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MoTe₂ Field-Effect Transistors with Low Contact Resistance through Phase Tuning by Laser Irradiation

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Abstract: Due to their extraordinary electrical and physical properties, two-dimensional (2D) transition metal dichalcogenides (TMDs) are considered promising for use in next-generation electrical devices. However, the application of TMD-based devices is limited because of the Schottky barrier interface resulting from the absence of dangling bonds on the TMDs' surface. Here, we introduce a facile phase-tuning approach for forming a homogenous interface between semiconducting hexagonal (2H) and semi-metallic monoclinic (1T') molybdenum ditelluride (MoTe₂). The formation of ohmic contacts increases the charge carrier mobility of MoTe₂ field-effect transistor devices to 16.1 cm² V⁻¹s⁻¹ with high reproducibility, while maintaining a high on/off current ratio by efficiently improving charge injection at the interface. The proposed method enables a simple fabrication process, local patterning, and large-area scaling for the creation of high-performance 2D electronic devices.

Keywords: transition metal dichalcogenides; 2D materials; chemical vapor deposition; phase; contact resistance



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1. Introduction

Two-dimensional (2D) materials have received much attention owing to their unique physical, chemical, and electronic properties [1–5]. In particular, hexagonal group-VI 2D transition metal dichalcogenides (TMDs) have received significant research interest due to their exotic bandgap opening and strong spin-orbit coupling, which shows their feasibility in various applications, such as logic transistors, memristor, piezoelectronics, spintronics, wearable/flexible devices, sensors, and valley optoelectronics, among many others [6–11]. Distorted octahedral (T) phase TMDs have recently received attention in the field because of their promising features in novel electronic devices and topological field-effect transistors (FETs) based on quantum spin Hall effects [12]. Unlike the semiconducting (2H) phase, which has a relatively high bandgap opening (≥ 1 eV) and high structural and environmental stability, the semi-metallic (1T') phase with a slight band overlap near the Fermi level is often found to be metastable [12–18]. When this phase is used in FETs, the semiconducting layer comprises a single layer or a few layers and has a covalently bonded lattice, where all charge carriers are confined in an atomically thin channel path, resulting in excellent gate tunability and a high on/off current ratio (I_{on}/I_{off}). Hence, it has significant potential in the optimal scaling of transistors for single-atom devices. However, only inhomogeneous centimeter-scale 1T' MoS₂ has been reported [19]; this limits the size of the 1T' TMD region for scalable applications. Thin-film tellurides, such as MoTe₂ and WTe₂, feature the most optimized electronic features, including superconductivity, quantum spin Hall state, and 2H, 1T, and 1T' phases [12,20–25]. In particular, MoTe₂ is receiving broad

attention for phase-tunable memory devices owing to the small energy gap between the 2H and 1T' phases, and strain-induced phase switching, gating, and heating have been experimentally demonstrated [26–30]. Despite increasing interest in tellurides, creating large-scale 1T'-phase MoTe₂ thin films in electronic applications remains a challenge, as does precisely controlling the number of layer depositions.

Creating an ohmic contact is another obstacle in developing high-performance ultrathin TMD devices because of the lack of surface dangling bonds and the high surface-to-volume ratio. To minimize the Schottky barrier between the semiconducting channel and metallic electrode, previous studies have focused on the contact interface, including the alignment of the work functions of Co, Sc, and Au/Ti electrodes with the conduction and valence band edges of the active layer [31–35]. Fermi-level pinning near the interface has been found to significantly affect device characteristics. In addition, the abundant metal electrodes near the conduction band were also found to improve device performance, owing to the effective carrier injection, which resulted in a decrease in the contact resistance [31]. For example, graphene-electrode-based FET devices show the advantages of electrical properties such as a tunable work function, high electrical conductivity, and nominal damage to the active layer. Transfer-free direct graphene synthesis on the MoS₂ layer resulted in a barrier height of 0 meV at a high gate voltage. Consequently, a high charge carrier mobility can be obtained due to the modified contact resistance and ohmic contact of the graphene and the MoS₂ layer [36]. The contact materials are suggested to form a highly conductive state for the active channel; this ensures a low interface resistance and facilitates carrier injection. However, doping mechanisms for nanoelectronics are typically not straightforward due to their limited doping levels, stability, and the complexity of the process [37]. Therefore, methods for obtaining two different electrical characteristics within a single element layer should be explored to fabricate high-performance electronic devices.

Here, we demonstrate a robust phase-tuning method that enables high-performance MoTe₂ FETs with a heterophase and homojunction interface. We developed a locally patterned semi-metallic 1T' phase on ultrathin semiconducting MoTe₂ nanosheets by laser irradiation. Laser-irradiated 1T' MoTe₂ FET devices exhibited enhanced electrical performances due to the effective charge injection via the homojunction interface. Additionally, we demonstrated a contact resistance of 93 kΩ·μm. This low contact resistance resulted from the work function of the 1T' MoTe₂ and the 2H conduction band near the vacuum level. Furthermore, 1T' MoTe₂ FET devices showed better overall performance than that of the semiconducting 2H MoTe₂ FETs. In addition, we measured the electrical properties in the atmosphere and obtained an average mobility of 16.1 cm² V⁻¹S⁻¹, a high on/off ratio of more than 10⁵, and a subthreshold swing of 98 mV dec⁻¹ from 36 MoTe₂ FET devices. Laser-irradiation-assisted 2H–1T' phase conversion is highly reproducible, suitable for scaling up and applying to large areas, and feasible for engineering the contact interfaces of 2D layered materials for atomically thin nanoelectronics in the near future.

2. Materials and Methods

The mixture of MoO₃/NaCl (4.0 mg/2.3 mg) for MoTe₂ synthesis was placed in a sample boat, and the target substrate (SiO₂/Si substrate) was placed at the top of the boat. A tellurium powder in a crucible for the tellurization of MoO₃ was placed upstream of the Ar carrier gas. After evacuation, the SiO₂/Si substrate was heated to $T = 750$ °C at a pressure of $p = 350$ Torr.

A $p++$ Si/SiO₂ 300 nm wafer was used as a rigid substrate and gate electrode to fabricate the MoTe₂ FETs. Before fabricating the devices, the substrate was rinsed using ethanol, acetone, isopropyl alcohol, and deionized water. The device fabrication was completed by synthesizing a MoTe₂ active layer according to the method described above. To pattern S/D electrodes on the MoTe₂, the shadow mask, with various channel lengths from 30 to 200 μm, was aligned. Then, a Au (30 nm)/Ti (3 nm) electrode was deposited through the thermal evaporator. After fabrication of MoTe₂ FETs, the laser, having the

wavelength of 532 nm, was irradiated at the interface MoTe₂ and S/D electrodes to induce phase transition. The exposure time was varied from 1 to 8 s at an ambient atmosphere.

The crystalline properties of MoTe₂ were characterized via Raman spectroscopy (WITec Alpha 300R, Ulm, Germany), and its thickness was measured using atomic force microscopy (AFM; Bruker MultiMode 8, Camarillo, CA, USA). The chemical composition and bonding properties of MoTe₂ were determined using X-ray Photoelectron Spectroscopy (XPS, PHI VersaProbe, Chanhassen, MN, USA). The electrical characteristics were analyzed using a Keithley 2636A source meter (Tektronix, Beaverton, OR, USA) under an ambient atmosphere.

3. Results and Discussion

Figure 1a shows a schematic of the synthesis of monolayered MoTe₂ via chemical vapor deposition (CVD) on a SiO₂/Si substrate. Figure 1b and Figure S1. show the optical images of the obtained film. The material synthesis is described in detail in the Materials and Methods section. An AFM image and a height profile (Figure 1c) show the thickness and demonstrate that the MoTe₂ flakes on SiO₂/Si have a height of 0.87 nm, indicating that they form a monolayer. Figure 1d shows the Raman spectra of MoTe₂ on the SiO₂/Si substrate after laser irradiation from 1 to 8 s. Laser irradiation causes structural phase evolution in the exposed area. All MoTe₂ films exhibit a relatively weak peak at 235 cm⁻¹ (in-plane E_{12g} mode) but a strong prominent peak at 159 cm⁻¹ (out-of-plane A_g mode) under 532 nm laser excitation [38–40]. The E_{12g} mode signal is typically strong for 2D MoTe₂ flakes; however, the A_g mode signal was higher than that of the E_{12g} mode, indicating that the in-plane signal was diminished.

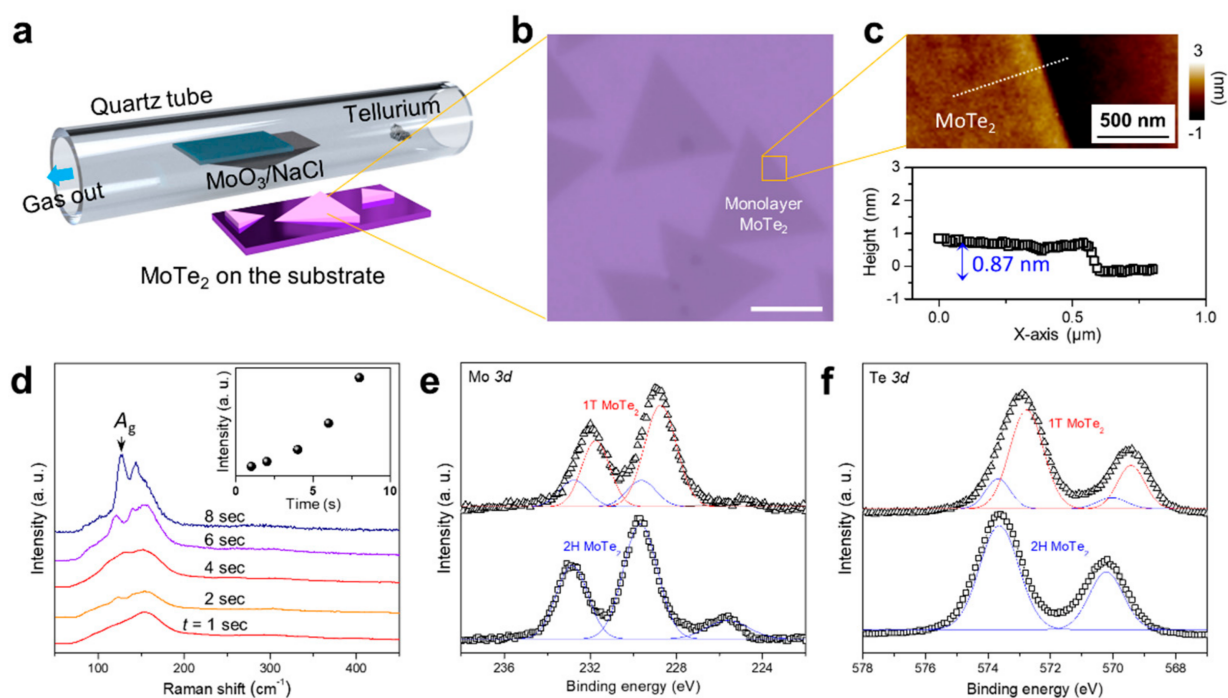


Figure 1. Characterization of CVD-grown MoTe₂: (a) Schematic of CVD synthesis of MoTe₂ on SiO₂/Si substrate using salt-assisted growth; (b) Optical microscopy image of triangular monolayered MoTe₂ nuclei on the SiO₂/Si substrate. The scale bar is 2 μm; (c) AFM image of MoTe₂ and the height profile corresponding to the white dashed line; the thickness of the MoTe₂ is 0.87 nm; (d) Raman spectra of MoTe₂ at various laser irradiation times. The inset shows the evolution of the A_g peak related to the 1T' phase of MoTe₂ with time. XPS spectra show (e) Mo 3d and (f) Te 3d peaks of the 1T' and 2H phases of MoTe₂.

To obtain the critical laser irradiation time for the phase transition in the MoTe₂ samples on the SiO₂/Si substrate, the desired area was gradually and carefully exposed to irradiation by a laser with a power of 2.6 mW and a wavelength of 532 nm. In addition, the Raman spectra were measured after laser irradiation for each duration. The evolution trend was similar for all cases. The E¹_{2g} peak intensity decreased gradually with increasing laser irradiation time, and when the laser irradiation time reached the critical value, two new peaks appeared at 127 and 141 cm⁻¹, which correspond to the A_g mode of 1T' MoTe₂ [30]. These peaks provide direct evidence of the phase transition from the 2H phase to the 1T' phase. The A_g intensity is plotted as a function of the irradiation time in the inset of Figure 1d. The critical laser irradiation time for the phase transition of MoTe₂ from the 2H phase to the 1T' phase was 8 s. The phase transition of MoTe₂ might be explained by the Te vacancy formation in MoTe₂, which results in alterations in the stoichiometry of Mo atoms; this leads to reconstruction of its atomic structure, and phase transition of the 2H phase to the 1T' phase.

Figure 1e,f show the XPS spectra of Mo and Te, respectively. The spectra were obtained before and after laser irradiation for 8 s. As shown in Figure 1e, prominent peaks at 229.4 eV (Mo 3d_{5/2}), 233.0 eV (Mo 3d_{3/2}), 570.3 eV (Te 3d_{5/2}), and 573.5 eV (Te 3d_{3/2}) can be observed in the pristine sample, corresponding to the 2H phase. By contrast, the XPS peaks for 1T' MoTe₂ can be observed at 229.0 eV (Mo 3d_{5/2}), 232.2 eV (Mo 3d_{3/2}), 569.6 eV (Te 3d_{5/2}), and 572.9 eV (Te 3d_{3/2}). The binding energies of Mo and Te atoms in the irradiated area exhibit an energy shift of 0.4–0.8 eV between two different phases on MoTe₂. This shift can be attributed to the distinct lattice symmetry of the 2H and 1T' phases of MoTe₂ [41]. This value is similar to previously reported energy shift values ranging from 0.4 to 0.6 eV [41–43]. Moreover, we observed that in 1T' MoTe₂, Te 3d peaks are blue-shifted by 0.7 eV, whereas the Mo 3d peaks are blue-shifted by 0.4 eV. It has also been previously reported that chalcogen deficiency in TMDs decreases the binding energy of the transition metal by approximately the same amount [44].

To evaluate the feasibility of MoTe₂ with a heterophase homojunction interface for electronic applications, MoTe₂ FET devices were fabricated on a SiO₂/Si substrate. Figure 2a,b show a schematic of the MoTe₂ FET device structure and its A_g Raman mapping image after laser irradiation, respectively. The A_g Raman mappings show that the MoTe₂ monolayer is not limited to deposit uniformly over the entire area but also includes the channel region, resulting in 2H to 1T' phase-transformed MoTe₂ devices. Overall, the laser irradiation approach efficiently enabled the fabrication of MoTe₂ FETs by simple phase tuning. The transfer characteristics of back-gate FETs were investigated under laser irradiation. Figure 2c,d show the drain current (*I*_D) as a function of the gate voltage (*V*_G) of the 2H and 1T' MoTe₂ FETs, respectively. The measured curves show a high on/off ratio of more than 10⁵ and typical p-type transistor characteristics. From the obtained *I*-*V* curves of the MoTe₂ FETs, we determined the field-effect mobility (*μ*) as follows:

$$I_D = \frac{W}{2L} C u (V_{GS} - V_T)^2 \quad (1)$$

where *I*_D is the drain current, *W* is the channel width of the electrode, *L* is the channel length of the electrode, *C* is the area capacitance of the SiO₂ layer, *V*_{GS} is the gate voltage, and *V*_T is the threshold voltage. At the saturation regime, the average mobility values of 13.6 cm² V⁻¹s⁻¹ for the 36 2H-phase devices and 16.1 cm² V⁻¹s⁻¹ for the 36 1T'-phase devices were obtained, respectively; these values represent an increase of nearly 20% (Figure 2e). The high on/off ratios for the 36 2H MoTe₂ FETs devices were well preserved after phase tuning to 1T' by laser irradiation, as shown in Figure 2f. Moreover, 1T' phase transformed FETs showed a high on-current level (~3.1 mA), and threshold voltage. This indicates that charge carrier movements were well preserved or improved after forming the homojunction interface. Interestingly, according to the statistical results, the on-current, as well as the off-current, slightly increased, so the carrier mobility of the 1T' MoTe₂ FETs was increased overall, but the average on/off ratio decreased. These results demonstrate that

the heterophase homojunction interface between the active layer and electrodes enhanced the electrical performance of the MoTe₂ FET devices and indicates the feasibility of the laser irradiation approach.

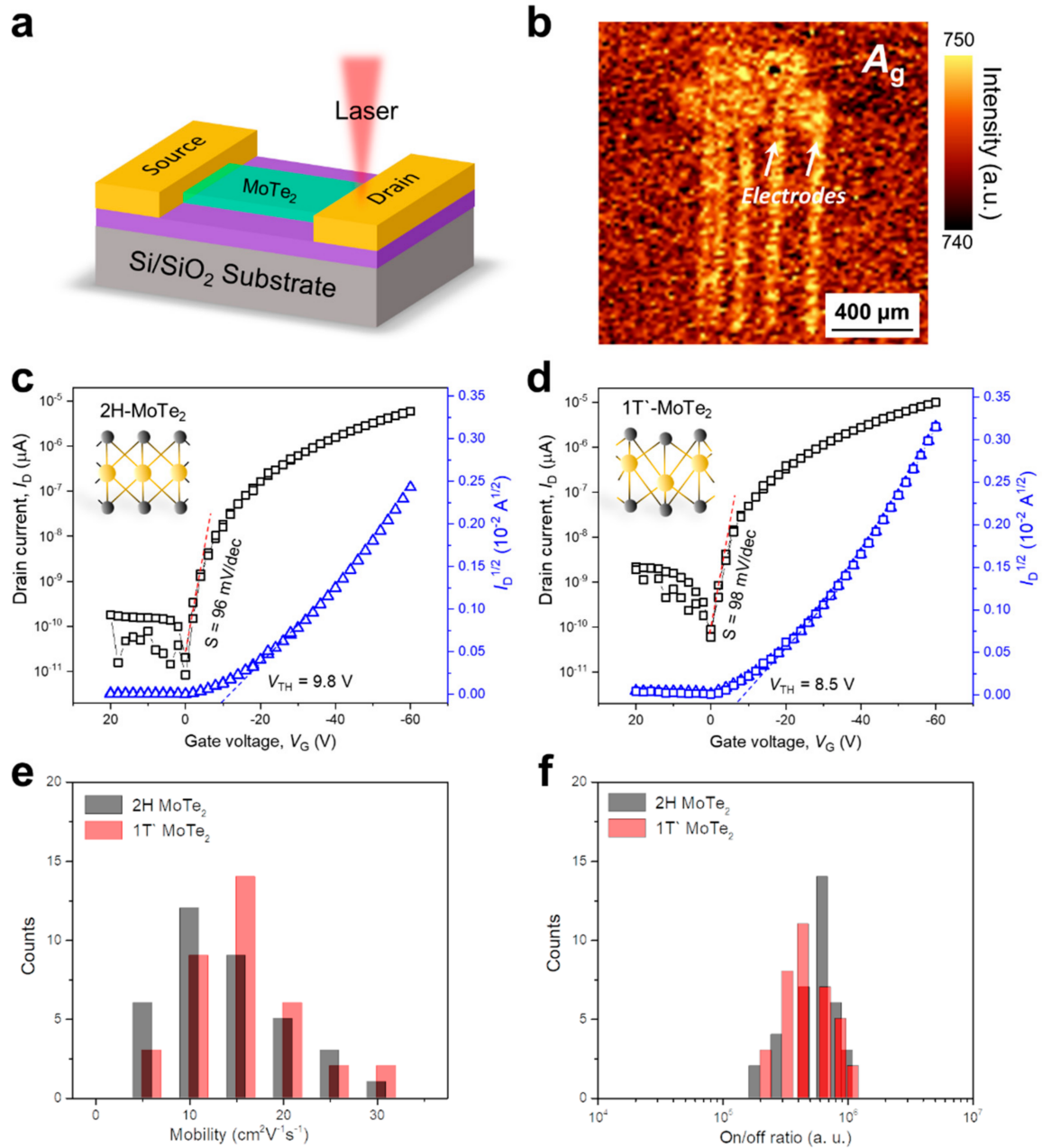


Figure 2. Electrical characterization of 2H and 1T'-phase of MoTe₂ FETs: (a) Schematic structure of MoTe₂ FETs on SiO₂/Si substrate; the dimensions of the MoTe₂ FETs are the following: channel length 20 μm and width 400 μm. (b) Two-dimensional A_g Raman mapping image of MoTe₂ FETs on SiO₂/Si substrate after laser irradiation. Transfer curves of (c) 2H MoTe₂ and (d) 1T' MoTe₂ FETs after laser irradiation; (e) Statistical diagram of the measured carrier mobilities; (f) On/off ratio of the 36 fabricated 2H and 1T' FETs.

The contact resistances were obtained via the transfer length method (TLM) to demonstrate the reasons for the improvement in the electrical characteristics. From the TLM plots, the contact resistance values were extracted from the value for a channel length of zero. To obtain an excellent statistical agreement, at least 36 samples were measured and analyzed. Figure 3a,b show that the resistance of 1T' MoTe₂ decreased by 22.5%, from 120 to 93 kΩ·μm. Many studies have reported that the interface characteristics are among the

main factors affecting contact resistance [45,46]. In the $1T'$ MoTe₂ device, phase tuning by laser irradiation helps reduce the contact resistance between the semiconducting layer and the electrodes. This increases the mobility and decreases the threshold voltage by reducing the contact resistance. Figure 3c shows that the 2H MoTe₂ device exhibits nonlinear I_D - V_{DS} characteristics, indicating Schottky behavior, whereas an ohmic-like behavior is observed in the $1T'$ MoTe₂ device performance, as shown in Figure 3d.

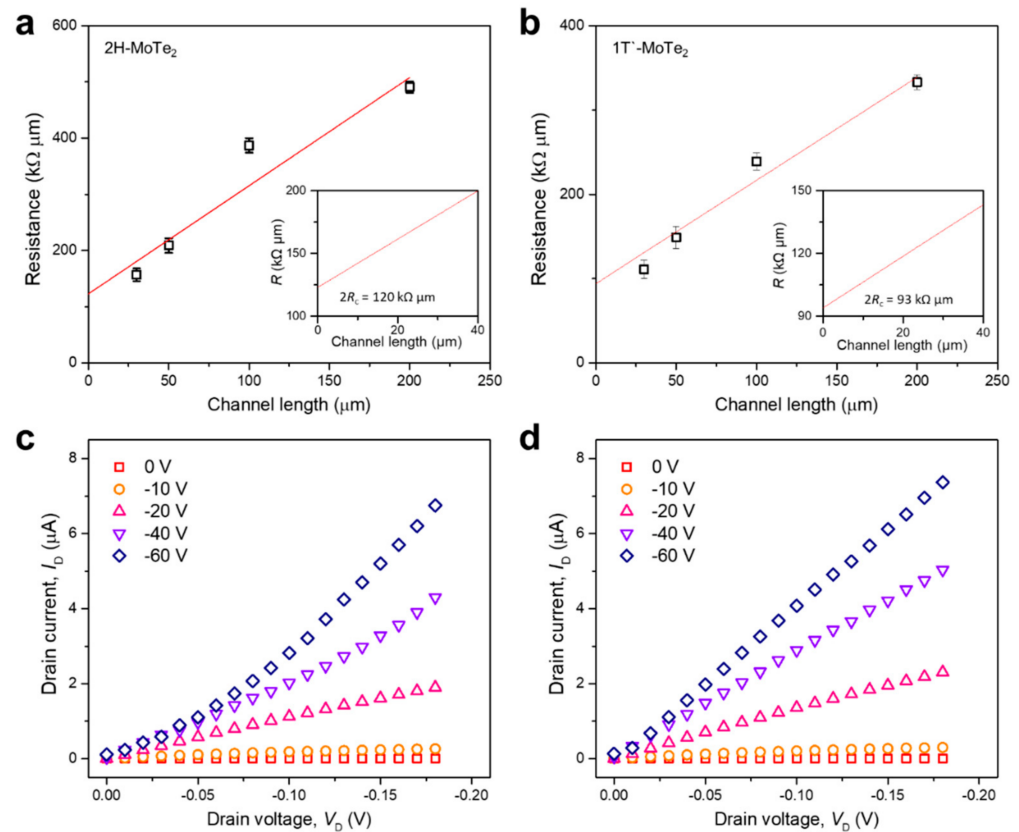


Figure 3. Contact resistance of the 2H-phase and $1T'$ -phase of MoTe₂ FETs: (a) 2H MoTe₂ and (b) $1T'$ MoTe₂ FETs after laser irradiation. The channel width is normalized using the resistance obtained from 2H MoTe₂ FETs; Output curves of (c) 2H and (d) $1T'$ -phase MoTe₂ FETs after laser irradiation; the FETs have gate voltages ranging from 0 to -60 V.

To further quantify the ohmic contact in these devices, the barrier height at the interface between the S/D electrodes and the MoTe₂ channel was studied [36,47]. The electrical properties were characterized at various temperatures to measure the energy levels of the metallic and semiconducting layers in the $1T'$ -phase MoTe₂ devices, as shown in Figure 4.

As the temperature increases, the drain current (I_D) also increases due to the hopping transport, which dominates the charge transport in MoTe₂ [48]. The increase in the current with temperature is due to the increased charge carriers at elevated temperatures, where the thermal energy is sufficient to overcome the activation energy. The results were fitted using the following thermal emission equation to obtain an Arrhenius plot of the laser-irradiated $1T'$ MoTe₂ devices at different gate voltages:

$$I_{DS} = AT^{3/2} \exp\left(-\frac{q\phi_B}{k_B T}\right) \left[\exp\left(\frac{qV_{DS}}{nk_B T}\right) - 1 \right] \quad (2)$$

where I_{DS} is current, A is the Richardson's constant, T is the temperature, ϕ_B is the Schottky barrier, q is the charge constant, k_B is the Boltzmann constant, V_{DS} is the drain voltage, and n is the nonideal factor [32,49]. When $V_{DS} \gg k_B T$, Equation (2) can be simplified as:

$$\ln\left(\frac{I_{DS}}{T^{3/2}}\right) = -\frac{q\left(\phi_B - \frac{V_{DS}}{n}\right)}{k_B T} + \ln(A) \quad (3)$$

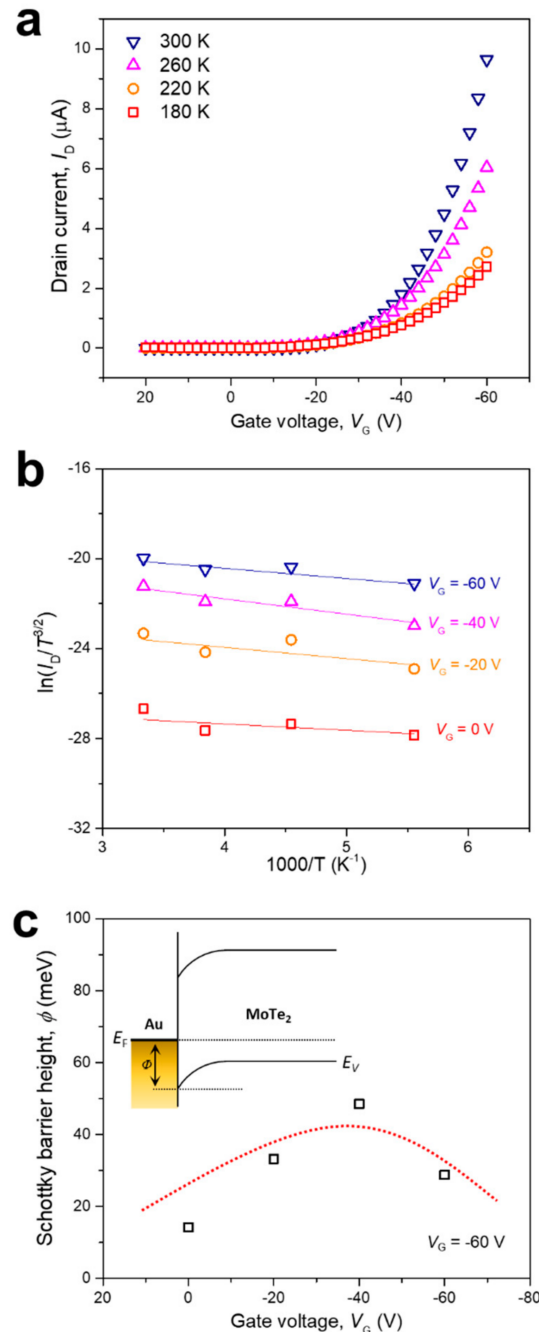


Figure 4. Transfer curves and Schottky barrier of 1T' MoTe₂ FETs at various temperatures: (a) Transfer curves obtained from laser-irradiated 1T' MoTe₂ FETs at temperatures ranging from 180 to 300 K; (b) Arrhenius plot of laser-irradiated 1T' MoTe₂ FETs with gate voltages ranging from 0 V to −60 V; (c) Schottky barrier height of laser-irradiated 1T' MoTe₂ FETs. The inset shows the schematic of the Schottky barrier of p-type MoTe₂ with contact to the Au electrode.

The Schottky energy barrier (ϕ_B) can be expressed as the slope of the simplified equation in a curve of $\ln(I_{DS}/T^{3/2})$ versus $1000/T$. The results were plotted at different V_G values for the 1T' MoTe₂ FETs (Figure 4b). Finally, to obtain the nonideal factor (n), which is required to calculate ϕ_B for each slope, we used the following equation:

$$\frac{dV_{DS}}{dI_{DS}} = \frac{nk_B T}{q} \frac{1}{I_{DS}} + R_s \quad (4)$$

where R_s is the series resistance. We calculated n as the slope of a curve of dV_{DS}/dI_{DS} versus $1/I_{DS}$ and obtained a value of 9.93 (Figure S2) [36,49]. The value of ϕ_B was calculated from these results (Figure 4c). When a zero-gate voltage was applied, the Schottky energy barrier was lower in the 1T' MoTe₂ device than in the 2H MoTe₂ device, demonstrating that 1T' MoTe₂ forms a sharper interface between the electrode and the semiconducting layer than 2H MoTe₂ (Figure S3). Note that the difference in transistor polarity between the 2H and 1T' devices is explained simply by modifying the Schottky barrier height by laser irradiation. An advantage of the 1T' device is that the S/D electrodes are side contacts rather than vertical contacts, providing better device properties [50,51]. Furthermore, this result is fundamentally different from previously reported results on chemical doping. The transistor polarity change mechanism preserves the electron and hole mobilities. It efficiently improves electron mobility with a facile approach. Moreover, 1T'-phase devices are highly independent of the metal of the S/D electrodes. We designed 1T' MoTe₂ devices with the same electrodes and achieved a yield of nearly 100% with consistent performance. Additionally, an ohmic-like contact was achieved at the interface between the active layer and electrodes, which improved the charge carrier injection without barriers. Although the 1T' phase is metastable, it has been demonstrated that stability can be maintained under environmental conditions after facile laser irradiation.

4. Conclusions

In summary, we developed a facile method for phase tuning from 2H to 1T' MoTe₂ by laser irradiation. This method produces a 1T' MoTe₂ monolayer that facilitates charge injection at the interface and thus improves the electrical characteristics of the MoTe₂ FETs. Moreover, we showed that phase tunability could help effectively reduce the contact resistance at the interface between the S/D electrodes and the MoTe₂ channel, thus providing high carrier mobility of the FETs. Additionally, we demonstrated that the 1T' phase of MoTe₂ is a heterophase homojunction electrode with a low contact resistance value of 93 k Ω · μ m at zero gate bias. The low contact resistance results in a high on-current of 3.1 mA^{1/2}, and a high charge carrier mobility of 16.1 cm² V⁻¹s⁻¹. Our work demonstrates that phase engineering is a practical approach for improving the performance of MoTe₂ devices.

Supplementary Materials: The following are available online at <https://www.mdpi.com/article/10.3390/nano11112805/s1>, Figure S1: Optical image of the CVD-grown MoTe₂ on the SiO₂/Si substrate, Figure S2: I-V curve of MoTe₂ FETs, Figure S3: Schottky barrier height of MoTe₂ FETs. Table S1. Comparison the device performances.

Author Contributions: Conceptualization, J.K. (Junyoung Kim) and E.L.; methodology, G.Y.B.; software, E.L.; validation, G.Y.B., J.K. (Jinsung Kim) and S.L.; formal analysis, J.K. (Junyoung Kim); investigation, G.Y.B. and J.K. (Jinsung Kim); resources, J.K. (Junyoung Kim); data curation, J.K. (Junyoung Kim) and S.L.; writing—original draft preparation, J.K. (Junyoung Kim) and E.L.; writing—review and editing, J.K. (Junyoung Kim) and E.L.; visualization, E.L.; supervision, E.L.; project administration, E.L.; funding acquisition, E.L.; contributed equally, G.Y.B., J.K. (Jinsung Kim) and J.K. (Junyoung Kim). All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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