

MoTe₂-Based Schottky Barrier Photodiode Enabled by Contact Engineering

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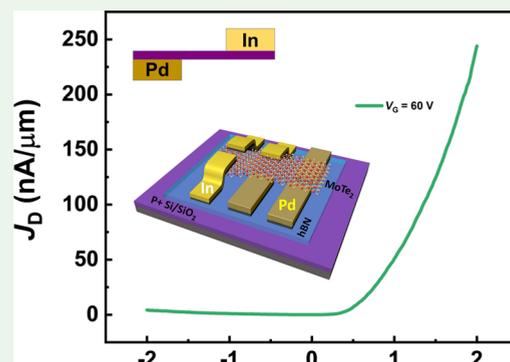
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Supporting Information

ABSTRACT: Two-dimensional (2D) atomic crystalline materials have attracted the scientific community owing to their remarkable electrical and optical applications, such as solar cells, light-emitting diodes, and photodiodes. This study demonstrates a Schottky barrier photodiode (SPD) using different metal architectures in lateral and vertical contacts on n-type 2H phase semiconducting molybdenum ditelluride (MoTe₂) synthesized by the self-flux crystal growth method. High-work-function palladium and low-work-function indium metals have been deposited on MoTe₂ to fabricate a field-effect transistor confirming diode characteristics. The device shows an ideality factor of 1.09 and a rectification ratio of 10², indicating ideal diode characteristics based on a single MoTe₂ channel. In addition, we measured the device in the dark and used the green laser to analyze the photodiode behavior of SPD in a wide range of light intensity. A single channel using contact architecture-based study is helpful to apply in other 2D materials to achieve the simplest fundamental diodes for future nanoelectrical and optoelectronic devices.

KEYWORDS: Schottky photodiode, MoTe₂, flux method, contact engineering, p-n junction



1. INTRODUCTION

Transition metal dichalcogenides (TMDs) have become the focus of material class for intense research due to their exceptional physical and chemical features, such as thickness-dependent electronic properties, free dangling bond surface, mechanical flexibility, and excellent intrinsic carrier mobility.^{1–4} TMDs, due to their flexibility, are stacked in different configurations to fabricate devices, allowing for modifying the physical properties based on the structure for future applications.^{5,6} As a result, they have received a lot of interest in electronic and optoelectronic applications, such as field-effect transistors (FETs), spintronics, solar cells, light-emitting diodes, and photodetectors.^{7–11} Specifically, extensive studies have been done, where TMD diodes are stacked in p- and n-type materials for a variety of applications as switching devices, photodetectors, and self-powered sensors, by producing depletion regions to rectify unidirectional current.^{12–14} Among them, 2H phase semiconducting molybdenum ditelluride (MoTe₂) exhibits a relatively low band gap such as a direct optical band gap of 1.02 eV in a monolayer. In comparison, its multilayered form shows an indirect band gap of 0.85 eV, which benefits it in a broader spectrum response.^{15–17} In addition, MoTe₂ absorbs light from visible to near-infrared wavelengths, making it an excellent material for photoresponse behavior.¹⁸ The Schottky barrier photodiode (SPD) is a structure that does not stack in a

heterostructure and consumes less power than conventional p-n diodes due to its rapid response and minimal voltage drop in the forward direction.¹⁹ Studies have been reported that revealed exceptional diode properties in two-dimensional (2D) p-n junction heterostructures. However, the SPD based on contact engineering is yet to be explored extensively for practical use.²⁰

In this work, a single-channel MoTe₂-based FET device is fabricated in a lateral and vertical structure using high and low work function metal contact to study the comparative electrical charge transport properties. The designed electrodes consist of direct palladium (Pd) contact with MoTe₂ on the bottom and indium (In) on the top of the channel covered by gold (Au) metal layers to avoid degradation due to oxidation. The electrical charge transport properties are investigated in lateral and vertical structures, such as on bottom contacts (Pd–Pd), top contacts (In–In), and from top to bottom contacts (In–Pd) in an intrinsic MoTe₂ FET. In the lateral structures, the device shows ohmic and Schottky contacts, whereas the

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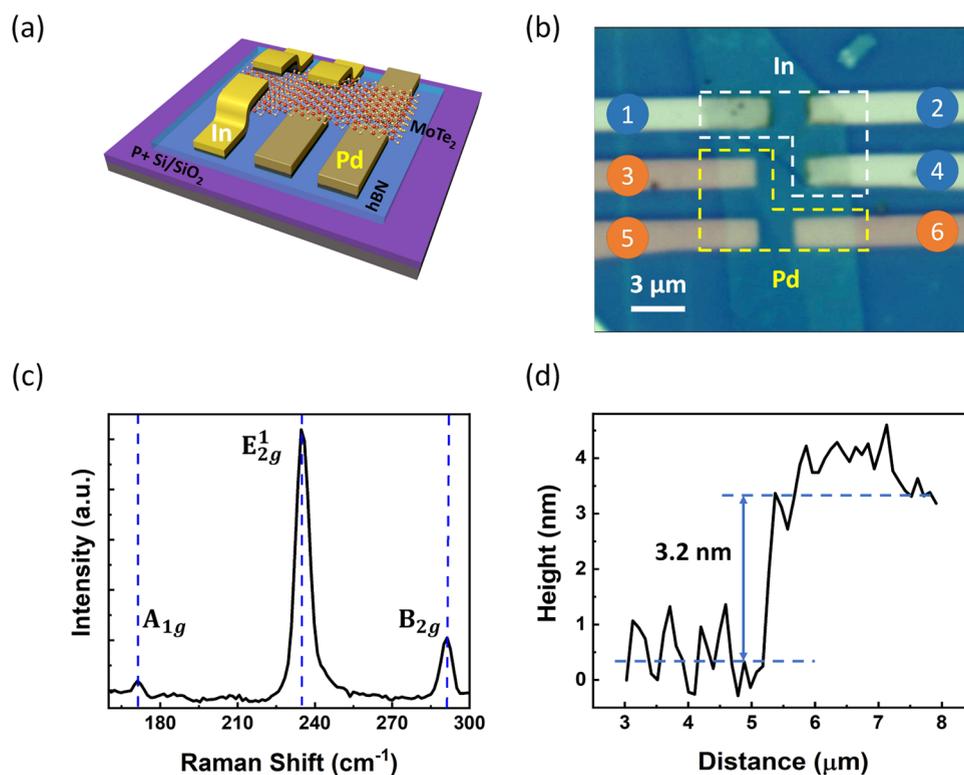


Figure 1. Lateral and vertical metal contact engineering in the MoTe₂ FET. (a) Schematic illustration of the MoTe₂ SPD device. (b) Optical image of the MoTe₂ SPD device with top In and bottom Pd metal contacts. (c) Raman spectra obtained from the MoTe₂ flake with atomic displacements showing vibrational modes. (d) AFM line profile shows the thickness of the MoTe₂ flake.

vertical structure across top–bottom contacts shows p–n junction characteristics. In fabrication, Pd metal with high work function ($\Phi = 5.6$ eV)²¹ due to its good air stability and resistance to oxidation is deposited on the bottom of a mechanically exfoliated multilayer MoTe₂, while In metal with low work function ($\Phi = 4.1$ eV)²² is deposited on the top surface of the flake, which exhibits as an ideal-like diode upon measuring from top to bottom contacts. Uneven metal contacts resulted in an ideality factor of 1.09, and a rectification ratio of up to 10^2 was observed at room temperature. Unlike complex p–n junction diodes, our simplified structure can be applied to compare charge transport properties at various temperatures and photodiode behavior, such as mobility, ideality factor, and tunneling barrier height.

2. RESULTS AND DISCUSSION

Figure 1a illustrates the schematic diagram of MoTe₂ SPD, whereas Figure 1b shows the optical image of the device on which Pd high-work-function electrodes are designed to the bottom and In low-work-function electrodes on the top of the multilayered MoTe₂. First, thick hexagonal boron nitride (h-BN) was transferred on a precleaned Si/SiO₂ 1×1 cm² wafer to prevent the leakage current and interface traps.²³ Pd owing to its stability in the air was patterned on h-BN and mechanically exfoliated multilayered flakes from our grown MoTe₂ bulk crystals transferred onto prepatterned Pd electrodes using a dry transfer method with poly-(dimethylsiloxane) (PDMS) stamp. Later, In electrodes were designed on the top surface of the MoTe₂ multilayered channel. To obtain Raman spectra of the n-type MoTe₂ flake, we used the excitation laser of 532 nm wavelength in an ambient environment at room temperature.

The Raman spectrum in Figure 1c shows three peaks of MoTe₂: A_{1g} mode at a peak value of 170 cm⁻¹, E_{2g}¹ mode at a peak value of 234 cm⁻¹, and B_{2g}¹ mode at a peak value of 289 cm⁻¹.²⁴ Atomic force microscopy (AFM) was performed to confirm the layer thickness and the number of layers of the MoTe₂ flake, as shown in Figure 1d. The layer thickness of MoTe₂ was 3.2 nm, confirming that the MoTe₂ flake was approximately four layers,²⁵ as shown by drawing a white line in Figure S2.

The output characteristics normalized by the channel width of In–In contacts on MoTe₂ at various gate biases are shown in Figure 2a, which shows an ohmic-like junction with low resistance based on the linear output characteristics. On the other hand, bottom (Pd–Pd) contacts have nonlinear output characteristics, generating Schottky junctions with high resistive contacts, as shown in Figure 2c. The transfer characteristics normalized by the channel width of the lateral metal–MoTe₂–metal with top In–In contact and bottom Pd–Pd contact are shown in Figure 2b,d as a function of the drain voltages. The device demonstrated ambipolar behavior with n-type electron dominant features for both connections at different drain voltages. Due to the difference in Fermi levels, when an n-type semiconductor meets metal with a higher work function, the electrons of the semiconductor diffuse into the metal with a lower energy state, generating a depletion area until aligned Fermi level. Thus, a potential is created, which blocks carrier injection into metal and forms the Schottky junction. On the other hand, when the n-type semiconductor meets low-work-function metal, it starts aligning the Fermi level and the electrons move from semiconductor to metal due to its lower work function than metal, forming ohmic contact.^{26,27} The field-effect mobility (μ_{FE}) of the MoTe₂

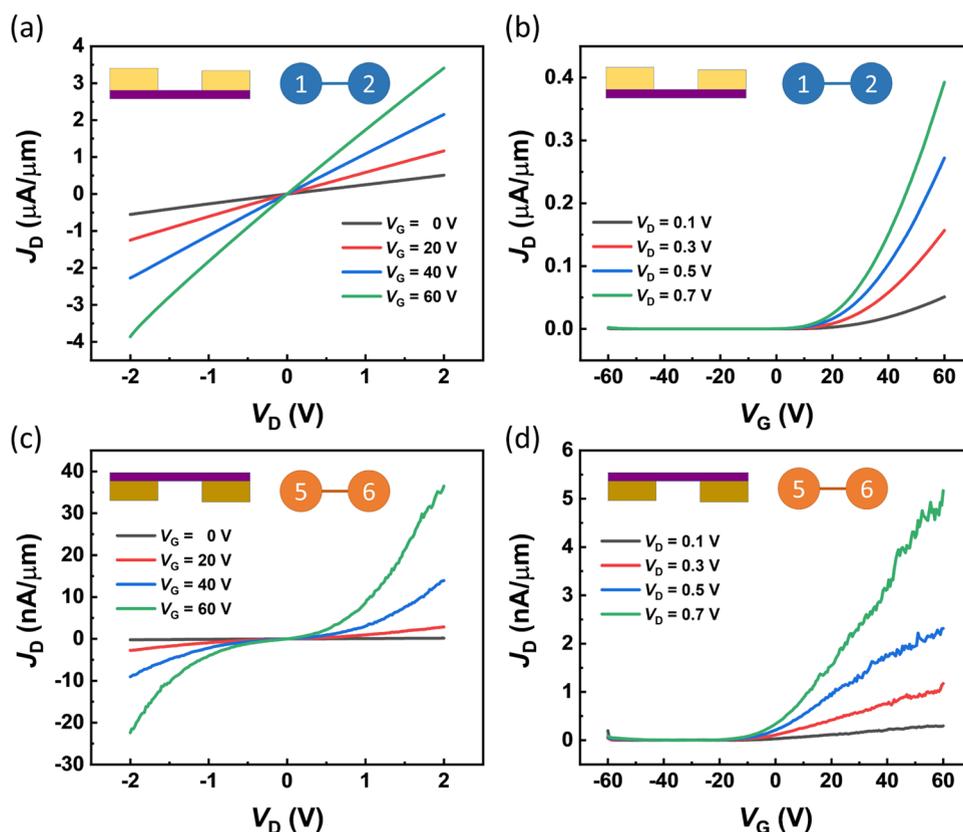


Figure 2. Comparison plot of output and transfer characteristics normalized by the channel width for different electrode architectures at a temperature of 300 K. (a) Output characteristics of In–In contact at the top of the MoTe₂ single channel depending on different gate biases. (b) Transfer characteristic plot of the MoTe₂ single channel at different drain voltages. (c) Output characteristic plot of Pd–Pd contact at the top of the MoTe₂ single channel depending on different gate biases. (d) Transfer characteristic plot of the MoTe₂ single channel at different drain voltages.

device with different contact architectures (In–In, Pd–Pd, and In–Pd) is calculated using $\mu_{FE} = g_m \times \left[\frac{L}{WC_{ox}V_{DS}} \right]$, where $g_m = (dI_{DS}/dV_G)$ is the transconductance, L is the channel length, W is the width, C_{ox} is the capacitance of 285 nm thick SiO₂, I_D is the source–drain current, V_D represents the drain voltage, and V_G is the gate voltage.²⁸ The electron mobility at room temperature in the lateral structure for In–In contact is 60.04 cm² V^{−1} s^{−1} with a current on/off ratio of 10⁵ at $V_D = 0.1$ V, while for Pd–Pd connection, the electron mobility is found to be 0.31 cm² V^{−1} s^{−1} with a current on/off ratio of 10². By contrast, the electron mobility in the vertical structure for In–Pd contacts is obtained at 0.17 cm² V^{−1} s^{−1} with a current on/off ratio of 10³. Thus, our findings suggest that the electrical connection is much improved in the top contact with In–MoTe₂ instead of Pd–MoTe₂. It can be referred to as the good ohmic contact formation at the metal–semiconductor junction due to the low-work-function value of In and n-type dominant nature of MoTe₂. The on/off ratios for lateral and vertical structures are extracted from the logarithmic scale of transfer characteristic curves as shown in Figure S3.

At the metal–semiconductor interface, a charge carrier hinders by a barrier that includes a major carrier transport mechanism explained by thermionic emission theory.^{29–32} In Figure 3a, the output characteristics normalized by the channel width (inset: logarithmic scale) of In and Pd electrodes show strong rectification behavior as a function of gate biases ranging from 0 to 60 V. At the Pd semiconductor junction, a

high barrier is formed in the reverse direction of the diode. While in contact with low work function, the In semiconductor junction forms an ohmic junction with MoTe₂ in the diode’s forward direction. Thus, a high resistive barrier at the junction of Pd exists, and the MoTe₂ device exhibits an ideal diode behavior. Figure 3b shows the transfer characteristics normalized by the channel width for different metal architectures, showing an n-type MoTe₂ behavior and an increase in the current varying drain voltages at room temperature. Figure 3c shows the logarithmic plot of output characteristics at room temperature. The MoTe₂ device exhibits rectifying behavior with increased drain current by increasing the gate voltage. The rectifying performance of the Schottky diode in different metal architectures can be estimated by evaluating the ideality factor (η), which can be determined using the Shockley diode equation.^{33,34} The ideality factor can be figured out from a plot of the natural logarithm of the diode current vs drain voltage by taking the slope, which gives $e/\eta k_B T$.^{19,20} In our device, the lowest ideality factor is found to be 1.09 at room temperature, which is close to unity. The ideal diode-like behavior can be obtained if the ideality factor is closer to unity. The ideality factor increases upon the decrease in current as the forward current is mainly based on the diffusion of electrons. The ideality factor is strongly temperature-dependent, which is calculated at the slope of forward-biased region temperature ranging from 77 to 300 K. Figure 3d shows the variation in ideality factor with a decrease in temperature from 300 to 77 K. Our device has a

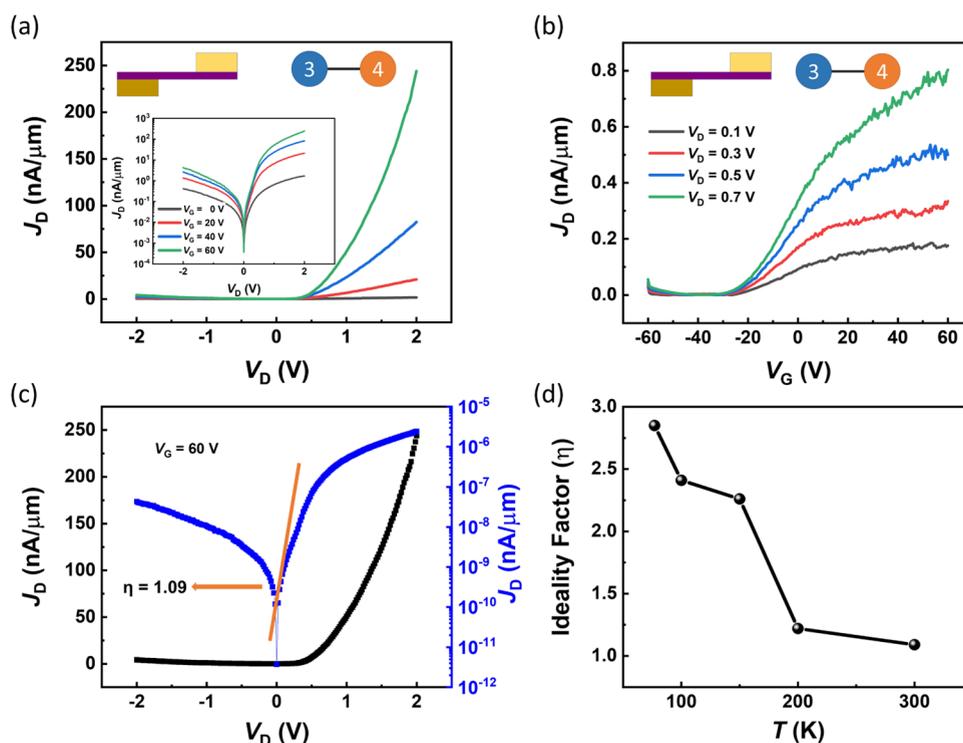


Figure 3. Plot of SPD behavior of the MoTe₂-based device at a temperature of 300 K. (a) Output characteristics normalized by the channel width of the MoTe₂ device on the top to bottom contact as a function of gate bias. Inset: Logarithmic scale of output characteristics at various gate biases. (b) Transfer characteristics normalized by the channel width of the MoTe₂ device on the top to bottom contact at different drain voltages. (c) Plot of the natural logarithm of the diode current vs drain voltage to determine the ideality factor. (d) Ideality factor of the SPD as a function of temperature.

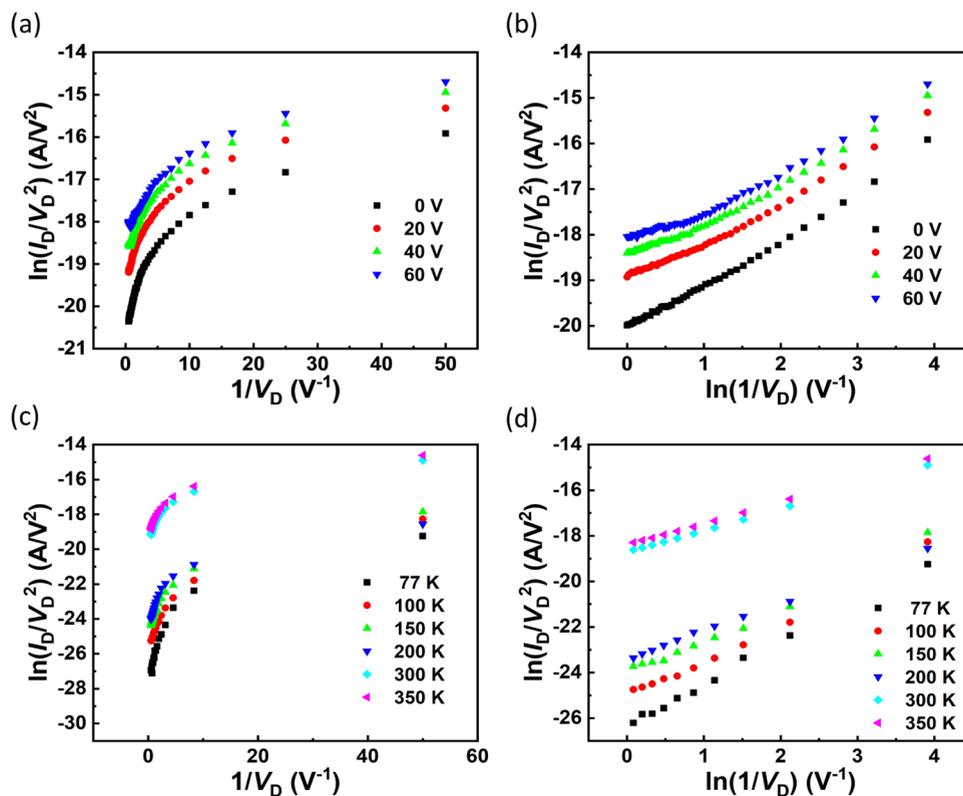


Figure 4. (a) Plot of $\ln(I/V_D^2)$ as a function of $(1/V_D)$ illustration of the DT behavior of the device at various gate biases at room temperature. (b) Plot of $\ln(I/V_D^2)$ vs $\ln(1/V_D)$ at different gate biases. (c) Plot of $\ln(I/V_D^2)$ as a function of $(1/V_D)$ illustration of the DT behavior of the device at $V_G = 60$ V. (d) Plot of $\ln(I/V_D^2)$ vs $\ln(1/V_D)$ at different temperatures showing linear relation of DT of the device.

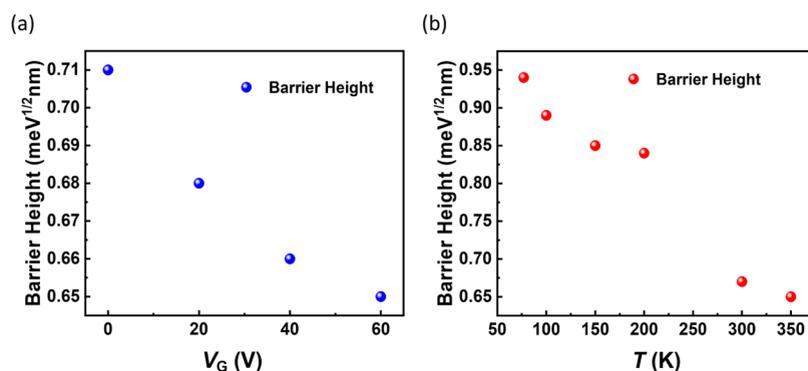


Figure 5. Barrier parameter calculation. (a) Tunneling barrier height of the MoTe₂ device as a function of gate bias at a temperature of 300 K. (b) Tunneling barrier height of the MoTe₂ device as a function of temperature.

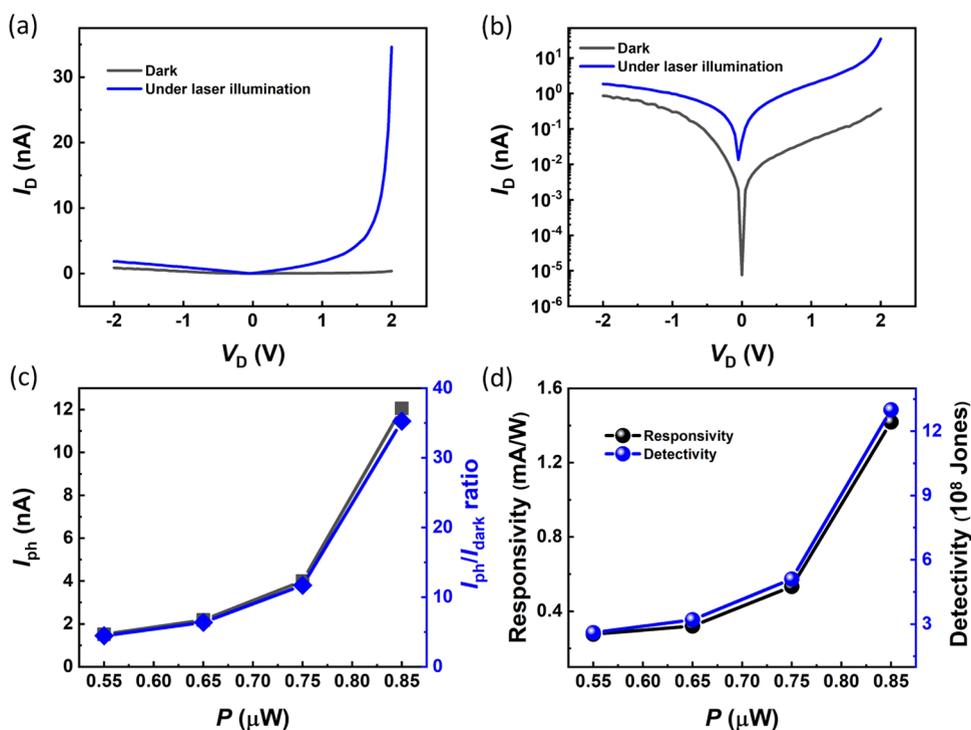


Figure 6. Photoresponse of the MoTe₂ SPD at room temperature in an ambient environment (a) Output characteristic plot of the MoTe₂ device in the top-to-bottom contact under the $\lambda = 520$ nm green laser illumination and dark conditions. (b) Logarithmic scale of output characteristic plots of the MoTe₂ device. (c) Plot of the photocurrent (I_{ph}) and ($I_{\text{ph}}/I_{\text{dark}}$) ratio as a function of power. (d) Plot of the responsivity and detectivity of the MoTe₂ SPD as a function of power.

lower ideality factor of 1.09, which is close to unity at 300 K, while a higher ideality factor of 2.85 at a lower temperature of 77 K, showing strong dependence on temperature, as shown in Figure S4. We refer to the higher ideality factor at low temperatures as the current through the metal–semiconductor junction depends upon temperature and electrons only can overwhelm the lower barrier resulting in deviation in ideality.

To explore the vertical charge transport, we measured our device in a range of temperatures from 77 to 350 K shown in Figures S5 and S6, as both contact resistance and interlayer resistance effectively contribute to the total resistance. Our device shows Schottky contact at the barrier interface, and charge transfer across the barrier is dominant due to the tunneling behavior. At low bias voltages, direct tunneling (DT) is dominated, which can be approximated by the following equation^{35–38}

$$I = \frac{Aq^2V\sqrt{2m^*\phi_B}}{h^2d} \exp\left(-\frac{4\pi d\sqrt{2m^*\phi_B}}{h}\right) \quad (1)$$

At higher bias voltages, the probability of tunneling is increased as the barrier is reshaped from a trapezoid to a triangle, where the Fowler–Nordheim tunneling (FNT) is dominant, which can be described by the following equation^{35–38}

$$I = \frac{Aq^3m_0V^2\sqrt{2m^*\phi_B}}{8\pi h\phi_B d^2 m^*} \exp\left(-\frac{8\pi d\sqrt{2m^*\phi_B}^{3/2}}{3hqV}\right) \quad (2)$$

In eqs 1 and 2, ϕ_B is the tunneling barrier height, m_0 is the rest electron mass, m^* ($0.46 m_0$)³² is the effective mass of electrons in the MoTe₂ flake, q is an electronic charge, h is Planck's constant, A is the electrical contact area, and d is the

width of the interface barrier. The plot of $\ln(I/V_D^2)$ vs $(1/V_D)$ curves shown in Figure 4a,c illustrated two different plots at various gate voltages and temperatures ranging from 77 to 350 K. The device exhibits exponential behavior, indicating that DT is the dominant charge carrier mechanism, while FNT is not observed. However, the DT and FNT are observed at a transition voltage of $V_T = 1.1$ V by measuring the device in an ambient environment under green laser illumination using the wavelength of 520 nm shown in Figure S7. To further confirm the DT mechanism, we plotted $\ln(I/V_D^2)$ as a function of $\ln(1/V_D)$, where the plots illustrated the linear relation as shown in Figure 4b,d. Furthermore, it is used to obtain the DT barrier parameters $d\sqrt{(\phi_B)}$ of the SPD device by linear fitting. Figure 5a shows the tunneling barrier height vs the gate voltage of the SPD device. The tunneling barrier height has a decreasing trend and varies in the range of (0.71–0.65 meV^{1/2}) nm.

Similarly, a decrease in tunneling barrier height in the range of (0.94–0.65) meV^{1/2} nm is observed depending on the temperature range from 77 to 350 K. High tunneling barrier height can be seen at a low temperature (77 K) while low tunneling barrier height is observed at a high temperature (350 K) as shown in Figure 5b. The above results illustrate that tunneling barrier height depends upon gate voltage and temperature. We refer to these variations in barrier parameters as the inhomogeneity and interfacial surface defects that are present at the metal–semiconductor interface.¹⁹

In addition, we demonstrated the photodiode behavior of the MoTe₂ device under green laser light illumination with a wavelength (λ) of 520 nm and a wide range of power intensity in the top to bottom contacts (In–Pd). Under the illumination of green laser light of a wide range of power intensities, the SPD shows good photodiode behavior as shown in Figure 6a,b. The responsivity and detectivity are the key parameters that were evaluated for the MoTe₂ SPD device at $V_D = 1$ V. The responsivity is defined as $R = I_{ph}/P$, in which P is the power of incident illumination, and I_{ph} is the photocurrent, which is calculated by the equation: $I_{ph} = I_{light} - I_{dark}$, where I_{light} and I_{dark} are the current measured under light illumination and dark conditions. The photocurrent is increasing with an increase in incident laser power intensity as shown in Figure 6c. The detectivity is defined by the equation $D = R/\sqrt{2qI_{dark}}$, where both the responsivity and detectivity are estimated as 1.4 mA W⁻¹ and 13.5×10^8 Jones, respectively, at $P = 0.85 \mu\text{W}$ under the $\lambda = 520$ nm green laser light as shown in Figure 6d. Moreover, the device was measured in an ambient environment due to the limitation of the facility. Thus, the polarity was changed from n-type to p-type, as shown in Figure S8. As earlier published reports have revealed that deep ultraviolet irradiations, electrostatic gating, and chemical doping of the TMDs can change the intrinsic operation of the device.^{39–42}

However, in our device, we refer to polarity conversion to the hole doping in the air as the energy level difference that exists between MoTe₂ (4.1 eV) and oxygen/water redox couple (4.83 eV) due to which the electrons are injected from MoTe₂ to oxygen/water redox couple.⁴³

3. CONCLUSIONS

In summary, we have fabricated FETs using different metal architectures in lateral and vertical contacts on n-type MoTe₂ grown by the self-flux method to study electrical charge transport properties. Due to the difference in the work function

of deposited metal electrodes, the electrical charge transport in the top to bottom contacts at various gate biases confirmed ideal-like diode characteristics giving an ideality factor of 1.09, which is close to the unity. This value demonstrates an ideal-like diode behavior obtained in a simple single channel regardless of the complex diode structure that usually requires a heterostructure based on p- and n-type materials. The device shows excellent electrical properties on the top contacts between (In–In) contacts over a wide range of temperatures. In addition, the device shows a stable photodiode response to the green laser light, confirming the application of a single-channel diode for future electrical and optoelectronic devices.

4. EXPERIMENTAL METHODS

4.1. MoTe₂ Crystals Grown by the Self-Flux Method. The self-flux method is used to grow MoTe₂ crystals, and tellurium was used as a self-flux material and a reactive agent as shown in Figure S1a. The growth of exceptionally pure crystals is made possible by the flux material having a lower melting point, which aids in dissolving solute for the chemical reaction and crystallization process. Removing the flux material from the grown crystals requires immediate centrifugation. The source materials molybdenum powder of purity 99.99% (Sigma Aldrich) and tellurium 99.999% (Alfa Aesar) were measured and added into an alumina crucible fixed into a quartz tube ampule in Argon (Ar) environment inside a glovebox. A cylindrical shape of high purity 99.99% SiO₂ quartz wool with excellent insulation performance was also loaded above one cm of alumina crucible to absorb excessive solvent in centrifugation. It is then placed into a high-temperature furnace while being vacuum sealed. The furnace temperature program was set at 1100 °C as a ramp-up temperature at 50 °C h⁻¹. The temperature dwelling time was set for 36 h, and then, the ampule was allowed to cool down at 2.5 °C h⁻¹ until it reached 550 °C. The resultant ampule was centrifuged at 2000 rpm for 120 s to the obtained large size of about 0.6 cm crystals as shown in Figure S1b. X-ray diffraction was used to characterize the grown crystals to confirm crystal formation and quality as shown in Figure S1c. The formed crystals were then used for device fabrication.

4.2. Device Fabrication. Using the photolithography method, a highly p-doped 285 nm Si/SiO₂ substrate was used to draw an outer electrode pattern, and Au/In metal layers were deposited using an electron beam evaporator before being lifted off in acetone. To prevent leakage current and lessen the scattering, a thick mechanically exfoliated h-BN was transferred using the traditional PDMS stamping method onto a prepatterned substrate. The sample was then cleaned with acetone and isopropyl alcohol, submerged in chloroform and dried using a nitrogen gun. Next, poly(methyl methacrylate) was spin-coated onto the sample and using an e-beam lithography technique, the inner bottom electrodes were drawn on h-BN. After that, Pd/In electrodes were kept at a thickness of 10/5 nm using an electron beam evaporator since Pd exhibits high resistance to oxidation under ambient conditions. Mechanically exfoliated MoTe₂ flake was transferred onto metal electrodes by the dry transfer method. Again, metal layers of Au/In 30/10 nm were then deposited onto the MoTe₂ flake as top electrodes.

4.3. Device Characterization. X-ray diffraction (XRD) was performed to confirm the MoTe₂ crystal growth. After device fabrication, the thickness of MoTe₂ was analyzed by AFM in noncontact mode. The Raman analysis of MoTe₂ was performed using a laser wavelength of 532 nm under ambient conditions to characterize the flake quality and surface.

4.4. Electrical and Photo Measurements. The electrical transport measurement of the MoTe₂ device was performed under a vacuum using a Keithley 4200-SCS parameter analyzer. Photo measurement was performed in the dark and using a 520 nm green laser in the ambient environment using different power intensities.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsanm.2c04569>.

Crystal growth by the self-flux method; AFM image and line scan profile for MoTe₂; electrical characterization of the MoTe₂ device at 300 K; ideality factor of MoTe₂ SPDs at different temperatures; temperature-dependent electrical characterization; FNT and DT behavior; and optical characteristics (PDF)

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Notes

The authors declare no competing financial interest.

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■ ABBREVIATIONS

2D, two-dimensional
SPD, Schottky barrier photodiode
MoTe₂, molybdenum ditelluride
TMDs, transition metal dichalcogenides
FETs, field-effect transistors
Pd, palladium
In, indium
Au, gold
h-BN, hexagonal boron nitride
PDMS, poly(dimethylsiloxane)
AFM, atomic force microscopy
 μ_{FE} , field-effect mobility

(dI_{DS}/dV_G), transconductance
 L , channel length
 W , width
 C_{ox} , capacitance of 285 nm thick SiO₂
 I_D , source–drain current
 V_D , drain voltage
 V_G , gate voltage
 η , ideality factor
DT, direct tunneling
FNT, Fowler–Nordheim tunneling
 ϕ_B , tunneling barrier height
 m_0 , rest electron mass
 m^* , effective mass of electrons
 q , electronic charge
 h , Planck's constant
 d , width of the interface barrier
 λ , wavelength

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