2D) van der Waals (vdW) transition-metal dichalcogenides (TMDCs) have aroused tremendous research interests in recent years as one type of promising materials to break Moore’s law and replace silicon, owing to their unique structures and novel physical properties.1–7 Compared with traditional bulk materials, TMDCs have the inherent vdW layered structures,8–11 which allow for the production of possible next-generation high speed, low power consumption, and flexible integrated multifunctional devices. However, although TMDCs have been investigated for over 10 years, they are still rare commercial products currently. One of the most critical limiting factors is the lack of large-scale production of high-quality 2D TMDC single-crystalline source materials. Several growth methods, including chemical-vapor deposition (CVD), molecular-beam epitaxy (MBE), and chemical-vapor transport (CVT), have been adopted to prepare 2D TMDC single crystals. CVD growth is feasible but often suffers from poor crystallization quality and low repeatability.12 Meanwhile, MBE growth is highly dependent on the surface properties of the substrates and thus is not suitable for scalable production due to its extremely time-consuming nature.13 Compared with the above two methods, the CVT method is highly reliable and scalable due to less influence of process conditions, so it is often used to synthesize high-quality layered TMDC single crystals. However, the yield is generally low and can only be used in the research field.14 So far, the CVT method has been used to synthesize various TMDC single-crystal materials, such as MoS2, MoSe2, MoTe2, WSe2, WTe2, VTe2, TaTe2, TiTe2, ZrTe2, etc.
Although the 2D TMDCs can be scalably produced by liquid-phase exfoliation of CVT crystals, they generally suffer from small size and poor quality.\textsuperscript{15} Recently, large-area exfoliation and assembly techniques have been reported in single-crystal TMDCs\textsuperscript{16–20} which allow for a top-down device fabrication process similar to single-crystal silicon. Nevertheless, the large-scale production of high-quality, large-size TMDC bulk crystals still remains a great challenge. Therefore, it is of great significance to develop a simple and feasible method for the large-scale growth of high-quality TMDC single crystals.

Transition-metal tellurides (TMTs) such as MoTe\textsubscript{2} and WTe\textsubscript{2} have attracted much attention in recent years owing to their ultrahigh mobility, suitable band gap, or topological states.\textsuperscript{21–30} Since tellurium has poorer oxidizing capability compared with sulfur and selenium, it is more difficult to grow high-quality TMT single crystals on the large scale within the TMDC family. Generally, the conventional CVT method is to seal the source material and transport agent in an elongated TMT single crystals on the large scale within the TMDC family. Generally, the conventional CVT method is to seal the source material and transport agent in an elongated quartz tube, and the carrier gas transports the source material to the crystalline region by diffusion without an external driving force, resulting in a slow transport rate. Thus, the conventional CVT method for growing TMTs usually has an extremely long reaction time of up to several weeks.\textsuperscript{22,31} Moreover, no investigation about the large-scale CVT growth of TMTs related to production level and yield has been reported to date.

Here, we demonstrate large-scale production of high-quality, centimeter-sized TMT single crystals (MoTe\textsubscript{2}, WTe\textsubscript{2}) through a chloride-mediated CVT method with strong convective driving force. The grown TMT single crystals exhibit extremely high crystalline quality and good electrical transport properties, which allows the large-area exfoliation and excellent device performance. Moreover, this method could also be generalized to large-scale production of other TMTs like NbTe\textsubscript{2} and TaTe\textsubscript{2} single crystals. This high-efficiency, high-quality, reproducible, energy-saving, low-pollution, and low-cost crystal-preparation approach paves the way for large-scale applications in next-generation 2D electronics, optoelectronics, and spintronics devices.

RESULTS AND DISCUSSION

**Growth and phase characterizations of the MoTe\textsubscript{2} and WTe\textsubscript{2} single crystals**

Bulk MoTe\textsubscript{2} and WTe\textsubscript{2} single crystals were grown by the chloride-mediated CVT method, in which the highly reactive transition metal chloride was used as the transport agent, and the transport-agent flow resistance was reduced by the increased diameter of the quartz ampoule to provide a stable driving force for the CVT process. For a typical growth process, 0.1 mol high-purity transition metal (Mo or W) powder (Aladdin, purity 99.99%), 0.2 mol Te granule (Aladdin, purity 99.99%), and approximately 0.2–1 g of transition metal chloride (MoCl\textsubscript{5} or WCl\textsubscript{6}, Aladdin, the purity is 99.6 and 99.9%, respectively) used as transport agent were put into a vacuum-sealed quartz ampoule with an inner diameter of 4 cm and a length of 30 cm. Then, the ampoule with chemicals was placed horizontally into a two-zone furnace with a furnace chamber diameter of 60 mm, where the temperatures of source and growth zones were heated to 900\,°C and 700\,°C within 1 h, respectively. After 2 (for 1 g transport agent) to 7 days (for 0.2 g transport agent) of growth, the quartz ampoule was directly cooled down to room temperature by furnace cooling. Figure 1A shows the in situ optical images of as-synthesized MoTe\textsubscript{2} and WTe\textsubscript{2} after 2 days of CVT growth, revealing that almost no residual source material remained at the high-temperature end of the ampoule wall, while the low-temperature end was found to be covered with the target crystals. For MoTe\textsubscript{2} growth, the source materials used were Mo 9.59 g and Te 25.52 g, the mass of the as-grown MoTe\textsubscript{2} single crystals was 35.10 g, and the yield was about 99.97% with a transport rate of 731.25 mg/h. For WTe\textsubscript{2}
growth, the source materials used were W 18.38 g and Te 25.52 g, the mass of the grown WTe₂ single crystals was 43.89 g, and the yield was about 99.98% with a transport rate of 914.375 mg/h. The transport rates of our chloride-mediated CVT method were 73 to 9,144 times faster when compared with the conventional CVT growth, with a lower transport rate from 0.1 to 10 mg/h. The production yield of MoTe₂ and WTe₂ is nearly close to 100%, which avoids wasting of source materials. We noted that the remaining translucent transport-agent crystals were generally deposited upstream of the target crystal and were removed during the subsequent single-crystal collection. Even though the transport rate of this large-scale CVT growth is very fast, the as-obtained crystal size is very large, as shown in Figures 1B and 1C. The lateral size of the as-grown MoTe₂ and WTe₂ single crystals can reach 1.7 and 2.7 cm, respectively. It should be noted that most of the WTe₂ crystals were grown with a long-ribbon-like shape along the W-W chains (a axis).

Significantly, such a large-scale CVT method with high transport rate and high production yield benefits from the efficient carrying capacity and the fast flow rate of the transport agent. Unlike CVD growth, the gas flow during the CVT growth is not driven externally, resulting in a slow transport rate. To solve this problem, we use the highly reactive transport agent and increase the tube diameter in the vertical direction, which can ensure that after the transport agent reacts in the crystallization zone, the generated lighter gas product can move higher by convection and gravity, thereby providing a driving force for the gas flow during the CVT process. Meanwhile, the collision between the transport agent and the generated gas product is avoided, forming a high-speed transport-agent circulation loop. For a typical growth process, in the high-temperature zone, M powders react with Cl₂ to form MClₓ, while in the low-temperature zone, MClₓ (M = Mo, W; x = 5, 6) reacts with Te to form MTe₂ and Cl₂. MClₓ (M_{mol} = 272.9/396.2 for MoCl₅/WCl₆) is heavy, while Cl₂...
(M_{mol} = 70.8) is light, so Cl$_2$ flows from the low-temperature zone to the high-temperature zone at the head of the quartz ampoule, while MCl$_x$ flows from the high-temperature zone to the low-temperature zone at the end of the tube, thus forming an efficient circulation transport process (Figure S1).

To characterize the quality and structural properties of the large-scale CVT-grown TMT crystals, X-ray diffraction (XRD) and Raman spectroscopy (Raman) were performed. Figure 1D exhibits the XRD patterns of the grown MoTe$_2$ and WTe$_2$ single crystals, and the corresponding standard diffraction peak patterns are shown below for reference. Obviously, only four equally spaced diffraction peaks in the range of 10° to 65° could be identified for both MoTe$_2$ and WTe$_2$ and are in good agreement with the standard hexagonal 2H-MoTe$_2$ (PDF#73-1650, data from the Powder Diffraction File and same as follows) and orthorhombic Td-WTe$_2$ (PDF#71-2156) structures, respectively. All peaks can be indexed as (0 0 L) reflections, indicating that the as-grown MoTe$_2$ and WTe$_2$ single crystals are in good orientation and that the exposed surface belongs to the ab plane. Additionally, sharp (0 0 L) peaks confirm the extremely high crystallinity of the chloride-mediated CVT-grown MoTe$_2$ and WTe$_2$ single crystals with the full width at half maximum (FWHM) of about 0.0335 and 0.0323° for the (0 0 8) peak, respectively. Next, Figure 1E shows the Raman spectra of the as-grown MoTe$_2$ and WTe$_2$ single crystals with the excitation at a wavelength of 532 nm. For MoTe$_2$, only two optical vibrational modes labeled as $A_{1g}$ (~173.06 cm$^{-1}$) and $E_{12g}$ (~233.01 cm$^{-1}$) are identified, consistent with that reported in the 2H-MoTe$_2$ crystal. As for WTe$_2$, five characteristic Raman peaks are observed at ~111.27, ~116.39, ~132.79, ~163.01, and ~210.14 cm$^{-1}$, corresponding to the $A_{42}$, $A_{31}$, $A_{41}$, $A_{71}$, and $A_{91}$ modes of Td-WTe$_2$, respectively. Therefore, synthesis of both 2H-MoTe$_2$ and Td-WTe$_2$ single crystals with good crystalline quality was confirmed by combining both the XRD and Raman characterizations.

### Structural characterizations of the MoTe$_2$ and WTe$_2$ single crystals

To further examine the quality of this large-scale CVT-grown TMT crystals, large-scale high-resolution transmission electron microscopy (HRTEM) analyses were conducted. Figures 2A and 2D show the HRTEM images of the few-layer MoTe$_2$ and WTe$_2$ obtained by ultrasonication of the original as-grown bulk crystals, respectively. Even if the imaging range of up to 4,756 nm$^2$ (~39,110 unit cells) for MoTe$_2$ and 2,922 nm$^2$ (~13,315 unit cells) for WTe$_2$, no obvious crystal defects were observed, indicating the high growth quality. Figures 2B and 2E show the magnified HRTEM images within the marked regions of Figures 2A and 2D, respectively, showing a nearly perfect periodic atomic arrangement like most ideal crystals. The marked interplane spacing in Figures 2B and 2E was determined to be ~0.302 and ~0.627 nm, respectively, corresponding to the (100) plane 2H-MoTe$_2$ hexagonal structure and the (100) plane Td-WTe$_2$ orthorhombic structure. The corresponding selected-area electron diffraction (SAED) patterns in Figures 2C and 2F show only a single array of diffraction points, strongly suggesting the high crystalline quality of the large-scale CVT-grown MoTe$_2$ and WTe$_2$ crystals.

Moreover, in order to identify the purity of the as-grown MoTe$_2$ and WTe$_2$ crystals and verify the uniform distribution of their components, energy-dispersive X-ray spectroscopy (EDAX) analysis was performed, as shown in Figure S2. The homogeneous distribution of metal (Mo or W) and tellurium (Te) throughout the entire investigated region indicates the absence of phase separation. Except for the Cu element from the TEM copper grids, only Mo, W, and Te as the constituent elements were detected (Figures S2C and S2D), and the ratio of transition metals (Mo or W) to Te were identified to be very close to 1:2, which is in good agreement with the stoichiometric ratio of MoTe$_2$ and WTe$_2$. 
Magnetoelectrical transport properties of MoTe$_2$ single crystals

Significantly, such large-scale CVT-grown TMT crystals not only have good crystalline quality but also show excellent electrical transport performance. Figure 3A and its inset show the electrical resistivity ($\rho_{xx}$) curve of the as-grown MoTe$_2$ crystal in the temperature range of 22 to 300 K, where two temperature-dependent regimes of resistivity could be clearly observed with a resistivity minimum at $\approx 127$ K, consistent with previous reports.\textsuperscript{42,47} We found that the resistivity increases sharply with the decrease in temperature when the temperature drops below 127 K, which is a typical feature of an $n$-type semiconductor in the freeze-out T region. While the resistivity slowly increases with increasing temperature when the temperature is between 127 and 300 K, which is a typical feature of an $n$-type semiconductor in the extrinsic T region.\textsuperscript{48} Figures 3B and 3C show the temperature-dependent Hall resistivity ($\rho_{xy}$) curves of the as-grown MoTe$_2$ crystal, and the linear $\rho_{xy}$ with a negative slope under the entire magnetic field range (-15 to 15 T) at various temperatures (50, 100, 200, and 300 K) were confirmed, which further indicates the $n$-type conductivity of the MoTe$_2$ crystal. The rapid increased slope was also observed with the decreased temperature, which is reasonable due to the rapid decrease of the thermally excited carrier concentration with the decreased temperature. Figure 3D and its inset show the magnetoresistance (MR) of the as-grown MoTe$_2$ crystal at different temperatures. We found that although the room-temperature (300 K) MR of MoTe$_2$ is less than 1% under a 14 T magnetic field, the MR rapidly increases to about 170% when the temperature is reduced to 30 K, which is the highest among the reported semiconducting 2H-phase MoTe$_2$. Detailed electrical transport characteristic parameters of the as-grown MoTe$_2$ crystals at different temperatures, including MR, Hall mobility ($\mu_H$), carrier density ($n$), and conductivity ($\sigma$), which were derived from its Hall-effect measurement, are further summarized in Figure 3E and Table S1. Clearly, the MR and $\mu_H$ decrease monotonically with increasing temperature and can reach 52%...
and 1,390.09 cm² V⁻¹ s⁻¹ at 50 K, respectively, and decrease to 0.6% and 48.73 cm² V⁻¹ s⁻¹ at room temperature (300 K), but these are still better than previous reports for the 2H-MoTe₂ crystals. Moreover, the electron density increases monotonically with the increase of temperature, which is caused by the increase of electrons thermally excited into the conduction band.

Additionally, further angle-dependent Hall measurements on the same MoTe₂ sample were conducted, as shown in Figure S3A. The inset angle θ represents the angle between the normal direction of the sample and the applied magnetic field (H), which is also applicable in the following discussion about WTe₂. Notably, the Hall resistivity of the MoTe₂ sample follows the standard cosine law with the angle at different temperatures. Meanwhile, the corresponding angle dependence of magnetoresistivity was also performed in a magnetic field of up to 14 T (Figure S4), which shows an approximate cos(2θ) relationship at a low temperature of 50 K. However, when the temperature rises to 100 K and above, the MR exhibits a strong asymmetry when the applied magnetic field is parallel or anti-parallel to the direction of the sample current (I), which has never been revealed before.

**Magnetoelectrical transport properties of WTe₂ single crystals**

Unlike MoTe₂, the resistivity of the as-grown WTe₂ crystal increases monotonously with increasing temperature over the whole temperature range, indicating its metallic transport nature (Figure 4A). Figure 4B and its inset further show its temperature-dependent Hall resistivity (ρₓᵧ) curves. Clearly, the Hall resistivity as a function of magnetic field shows a linear behavior at high temperatures (200 and 300 K), which implies that the as-grown WTe₂ crystal is an electron-dominant n-type conduction. Meanwhile, the Hall resistivity shows a negative slope as the temperature decreases, together with the gradual

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**Figure 3. Magnetoelectrical transport properties of the MoTe₂ single crystals**

(A) Temperature-dependent electrical resistivity (ρₓₓ) of the MoTe₂ crystal. Inset shows the change in ρₓₓ at a relatively high temperature regime (above 100 K). (B and C) Field dependence of Hall resistivity (ρₓᵧ) for the MoTe₂ crystal at different temperatures. (D) Magnetoresistance (MR) for the MoTe₂ crystal at 30 and 50 K. The inset shows a clear view at higher temperatures. The applied field is parallel to the c axis during the tests. (E) Dependence of MR, Hall mobility (μᵧ), carrier density (n), and conductivity (σ) as function of temperature derived from the Hall effect measurement on the MoTe₂ crystal. Error bars, SD, N = 25.
emergence of nonlinear behavior and Shubnikov-de Haas (SdH) quantum oscillations, which can be attributed to the existence of multiple Fermi pockets and the hole-electron carrier concentration compensation.\(51,52\) Figure 4C and its inset show the MR of the as-grown WTe\(_2\) crystal with the applied magnetic field parallel to the c axis of the sample measured at different temperatures. Although the room-temperature (300 K) MR of WTe\(_2\) is only 2.31\%, a significant unsaturated large MR (506,017\%) can be observed at 2 K under the high magnetic field \(B = 13.45\) T, which is stronger than most of values reported for the WTe\(_2\) crystals.\(53–55\) The pronounced SdH quantum oscillations at 2 K (Figure 4D) are clearly confirmed from the second-order derivatives \((d^2R/dB^2)\) of the data in Figure 4C. Such observations indicate the high quality of these large-scale CVT-grown WTe\(_2\) crystals.\(52\) Similar to MoTe\(_2\) crystals, we also summarize the transport characteristic parameters of the grown WTe\(_2\) crystals at different temperatures, as shown in Figure 4E and Table S1. Both the MR and electron mobility of WTe\(_2\) decrease with the increasing temperature, with a maximum value of 506,017\% and 11,952.10 cm\(^2\) V\(^{-1}\) s\(^{-1}\) at 2 K, respectively, which are higher than most of the reported values.\(53–55\) Since the carrier density exhibits inconspicuous change with temperature, the trend of the conductivity \((\sigma = \mu_H \cdot n)\) is basically the same as the mobility. Moreover, the reliability of the Hall signal of the WTe\(_2\) crystals was further verified through the angle-dependent Hall measurements. As shown in Figure S3B, the Hall resistivity as a function of angle also exhibits the standard cosine law at high temperatures (200 and 300 K) without quantum effects. Similar distortion and steps appear for the angle-dependent Hall resistivity below 100 K, which is attributed to the appearance of nonlinear Hall effect and SdH quantum oscillations. Figures S5 and S6 show the dependence of the magnetoresistivity on the angle between the applied field and the c axis of the same WTe\(_2\) sample at different temperatures. Apparently, the maximum and minimum MRs were observed with the condition of H//c axis and H \(\perp\) c axis, respectively. Some vibrations and steps due to SdH quantum oscillations can be observed.
oscillations were also observed at 2 K (Figure S5A). Such an SdH quantum oscillation becomes insignificant or disappears when the temperature is above 10 K, and the angle-dependent MR exhibits an approximate $\cos(2\theta)$ form.

Growth and phase characterizations of TaTe$_2$ and NbTe$_2$ single crystals
Significantly, we find that this large-scale CVT method is also applicable to other TMT single crystals such as TaTe$_2$ and NbTe$_2$. To illustrate it, we present the optical images of the as-grown TaTe$_2$ and NbTe$_2$ single crystals in Figures S7A and S7B, both of which exhibit sizes of centimeter level. The structures of TaTe$_2$ and NbTe$_2$ single crystals were further investigated using Raman, as shown in Figure S7C. Clear characteristic Raman peaks can be identified and are consistent with those of the reported TaTe$_2$ and NbTe$_2$ crystals.$^{56,57}$ Figure S7D shows their XRD patterns, which are consistent with the stand monoclinic TaTe$_2$ (PDF#21-1201) and monoclinic NbTe$_2$ (PDF#71-2196), respectively. Also, the sharp (0 0 L) peaks further confirm their high crystallinity quality with a FWHM of $\sim 0.0362^\circ$ and 0.0431$^\circ$ for the (0 0 2) peak, respectively.

Device applications of MoTe$_2$ and WTe$_2$ single crystals
Benefitting from the high crystallinity and good vdW layered structure of the as-synthesized TMT crystals, it is easy to exfoliate it down to a few layers to fabricate micro-nano devices. As is typically representative of semiconductor and Weyl semimetal in TMTs, MoTe$_2$ and WTe$_2$ are often employed to fabricate electrical devices such as transistors and micro-nano Hall elements, respectively. Figures 5A and 5B show the transfer characteristic of a typical 14-layer MoTe$_2$ transistor ($\sim 10.81$ nm thickness) under different drain-source voltage ($V_{ds}$) values on the linear and logarithmic...
We note that the leakage current of the 14-layer MoTe$_2$ transistor was on the order of tens of pico-amperes as the back-gate voltage ($V_{bg}$) was swept from -5 to 30 V (Figure S8). The corresponding optical image of the device is shown in the inset of Figure 5A. In contrast to previous reports, our as-fabricated MoTe$_2$ transistor shows a unipolar n-type response. Notably, the maximum current on/off ratio can reach $3.78 \times 10^3$, and the field-effect mobility can be derived as $10.86$ cm$^2$ V$^{-1}$ s$^{-1}$ under modulation of $V_{bg}$, which is comparable or even better than that of previously reported exfoliated 2H-MoTe$_2$ nanosheets. Figures 5C and 5D show the output characteristic of the MoTe$_2$ transistor at different $V_{bg}$ values in the positive and negative $V_{ds}$ regimes, respectively. As $V_{bg}$ increases linearly, the value of drain-source current ($I_{ds}$) increases exponentially, indicating the good modulation of back-gate voltage.

Furthermore, the temperature-dependent electrical resistivity ($\rho_{xx}$) of a typical 4-layer WTe$_2$ (~3.07 nm thickness) is determined by the measurement with the Hall bar configuration (Figure 6A). As shown in Figure 6B, the $\rho_{xx}$ decreases monotonically with the decreasing temperature from 300 to 2 K, exhibiting metallic behavior. Figures 6C and 6D show the corresponding Hall resistance ($R_{xy}$) and MR curves at different temperatures. Notably, the $R_{xy}$ shows a very smooth linear relationship with the variance of the applied magnetic field up to 5 T (Figure 6C). The sensitivities at 2 and 300 K are 5.4 and 2.1 $\mu$V/T, respectively, which make up the shortcoming that the working range and sensitivity of traditional Hall elements cannot be increased simultaneously. However, similar to earlier reports of the WTe$_2$, the giant MR effect of the as-exfoliated WTe$_2$ nanosheet is also suppressed, and the largest MR at 2 K is only about 5.17%.
Large-area exfoliation of MoTe$_2$ and WTe$_2$ single crystals

Ultimately, based on the high-yield, rapid, large-scale CVT-grown large-size TMTs, large-area exfoliations are available as shown in Figure 7 (more detail is in the experimental procedures). Notably, the as-grown MoTe$_2$ and WTe$_2$ bulk single crystals can be easily exfoliated to monolayers with lateral sizes reaching the level of millimeter scale (Figures 7A and 7B). For MoTe$_2$, two different color contrasts can be clearly identified within the same sheet (Figure 7A), and the corresponding Raman spectra collected at randomly distributed locations are shown in Figure 7C. The presence of the stronger $B_{1g}$ peak at $\sim$290.51 cm$^{-1}$ for the small-area MoTe$_2$ indicates its atomically thin thickness (Figure 7C, top panel), while the absence of two peaks of $E_{1g}$ ($\sim$118.72 cm$^{-1}$) and $B_{1g}$ indicates that the large-area MoTe$_2$ is monolayer (Figure 7C, bottom panel).$^{40,71}$ Also, the $E_{2g}$ and $A_{1g}$ modes show small negative and positive shifts as the sample thickness increases, respectively, similar to previous reports.$^{40}$ Moreover, the height profiles measured by atomic force microscopy (AFM) further reveal that the thickness of the exfoliated few-layer MoTe$_2$ with two different color contrasts is about 0.76 and 1.56 nm (Figure 7A, insets), respectively, which can be confirmed as monolayer and bilayer by its interlayer spacing of about 0.7 nm.$^{58,59}$ Similarly, a large-area WTe$_2$ monolayer can also be obtained (Figure 7B). As shown in Figure 7D, the corresponding Raman spectra of 30 randomly selected points on the sample are very similar, and all of the peak positions are consistent with those reported from exfoliated few-layer WTe$_2$.$^{24,72}$ In addition, the height profile with a step of approximately 0.83 nm illustrated in Figure 7D further confirms that it is a monolayer WTe$_2$. Therefore, the successful exfoliation of large-area few- and monolayer MoTe$_2$ and WTe$_2$ further confirms the large crystal domain and high crystalline quality of the large-scale chloride-mediated CVT-grown MoTe$_2$ and WTe$_2$ crystals and benefits device process such as photolithography. Although the scalable exfoliation of large-area, few-layer TMDCs still faces great challenges, there is no doubt that large single crystals make it easier. Our work provides a simple and feasible approach for the preparation of 2D electronics, optoelectronics, and spintronics devices.

Our work demonstrates a high-yield, large-scale, fast chloride-mediated CVT method to prepare centimeter-scale TMTs single crystals of extremely high quality represented by MoTe$_2$ and WTe$_2$, by both using highly reactive transition metal chloride as the transport agent and accelerating the circulation rate of the transport gas with the increase of the quartz ampoule diameter. The transport rate can reach 914.375 mg/h, and the yield is nearly 100%. Magnetoelastic transport studies prove the high quality of the as-grown crystals with ultrahigh carrier mobility, pronounced SdH quantum oscillations, and enormous unsaturated MR. On this basis, transistors and Hall element devices based on mechanically exfoliated atomically thin MoTe$_2$ and WTe$_2$ few layers are fabricated and exhibit excellent performance. Furthermore, MoTe$_2$ and WTe$_2$ monolayers with typical lateral dimensions of millimeter scale are easily obtained. This large-scale CVT-grown method has the advantages of high efficiency, high quality, reproducibility, energy saving, low pollution, and low cost, which will open up exciting opportunities for the large-scale production of TMDC single crystals for potential industrial applications of vdW electronics.

EXPERIMENTAL PROCEDURES

Resource availability

Lead contact

Further information and requests for resources and reagents should be directed to and will be fulfilled by the lead contact, Haixin Chang (hxchang@hust.edu.cn).
**Materials availability**

This study did not generate new unique reagents.

**Data and code availability**

The experimental data generated in this study are available from the lead contact upon request.

**Crystal characterizations and measurements**

Raman with 532 nm laser as the excitation source (Raman, LabRAM HR800, Horiba JobinYvon), XRD (SmartLab SE, Rigaku) with Cu Kα radiation (wavelength = 0.154 nm),
and field-emission TEM (FTEM, Tecnai G2 F30, FEI) were used to characterize the crystal phase and structure of the MoTe$_2$ and WTe$_2$ single crystals to confirm their high quality. AFM (SPM9700, Shimadzu) was used to determine the thickness of the exfoliated MoTe$_2$ and WTe$_2$ flakes. TEM samples were prepared by sonicating the bulk samples in ethanol and dropping the solution on TEM copper grids. The samples were dried naturally in vacuum conditions. The electrical and magnetic transport properties were measured in a Physical Property Measurement System (PPMS; DynaCool, Quantum Design) with a five-probe configuration using silver electrodes. Note that the MR is defined as $\frac{\rho_{xx}(H) - \rho_{xx}(0)}{\rho_{xx}(0)} \times 100\%$, and $\rho_{xx}(H)$ is the resistivity under an applied magnetic field $H$. The Hall mobility $\mu_H$ is defined as $\frac{R_H}{\rho_{xx}}$, where $R_H$ is the Hall coefficient ($R_H = \frac{\rho_{xy}}{B}$), and $\sigma$ is the conductivity ($\sigma = \frac{1}{\rho} \frac{1}{\rho_{xx}}$).

Electrical-device fabrication
For MoTe$_2$ transistor fabrication, few-layer MoTe$_2$ were mechanically exfoliated and transferred onto the heavily p-doped Si wafers covered with a 500 nm thick dielectric layer of SiO$_2$. The electrical contacts were patterned on top of the selected nano-sheet using standard lithography, followed by the electron-beam evaporation of a Cr/Au film (10/100 nm thickness). Electrical measurement of the MoTe$_2$ transistors was carried out by Keithley 4200 Semiconductor Characterization System (4200-SCS) Parameter Analyzer. To enhance the conductivity of the bottom gate, the natural oxidized SiO$_2$ on the back of the silicon wafer was scratched with a diamond pen, and an indium electrode was prepared by the indium pressing method as the bottom gate of the device. Note that the field-effect mobility $\mu_{FE}$ is defined as $\frac{L_w}{L_w \varepsilon_0 \varepsilon_r V_d} \frac{\Delta \varepsilon_r V_d}{\Delta V_{bg}}$, where $L_w$ is the channel length-to-width ratio, $\varepsilon_0$ is the permittivity in vacuum, $\varepsilon_r$ is 3.9 for SiO$_2$, and $d$ is the thickness of the dielectric layer.

For WTe$_2$ Hall element devices, few-layer WTe$_2$ with different thickness were mechanically exfoliated onto 500 nm SiO$_2$/Si substrate, and then Cr/Au (10 nm/100 nm) Hall bar electrodes were fabricated by standard lithography and electron-beam evaporation. The electrical and magnetic transport properties of the layered WTe$_2$ flakes were measured in a 14 T Physical Property Measurement System (PPMS, DynaCool, Quantum Design), which has a base temperature of 2 K. AFM was used to measure the thicknesses of MoTe$_2$ and WTe$_2$ samples.

Large-area crystal exfoliation
First, electron-beam evaporation of about 100 nm thick Au on the TMT bulk crystals freshly cleaved by scotch tape was performed. Since Au has a strong affinity for chalcogens, the as-evaporated Au atoms will bond with the tellurium atoms of the topmost layer of TMTs, thereby realizing large-area, single-layer exfoliation of the large-size TMT bulk crystals. Next, the top-most layer of the TMT bulk crystals can be peeled off using a thermal release tape (TRT) and stuck onto a 500 nm thick SiO$_2$/Si substrate. By heating on the hot plate ($\approx 100{^\circ}$C), TRT was released and left the ultrathin TMT crystal on the substrate. Then, the substrate was soaked in ethyl acetate to remove tape residue. The gold film was wet etched with potassium iodide and iodine (KI/I$_2$) (Gold Etch-Type TFA, Transene) without affecting the quality of the TMT flakes. Finally, the SiO$_2$/Si substrate with ultrathin TMT crystals was rinsed with deionized water repeatedly and dried under vacuum conditions.

SUPPLEMENTAL INFORMATION
Supplemental information can be found online at https://doi.org/10.1016/j.xcrp.2022.100953.
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AUTHOR CONTRIBUTIONS
H.C. supervised the project. L.Y. and H.W. designed the experiments. L.Y., H.W., G.Z., W.J., and L.L. conducted the experiments. L.Y. and H.W. wrote the manuscript. H.C. and W.Z. revised the manuscript. All authors contributed to the results analysis and discussions and approved the final version of the manuscript.

DECLARATION OF INTERESTS
The authors declare no competing interests.

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