

PAPER

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Transition metal dichalcogenides (TMDs) are promising candidates for the semiconductor industry owing to their superior electrical properties. Their surface oxidation is of interest because their electrical properties can be easily modulated by an oxidized layer on top of them. Here, we demonstrate the XeF₂-mediated surface oxidation of 2H-MoTe₂ (alpha phase MoTe₂). MoTe₂ exposed to XeF₂ gas forms a thin and uniform oxidized layer (~2.5 nm-thick MoO_x) on MoTe₂ regardless of the exposure time (within ~120 s) due to the passivation effect and simultaneous etching. We used the oxidized layer for contacts between the metal and MoTe₂, which help reduce the contact resistance by overcoming the Fermi level pinning effect by the direct metal deposition process. The MoTe₂ field-effect transistors (FETs) with a MoO_x interlayer exhibited two orders of magnitude higher field-effect hole mobility of 6.31 cm² V⁻¹ s⁻¹ with a high on/off current ratio of ~10⁵ than that of the MoTe₂ device with conventional metal contacts (0.07 cm² V⁻¹ s⁻¹). Our work shows a straightforward and effective method for forming a thin oxide layer for MoTe₂ devices, applicable for 2D electronics.

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Introduction

Transition metal dichalcogenide (TMD) semiconductors have been actively studied owing to their great potential in advanced electronics and unprecedented physical properties.^{1–8} Electronic devices based on exfoliated TMDs have a thickness limit of a few atomic layers with dangling-bond-free passivated surfaces because individual layers of TMDs are held together with weak van der Waals forces. Among these TMDs, 2H-MoTe₂ (alpha phase MoTe₂) has a moderate indirect band gap (E_g) of 0.88 eV in bulk^{9,10} and a direct band gap (E_g) of 1.1 eV in a monolayer form.¹¹ The moderate band gap of MoTe₂, similar

to that of bulk silicon, makes it a promising candidate for various electronic applications, such as transistors, complementary logic devices, optoelectronic devices, and memory.^{12,13} It is well known that the Schottky barrier height between MoTe₂ and the metal is very weakly dependent on the work function of the metal due to the strong Fermi level pinning effect, leading to a high contact resistance (R_c).¹⁴ Although the Schottky barrier height is determined by the work function of the contact metal according to the Schottky–Mott rule, a strong Fermi level pinning effect and high contact resistance (R_c) are caused by physical bombardment during the metal deposition process, leading to crystallinity damage in MoTe₂.¹⁵ It was recently found that transition metal oxides (TMOs) are suitable candidates as buffer layers between TMDs and metals.^{16,17} The fabrication of TMOs has been reported using various methods, such as oxygen plasma treatment,¹⁸ atomic layer deposition (ALD),¹⁹ UV–ozone treatment²⁰ and thermal evaporation.²¹ However, surface treatment methods can easily damage TMDs, leading to reduced crystallinity and increased surface roughness.^{15,22} The atomic thickness of TMD flakes gives rise to these issues, degrading the electronic properties of the device.

In this work, we demonstrate the fabrication of a thin and uniform oxidized layer on top of MoTe₂ by exposing it to XeF₂ gas. The oxidized layer had a uniform thickness of ~2.5 nm regardless of the exposure time (within ~120 s) due to the passivation effect and simultaneous etching. The non-stoichiometric oxide layer has a high work function, allowing for p-doping of the underlying MoTe₂. The MoTe₂ device showed a high field-effect hole mobility of 6.31 cm² V⁻¹ s⁻¹ with a high

^aDepartment of Material Science and Engineering, Yonsei University, Seoul, 03722, Korea^bDepartment of Energy Systems Research, Ajou University, Suwon 16499, Republic of Korea^cDepartment of Materials Science and Engineering, Ajou University, Suwon 16499, Republic of Korea^dDepartment of Materials Science and Engineering, Seoul National University, Seoul 08826, Korea^eResearch Institute of Advanced Materials (RIAM), Seoul National University, Seoul 08826, Korea^fDepartment of Physics and Pohang Accelerator Laboratory, Pohang University of Science and Technology, 37673, Pohang, Korea^gInstitute of Engineering Research, Seoul National University, Seoul 08826, Korea^hInstitute of Applied Physics, Seoul National University, Seoul 08826, Korea. E-mail: gwanlee@snu.ac.kr

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‡ These authors contributed equally: Eunji Ji and Jong Hun Kim.

on/off current ratio of $\sim 10^5$. The palladium (Pd) metal can easily tunnel to MeTe_2 layer by MoO_x layer. The mobility is two orders of magnitude higher than that of the MoTe_2 device with conventional metal contacts.

Results and discussions

The mechanically exfoliated MoTe_2 flakes were exposed to XeF_2 gas at 1.8 torr for different treatment times. The optical microscope images of the MoTe_2 flakes in Fig. 1a show the contrast change before and after XeF_2 exposure for 150 s. The corresponding surface morphology was also identified by using atomic force microscopy (AFM), as shown in Fig. 1b. As confirmed using AFM images, even after the treatment, the overall structure was not changed, exhibiting the relatively uniform surface morphologies; however, the optical contrast of the flakes was significantly suppressed after XeF_2 exposure. The depression in the optical contrast may be attributed to the change in the optical path caused by mediating the refractive properties of MoTe_2 with XeF_2 treatment.^{23,24} Fig. 1c shows the height profiles created by following the dashed lines in Fig. 1b. The thickness of the fourth layer (4 L) and third layer (3 L) MoTe_2 was measured to be ~ 3.1 nm and ~ 2.4 nm before XeF_2 exposure (blue dashed lines in Fig. 1b), respectively. However,

the thickness remained approximately the same even after XeF_2 exposure (red dashed lines in Fig. 1b), *i.e.*, ~ 3.0 nm and ~ 2.3 nm, respectively. Fig. 1d and e show the Raman spectra of the 3 L and 4 L regions (blue and red dots in Fig. 1a, respectively) of the MoTe_2 flakes. MoTe_2 showed the representative phonon modes of A_g^1 (~ 170 cm^{-1}), E_{2g}^1 (~ 234 cm^{-1}), and B_{2g}^1 (~ 289 cm^{-1}).²⁵ Meanwhile, monolayer (1 L) MoTe_2 was found to have a prominent peak of E_{2g}^1 and a weaker peak of A_{1g} at ~ 235 cm^{-1} and ~ 174 cm^{-1} , respectively.²⁶ The Raman spectra of 3 L MoTe_2 in Fig. 1d show that all Raman modes were depressed after XeF_2 exposure. However, only the B_{2g}^1 mode of 4 L MoTe_2 disappeared after XeF_2 exposure, maintaining other Raman peaks (Fig. 1e). This indicates that a few topmost layers of MoTe_2 were substituted with a different material with a comparable atomic thickness, while the subsurface remained as MoTe_2 . It was expected that unidentified layers were produced upon XeF_2 exposure. In particular, the Raman mode related to MoO_3 ²⁷ and MoO_2 ^{28,29} cannot be observed. Therefore, Raman spectroscopy failed to provide clear evidence of it.

ESI, Fig. S2a and b† show the optical microscope images and Raman spectra of 1 L and 2 L of MoTe_2 before and after XeF_2 exposure at 1.8 torr for 60 s, respectively. After XeF_2 treatment, the intensities of E_{2g}^1 and B_{2g}^1 in 2 L MoTe_2 were slightly lower

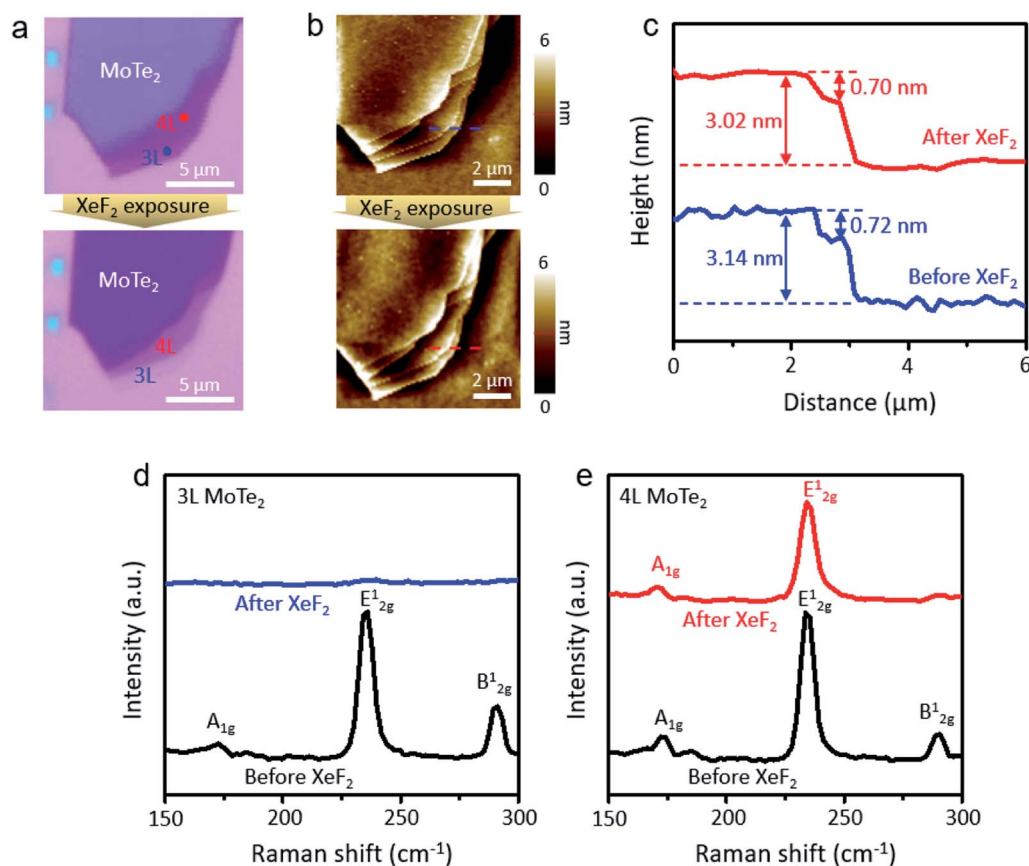


Fig. 1 (a) Optical microscope images of few layer MoTe_2 before and after XeF_2 exposure at 1.8 torr for 150 s, (b) AFM topology images of MoTe_2 before and after XeF_2 treatment, (c) height profile of MoTe_2 before (blue line) and after (red line) XeF_2 treatment, (d) the Raman spectra of 3 L MoTe_2 (blue dot region) in Fig. 1a before (black line) and after (red line) XeF_2 treatment, and (e) the Raman spectra of 4 L MoTe_2 (red dot region) in Fig. 1a before (black line) and after (red line) XeF_2 treatment.



than those before XeF₂ exposure. However, the B_{2g}¹ mode in 2 L disappeared after XeF₂ exposure in the Raman spectrum, indicating that 2 L transforms into 1 L at 1.8 torr for 60 s (the graph at the top in the red-colored region in ESI, Fig. S2b†). All modes in 1 L of MoTe₂ disappeared after XeF₂ exposure, indicating that its crystal structure vanished (the graphs in the blue-colored region of ESI, Fig. S2b†). The E_{2g}¹ mode of XeF₂-exposed 2 L was downshifted by 2.18 cm⁻¹ compared to that of pristine 1 L of MoTe₂ (before XeF₂ exposure). The A_g¹ mode showed an upshift from 171.94 cm⁻¹ (pristine 1 L of MoTe₂) to 172.15 cm⁻¹ (XeF₂-exposed 2 L of MoTe₂), which is another evidence for p-type doping of MoTe₂ after XeF₂ treatment.³⁰ We used the Raman mapping method to verify the uniform treatment of XeF₂ gas on the MoTe₂ surface (ESI, Fig. S2c and d,†). E_{2g}¹ and B_{2g}¹ intensities uniformly disappeared and remained after XeF₂ exposure. In particular, the E_{2g}¹ intensity remained and B_{2g}¹ mode completely disappeared in the 2 L region after XeF₂ exposure, implying that 2 L becomes uniform 1 L.

Scanning transmission electron microscopy (STEM) was used to identify the mysterious top layer of XeF₂-exposed MoTe₂. As shown in the cross-sectional STEM images of Fig. 2a, two MoTe₂ samples with different XeF₂ exposure times of 60 and 120 s at 1.8 torr showed a uniform and smooth layer of the same thickness (~2.5 nm) on top of intact MoTe₂. The underlying MoTe₂ might have remained intact, without any structural changes, even after XeF₂ exposure. The surface was continuously scanned using contact-mode AFM to evaluate the thickness of the oxidized layer. The topmost region of XeF₂-exposed MoTe₂ can be smoothly removed by using an AFM tip, as shown in Fig. 2b. The height profile based on the red dashed lines in Fig. 2b is shown in Fig. 2d. The exposed new surface had a difference in thickness of ~2.6 nm compared to the unexposed surface, which is consistent

with the TEM results. The peeled area appeared as flat as the non-peeled area. In addition, the energy-dispersive X-ray spectroscopy (EDS) map in Fig. 2c clearly shows that the top region of XeF₂-exposed MoTe₂ contains more oxygen than the underlying region. Although it was challenging to quantitatively analyze the composition of the top region due to the resolution limit of the EDS and ultrathin sample, we could confirm that the top region of MoTe₂ was predominantly oxidized and disordered by XeF₂-exposure (a green dotted region indicates oxygen), while the lower MoTe₂ remained approximately intact even after XeF₂ exposure, showing only the Mo (purple dots) and Te (cyan-colored dots) spectra in EDS.

The AFM images in Fig. 3a show the surface morphology of XeF₂-exposed MoTe₂ for different times. The surface of XeF₂-exposed MoTe₂ is cleaner and smoother than plasma-treated MoTe₂ for a long exposure time of 270 s owing to the suppression of ion-bombardment effect.^{31,32} The roughness and relative thickness of XeF₂-exposed MoTe₂ are plotted in Fig. 3b and c, respectively. The actual thicknesses of the flakes were measured using contact AFM. We did not observe a significant difference in the roughness of MoTe₂ even after a long exposure of 270 s (Fig. 3b). Thermodynamically, the XeF₂-treatment is possible to remove Mo or Te from the surface because the melting temperatures of MoF₆ and TeF₆ are lower than the room temperature.³³ Therefore, if a few top layers of MoTe₂ reacted with XeF₂, it would turn into a disordered structure with several broken bonds. Moreover, the disordered layer can be easily oxidized by exposing it to air after treating the surface with XeF₂ gas. Therefore, the broken structures can easily bond with oxygen when the sample is unloaded from the chamber. Some Mo–O bonds were expected to form at the surface defect sites, thinning the intrinsic MoTe₂.³⁴ In Fig. 3c, the relative thickness

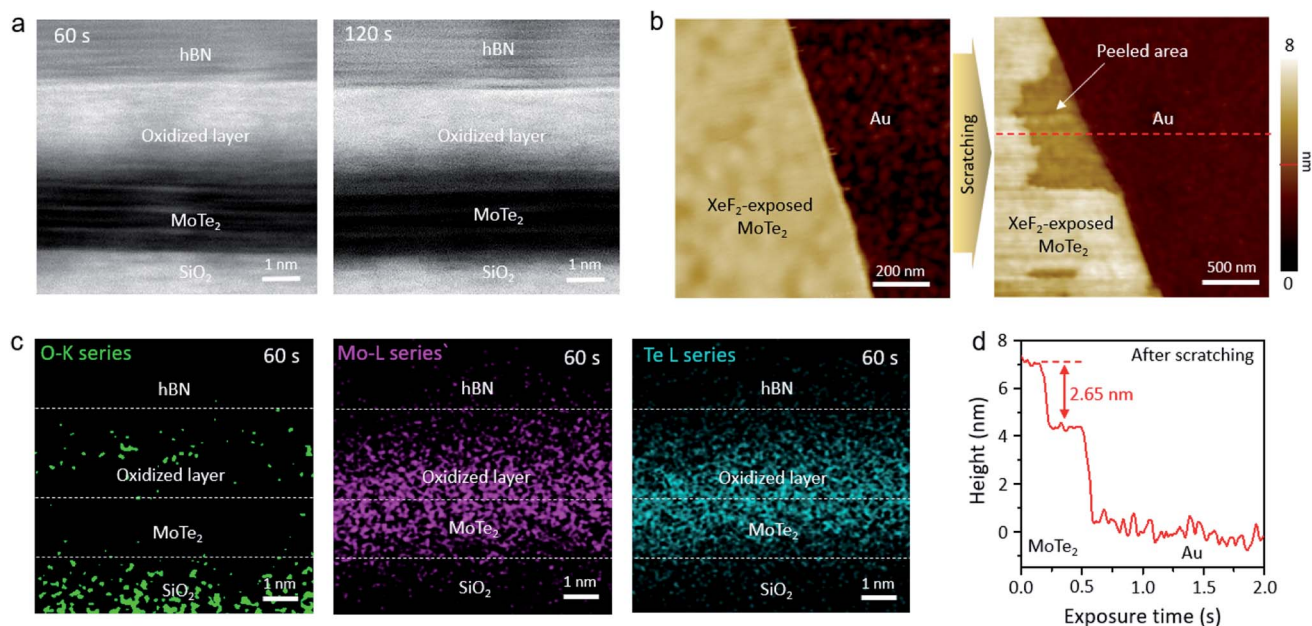


Fig. 2 (a) Cross-sectional STEM images of MoTe₂ after XeF₂ treatment at 1.8 torr for 60 s and 120 s, respectively, (b) AFM topography images of XeF₂ treated MoTe₂ before and after removing the surface layer by using a cantilever tip, (c) EDS mapping image of O-K, Mo-L and Te-L series of MoTe₂ after XeF₂ treatment at 1.8 torr for 60 s and (d) height profile of XeF₂ treated MoTe₂ after scratching by using a cantilever tip.



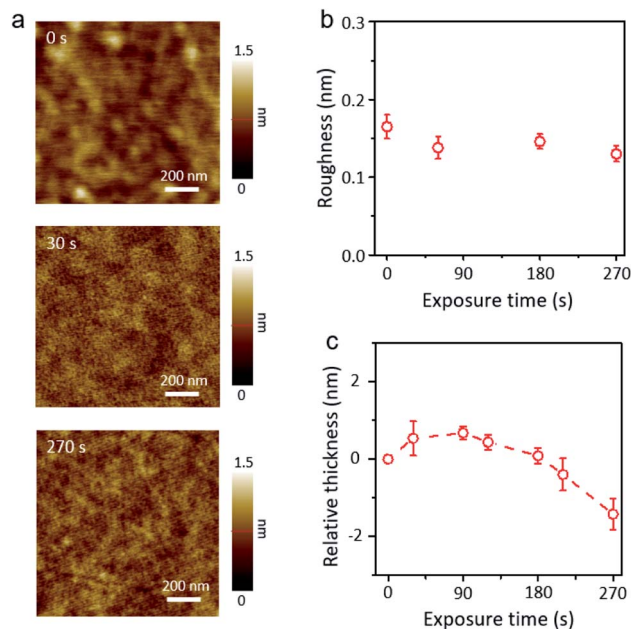


Fig. 3 (a) The AFM topography images of XeF₂ exposed MoTe₂ to show the uniform surface morphology after XeF₂ treatment for 0 s (as-exfoliated MoTe₂), 30 s and 270 s, respectively, (b) the root-mean-square (RMS) roughness of MoTe₂ after XeF₂ treatment at 1.8 torr with appropriate exposure times (from 0 s to 270 s) and (c) the relative height change of MoTe₂ as a function of increasing XeF₂ treatment time at 1.8 torr.

(the difference in thickness before and after XeF₂ exposure) was plotted as a function of the treatment time. The relative thickness of MoTe₂ gradually increased with the treatment time (from 0 to 120 s) after XeF₂ exposure because of the self-limiting growth mechanism of MoO_x.^{35,36} This is consistent with the results illustrated in Fig. 2a (similar thicknesses of the oxidized layer after XeF₂ exposure for 60 and 120 s). However, the relative thickness decreased rather rapidly with the exposure time from 120 to 270 s. The formation of MoF₆ in the chamber can become more dominant than MoO_x due to the Gibbs free energies of MoF₆, MoO₂, and MoO₃ (*i.e.*, -1473.17 , -533.0 , and -668.1 J mol⁻¹, respectively).³³ Therefore, such a decrease in the thickness can be observed as MoF₆ can be easily removed owing to its low melting and boiling temperatures. When the available number of fluorine atoms decreased at a reduced pressure of 1 torr-60 s XeF₂ (ESI, Fig. S3a and b,†), the number of bonding between carbon (C) and fluorine (F) atom should be limited, which revealed the nanohole surface on the MoTe₂ surface. In this case, while the relative thickness was just negligibly changed (ESI, Fig. S3d,†), the surface roughness became deteriorated, as shown in ESI, Fig. S3c.† We used the same conditions—fabrication of a uniform oxidized layer with no nanoholes on MoTe₂ with XeF₂ gas of 1.8 torr—for other experiments. Changes in the morphology due to various treatments in 2D materials devices often lead to reduced electrical contact.^{37–39} We directly fabricated smooth and uniform oxidized layers on MoTe₂ through XeF₂ gas under the conditions of 1.8 torr within 120 s. The XeF₂ exposure method is

easier to scale-up compared with previous methods, such as laser, plasma and thermal annealing.^{40–42}

Photoelectron spectroscopy (PES) with a photon energy of 780 eV was performed under ultra-high vacuum conditions to identify the top layer of MoTe₂ before (black dotted lines) and after (red dotted lines) exposure to XeF₂ (Fig. 4a–d). In each figure, the top-(bottom-) panel shows the core-level spectrum before (after) the treatment. We note that the MoTe₂ flakes were treated with XeF₂ gas at 1.8 torr for 60 s for PES-characterization. The Mo 3d core level from MoTe₂ showed the binding energies of Mo 3d_{5/2} at 228.4 eV and Mo 3d_{3/2} at 231.9 eV before XeF₂ exposure, agreeing with the previous report (Fig. 4a).⁴³ This indicates that the as-prepared MoTe₂ sample has high crystallinity. However, mixed states of Mo⁴⁺ and Mo⁶⁺ were observed after XeF₂ exposure of 60 s (red dotted lines), supporting the partial oxidation of molybdenum. Additionally, the Te–O bonds in the XPS spectra showed two peaks at 576.3 eV and 587.1 eV (upper graph of Fig. 4b, red dotted lines). The more interesting result is that the Mo 3d and Te 3d peaks are downshifted to almost the same extent (~ 0.7 eV). The peak downshifts can be assigned to hole-doping, since the Fermi level, which represents zero energy, moves further away from the conduction band with hole-doping.^{44,45} Besides, the previously suggested Raman spectra also support that the p-type doping occurred after XeF₂ exposure (ESI, Fig. S2b,†). As expected, the O 1s core level appeared after XeF₂ exposure (Fig. 4c). No peak related to H₂O (~ 532 eV³⁰) was found because PES was performed under ultra-high vacuum conditions immediately after mechanically exfoliating MoTe₂. The O 1s core-level spectrum together with the occurrence of Mo⁶⁺ in Mo 3d strongly indicates that an oxidized layer was formed, such as MoO_x.⁴⁶ Lastly, the F 1s peak (Fig. 4d) shows that a small number of fluorine atoms are chemically bonded. Hence, MoO_x, a disordered oxide layer containing negligible fluorine atoms, was formed, playing an important role in doping.

The valence band edges of the samples were investigated using PES ($h\nu = 90$ eV), as shown in Fig. 4e. The valence band edge of MoTe₂ before (black) and after (red) XeF₂ exposure was found to be positioned at ~ 0.8 eV and ~ 3.0 eV below the Fermi level, respectively. The valence band edge of XeF₂-treated MoTe₂ shifted to about 3.0 eV, which is close to the value of the MoO_x phase.⁴⁷ Interestingly, MoO_x does not contain a high density of defects (gap states) inside both MoTe₂ surfaces before (black) and after (red) XeF₂ exposure. The ultrathin oxidized layer can completely change the band diagram. Also, the secondary electron cutoffs (Fig. 4f) show that the work functions of the samples were positioned at ~ 4.8 (pristine MoTe₂) and ~ 5.6 eV (MoO_x), respectively, due to the formation of a non-stoichiometric oxidation phase after XeF₂ exposure. The work function of XeF₂-exposed MoTe₂ was smaller than that (~ 6.8 eV) of MoO₃.⁴⁸ Therefore, the result indicates that only a few topmost surface layers of MoTe₂ (3 L in Fig. 1) were substituted with MoO_x with a comparable atomic thickness, while the subsurface (1 L in Fig. 1) still remained as MoTe₂; however, the overall thickness was scarcely changed by XeF₂-treatment within a limited exposure time. A schematic image of MoTe₂ before and after XeF₂ exposure is shown in ESI, Fig. S4.† The 4 L of MoTe₂ was



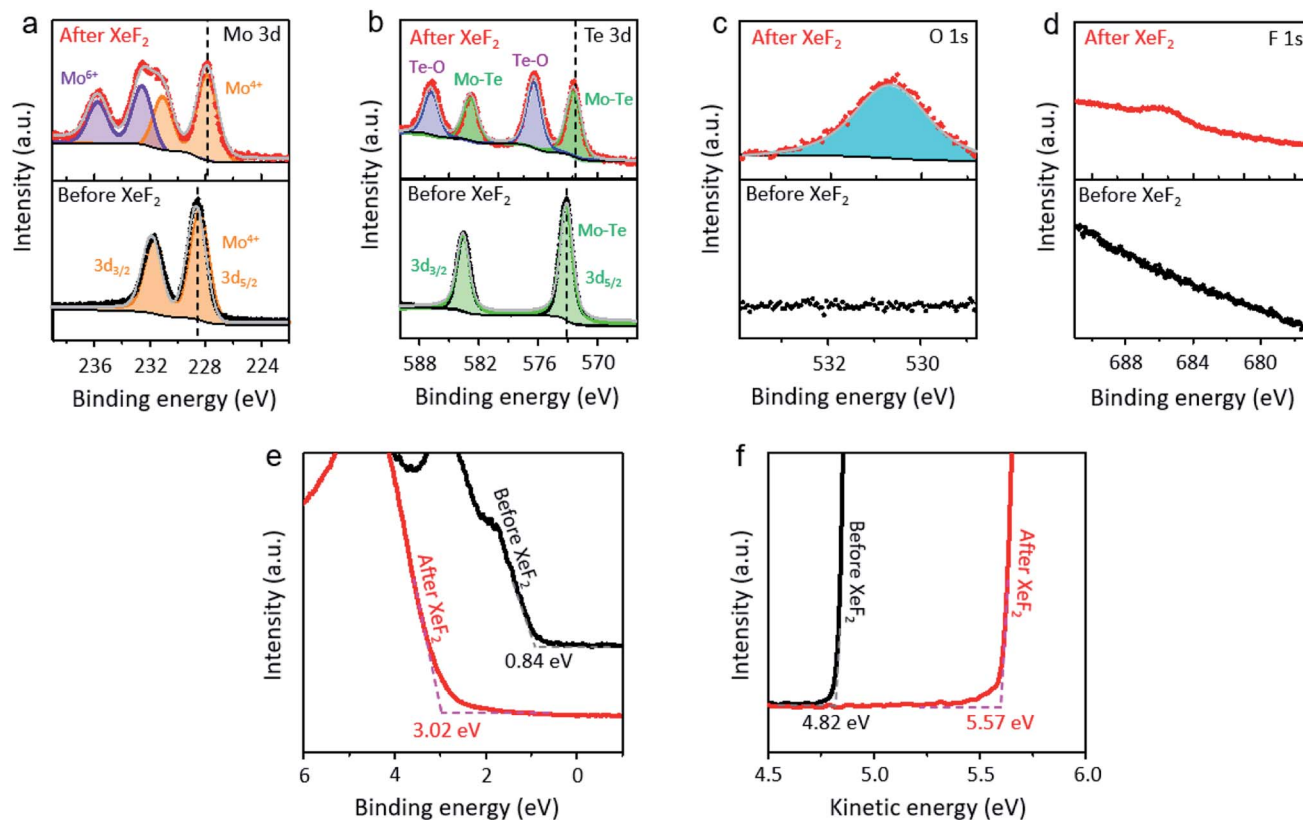


Fig. 4 (a–d) PES spectra with a photon energy of 780 eV of 2H-MoTe₂ without and with XeF₂ treatment at 1.8 torr for 60 s; the bottom spectra of PES show MoTe₂ before XeF₂ treatment and the top spectra of PES show that after XeF₂ treatment at 1.8 torr for 60 s. (a) Mo 3d spectra, (b) Te 3d spectra, (c) O 1s spectra and (d) F 1s spectra. (e and f) PES ($h\nu = 90$ eV) of 2H-MoTe₂ without and with XeF₂ treatment at 1.8 torr for 60 s; the black lines show the spectra of PES before XeF₂ treatment and the red lines show the spectra of PES after XeF₂ treatment at 1.8 torr for 60 s. (e) The valence edge of MoTe₂ before and after XeF₂ exposure and (f) the secondary electron cutoffs of MoTe₂ before and after XeF₂ exposure.

converted to a 1 L of MoTe₂ with an oxidized layer at 1.8 torr for 150 s. The 3 L of the MoTe₂ crystal structure disappeared after XeF₂ exposure, forming the remaining oxidized layer.

We fabricated MoTe₂ FETs by forming contacts in the XeF₂-exposed regions because the ultrathin oxidized layer of XeF₂-exposed MoTe₂ can be used as a buffer layer between MoTe₂ and the metal. Multilayer MoTe₂ with a thickness of ~ 3 nm was used for device fabrication. Two different types of contacts were fabricated on the same MoTe₂ flakes (ESI, Fig. S5† shows the detailed device fabrication process). Electrodes were patterned by e-beam lithography and exposed to XeF₂ for 60 s, followed by e-beam deposition of metals (Cr 1 nm/Pd 30 nm/Au 40 nm) to fabricate the contacts in the XeF₂-exposed regions. Cr is an adhesion layer; Pd can be connected to MoTe₂ for p-type FETs. Metals were also deposited on the surface of the pristine MoTe₂ for comparison. As shown in Fig. 5a, the MoTe₂ FET with conventional metal contacts showed a low field-effect hole mobility of $0.07 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ with an on/off current ratio of $\sim 10^4$, probably because of the high contact resistance (see the non-linear output curves ($I_{\text{ds}}-V_{\text{ds}}$) in the inset). Furthermore, the MoTe₂ FET with XeF₂-exposed contacts exhibited two orders of magnitude higher hole mobility ($6.31 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) with a higher on/off current ratio of $\sim 10^5$. Particularly, the linear output curves in the inset of Fig. 5b show the ohmic nature of

XeF₂-exposed contacts. The linear-scale transfer curves of the two devices in Fig. 5c clearly show that the on-current of the MoTe₂ FET with XeF₂-exposed contacts is significantly higher than that of the MoTe₂ FET with conventional metal contacts. The type and height of the Schottky barrier formed at the contact between the metal and the TMDs can determine the contact resistivity. However, the metal-insulator-semiconductor structure with an ultrathin insulating layer can substantially relieve the Fermi level pinning effect by reducing the direct metal contact to the interfacial states of MoTe₂.⁴⁹ The XeF₂-vapor process can chemically react with only a few topmost layers of TMDs, minimizing the degradation of underlying MoTe₂ crystallinity.³⁴ In addition, the oxidized layer induces p-doping of the underlying MoTe₂. Therefore, the ultrathin oxide interlayer enables overcoming the Schottky barrier induced by Fermi level pinning at the semiconductor-metal junction, reducing the contact resistance in the MoTe₂ devices (see the band diagram in Fig. 5d). The uniform oxide interlayer only with several nm-thickness can be easily and precisely produced through our method. In particular, the oxygen plasma treatment method was suitable for scale up, but uniform surface cannot be achieved due to ion bombardment on the MoTe₂ surface.³⁵ Laser treatment is also widely used to improve the properties of contact between the metal and MoTe₂,⁵⁰ but there



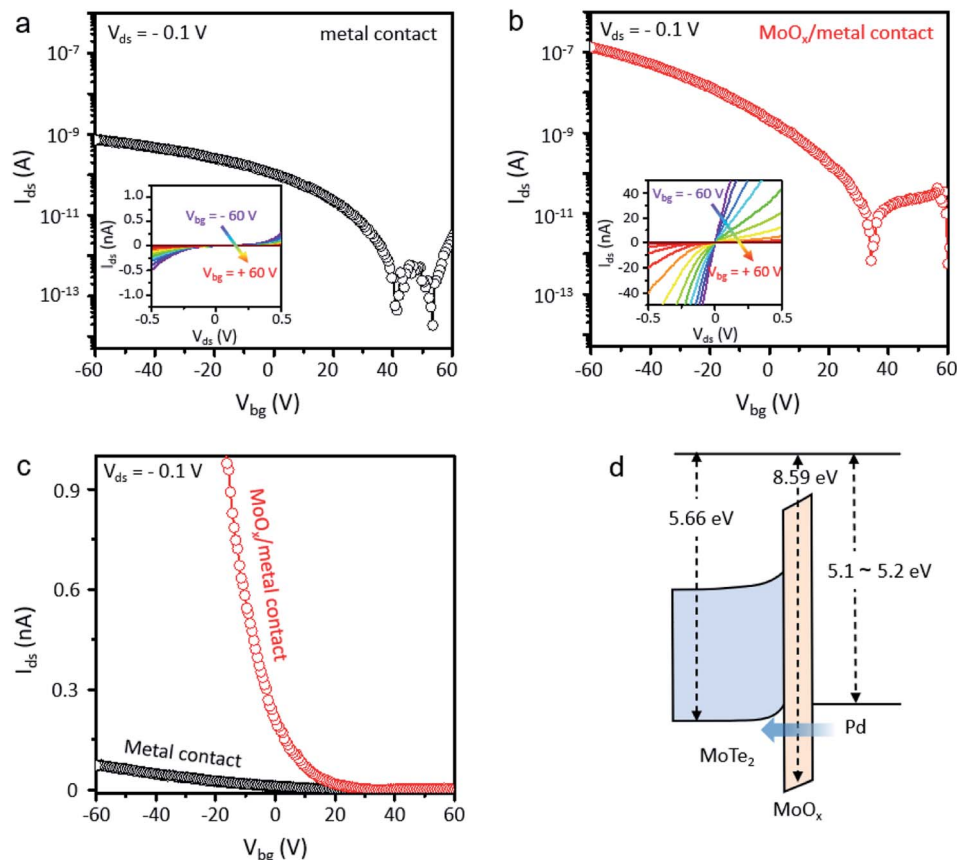


Fig. 5 (a) I_{ds} – V_g transfer characteristics of MoTe₂ FETs with and without the XeF₂ treatment process in the logarithmic scale. The black dotted line curve shows the transfer characteristics of the MoTe₂ channel with direct contact between the metal and MoTe₂. (b) The red dotted line curve shows the transfer characteristics of the MoTe₂ channel with contact between the metal and MoO_x/MoTe₂ in the logarithmic scale. The insets of Fig. 5a and b show I_{ds} – V_{ds} output characteristics of the MoTe₂ FET without XeF₂ treatment in the contact region (Fig. 5a) and with XeF₂ treatment in the contact region (Fig. 5b) before metal deposition. (c) I_{ds} – V_g transfer characteristics of MoTe₂ FETs both conventional metal contacts (black dots) and MoO_x/metal contacts (red dots) in the linear scale and (d) the schematic image of the band diagram for MoTe₂,¹⁵ oxidized layer, and Pd.^{21,51,52}

is a limitation to scale up. Therefore, our XeF₂-mediated oxidation technique is a promising candidate for fabricating high-performance electronic devices based on MoTe₂.

Conclusions

In conclusion, we demonstrate a controllable technique to produce a uniform oxidized layer on top of MoTe₂ by XeF₂ exposure. XeF₂-exposed MoTe₂ was formed with a thin and uniform oxidized layer (~2.5 nm-thick MoO_x) regardless of the exposure time (within ~120 s) because of the passivation effect and simultaneous etching. We could directly fabricate the ultra-smooth thin oxidized layer on MoTe₂ by the simple and easy XeF₂ vapor exposure method. In addition, the electrical properties of MoTe₂ FETs with and without XeF₂ treatment in the contact region were measured, including the hole mobility and on/off current ratio. The MoTe₂ FET with an ultra-smooth MoO_x interlayer shows a relatively high field-effect hole mobility of 6.31 cm² V^{−1} s^{−1} with a high on/off current ratio of ~10⁵. This result is two orders of magnitude higher than that of MoTe₂ FETs with conventional metal contacts (0.07 cm² V^{−1} s^{−1}). The

simplicity and applicability of our method will be useful for practical applications of MoTe₂ FETs in the future.

Experimental section

Sample preparation

MoTe₂ was mechanically exfoliated onto the SiO₂ (280 nm)/Si substrate after the substrate was cleaned with acetone and isopropyl alcohol (IPA) in an ultra-sonication bath for 10 min, respectively. The exfoliated MoTe₂ flakes were loaded into a XeF₂ chamber (ESI, Fig. S1†). The XeF₂ gas of high pressure (1.8 torr) was introduced into the chamber in a cyclic manner, where the chamber was filled with XeF₂ for different treatment times in a cycle and then pumped out.

Raman spectroscopy

The optical properties of MoTe₂ were observed through Raman spectroscopy (Renishaw, inVia), with a laser wavelength of 532 nm and a spot size of ~1 μm. Raman spectra at the same position on MoTe₂ were obtained before and after XeF₂ exposure.



Atomic force microscopy (AFM)

Atomic force microscopy (AFM, Park systems, NX10) was used to observe the surface morphology, roughness, and thickness of the MoTe₂ samples. The contact mode was used for exact thickness information. A silicon tip with an elastic constant of $\sim 0.2 \text{ N m}^{-1}$ was used in contact mode.

Transmission electron microscopy (TEM)

For cross-sectional TEM, a focused ion beam (FIB) system (Helios G4, Thermo Fisher Scientific, USA) was used. To protect the sample surface from e-beam, the XeF₂-exposed MoTe₂ flakes were covered with multilayer h-BN. We used TEM (JEM-F200, JEOL Ltd, Japan) with a low operating voltage of 80 keV. Transmission electron microscopy-energy dispersive X-ray spectroscopy (TEM-EDS) was used to analyze the atomic ratios.

Photoemission spectroscopy (PES)

MoTe₂ crystals were prepared on an Au-coated SiO₂ substrate. Photoelectron spectroscopy (PES) spectra of the sample were recorded at $h\nu = 780 \text{ eV}$. In addition, valence spectra and secondary electron cutoff were measured using a photon energy of $h\nu = 90 \text{ eV}$. Synchrotron-based PES spectroscopy was performed at room temperature at the 4D beamline in the Pohang Light Source II (PLS II), Korea.

Device fabrication and electrical measurements

MoTe₂ field-effect transistors (FETs) were fabricated on a SiO₂ (285 nm)/Si substrate using e-beam lithography (EBL, TESCAN). Metal contacts were deposited by using an e-beam evaporator (Temescal six pocket e-beam evaporation systems). The MoTe₂ FETs were measured by using a semiconductor parameter analyzer (Keithley 4200). The source-drain current (I_{ds}) was measured with a fixed source-drain voltage (V_{ds}) of -0.1 V and varying back-gate voltage (V_{bg}) from -60 V to 60 V . The field-effect mobility (μ_{FE}) was calculated, extracting from the linear area of the transfer curve. The equation is $\mu_{\text{FE}} = [(\Delta I_{\text{ds}}/\Delta V_{\text{bg}})(L/W)V_{\text{ds}}]/C_{\text{ox}}$, where Δ , W , L , and C_{ox} are the gradient of I_{ds} to V_{ds} , channel width, length and gate capacitance of SiO₂ (12.1 nF cm^{-2} for a SiO₂ thickness of 285 nm).

Author contributions

E. J. and J. H. K. contributed equally. E. J., J. H. K., and G. -H. L. designed this work. E. J. and J. H. K. performed all experiments including AFM, Raman spectroscopy, PES, and STEM analysis. W. L. performed the Xenon difluoride (XeF₂) experiments. K. I performed the photoemission spectroscopy (PES) system. E. J. and J.-C. S fabricated the device and analyzed device performance. All authors discussed the results. E. J. and J. H. K. wrote the manuscript together. J.-W. P, H. S and G.-H. L reviewed and edited the manuscript together. All authors have given approval to the final version of the manuscript.

Conflicts of interest

The authors declare no competing financial interest.

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