Highly Sensitive, Ultrafast, and Broadband Photo-Detecting Field-Effect Transistor with Transition-Metal Dichalcogenide van der Waals Heterostructures of MoTe$_2$ and PdSe$_2$

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Recently, van der Waals heterostructures (vdWHs) based on transition-metal dichalcogenides (TMDs) have attracted significant attention owing to their superior capabilities and multiple functionalities. Herein, a novel vdWH field-effect transistor (FET) composed of molybdenum ditelluride (MoTe$_2$) and palladium diselenide (PdSe$_2$) is studied for highly sensitive photodetection performance in the broad visible and near-infrared (VNIR) region. A high rectification ratio of $6.3 \times 10^5$ is obtained, stemming from the sharp interface and low Schottky barriers of the MoTe$_2$/PdSe$_2$ vdWHs. It is also successfully demonstrated that the vdWH FET exhibits highly sensitive photo-detecting abilities, such as noticeably high photoresponsivity ($1.24 \times 10^5$ A W$^{-1}$), specific detectivity ($2.42 \times 10^{14}$ Jones), and good external quantum efficiency ($3.5 \times 10^6$), not only due to the intra-TMD band-to-band transition but also due to the inter-TMD charge transfer (CT) transition. Further, rapid rise (16.1 $\mu$s) and decay (31.1 $\mu$s) times are obtained under incident light with a wavelength of 2000 nm due to the CT transition, representing an outcome one order of magnitude faster than values currently in the literature. Such TMD-based vdWH FETs would improve the photo-gating characteristics and provide a platform for the realization of a highly sensitive photodetector in the broad VNIR region.

1. Introduction

Highly sensitive photodetectors based on semiconducting materials as light-absorbing and charge-transporting materials in either the visible and infrared (IR) spectral ranges are extensively investigated in relation to environment monitoring, thermal imaging, gas sensing, and imaging optoelectronic devices due to their small volumes and compatibility with on-chip integration processes. For example, silicon-based metal-oxide-semiconductor (Si-MOS) field-effect transistor (FET) photodetectors have attracted the interest of the semiconductor industry over the past few decades. However, given the continuous decrease in their size and the exponential increase in the density of the transistors, the performance capabilities of Si-MOSFETs have started to degrade due to short channel effects, thus limiting further size reductions.

More recently, graphene, a representative 2D material, has attracted significant

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The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/advs.202003713

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DOI: 10.1002/advs.202003713
attention owing to its fascinating electronic, thermal, and mechanical properties. Its ultrahigh charge carrier mobility (>200 000 cm² V⁻¹ s⁻¹) offers the potential for developing fast electronic devices. Ultrahigh mobility, however, arises from its zero-bandgap characteristic, leading to low ON/OFF current ratios of graphene-based transistors, which limit the wider application of this material. Thus, it is necessary to obtain a new 2D material with a wide tunable bandgap so as to realize multifunctional nanoscale optoelectronic devices. Such a requirement has led researchers to investigate various transition-metal dichalcogenides (TMDs) and other 2D materials in addition to graphene. Among them, black phosphorus (BP), an intrinsically p-type TMD material, has been investigated due to its high hole mobility and ON/OFF current ratio. However, BP degrades easily under ambient air conditions, limiting its prospects in practical applications.

Recently, molybdenum ditelluride (MoTe₂), another p-type TMD semiconductor material, has attracted the attention of researchers owing to its remarkable electrical and optical properties. MoTe₂ has an indirect bandgap of 0.88 eV in a high-quality multilayered bulk form, while it also has a direct optical bandgap of 1.10 eV in a monolayer structure. It has thus tremendous potential for use in the construction of p-channel MOSFETs and for integration with MoS₂, n-channel MOSFETs, but it has limited applications in near-infrared (NIR) photodetection due to its broad bandgap. To overcome such constraints and to promote the high performance of MoTe₂ in optoelectronics, several different approaches have been investigated. In relation to this, Xie et al. introduced a BP/MoTe₂ junction to improve the photocurrent characteristics and observed relatively small values of the photoresponsivity (R ≈ 0.2 A W⁻¹) and external quantum efficiency (EQE ≈ 48.1%) with a slow response time (≈2 ms). These small values may be caused by BP, which is a critical material that easily oxidizes.

Recent advances have seen the discovery of another noble TMD semiconductor material, monochalcogenide palladium diselenide (PdSe₂), which has potential applications for IR photodetection due to its small bandgap. PdSe₂ exhibits a strong thickness-dependent bandgap of 1.4–0 eV, with a monolayer of PdSe₂ having a bandgap of ≈1.4 eV as well as bulk PdSe₂, which is metallic, having a bandgap approaching 0 eV. Specifically, it has a pentagonal puckered morphology which promotes stability in air and stable performance. PdSe₂ also has many unique electrical, photocurrent, and thermal characteristics as compared to conventional 2D graphene and other TMD materials. Thus, it is considered to be an extraordinary material for optoelectronic applications in the visible and NIR regions owing to its high mobility with a narrow and tunable bandgap.

Meanwhile, to fabricate efficacious electronic and optoelectronic devices, an exceptional approach related to van der Waals heterostructures (vdWHs) based on TMD semiconductors has been devised. Emerging devices based on TMD vdWHs have been successfully implemented in FETs, gas sensors, memory devices, high-speed integrated circuits, energy storage devices, diodes, inverters, negative differential resistance components, amplifiers, spin-FETs, and in water splitting. Further, this compact system of TMD vdWHs offers a new paradigm in which to overcome the limitations of conventional MOS heterostructures, especially for the engineering of state-of-the-art (opto)electronic devices such as (photo-)FETs, (optical) sensors, (photo-)MOSFETs, and photodetectors that operate in the visible-near-infrared (VNIR) range (400–1000 nm). TMD vdWHs p-n diodes are preferred over TMD homojunction-based p-n diodes owing to their high performance capabilities. Various methods have been used for the fabrication of TMD homojunction p-n diodes, including chemical doping for Fermi-level pinning, electrostatic doping to regulate the density of the carriers, and the creation of specific metal contacts to fabricate p-type semiconductors. All of these methods deteriorate the performance capabilities of devices and degrade the charge carrier density.

In such vdWH (opto)electronic devices, the photocurrent characteristics are highly enhanced up to a certain level owing to their sharp interfaces, tunneling mechanism, and band alignments. However, large Schottky contact barrier heights (φₛ) between the metal contact and the TMD material (metal-TMD heterojunction) obstruct the transport of photo-excited charge carriers, which leads to the suppression of photocurrent generation. From this perspective, the role of φₛ at the metal-TMD junction should be emphasized to enhance the photocurrent characteristics. Hence, the successful use of TMD vdWHs as high-performance VNIR photodetectors with highly improved characteristics, such as the photoresponsivity R and specific detectivity (D*), is yet to be realized.

In this work, we demonstrate for the first time a novel p-MoTe₂/n-PdSe₂ vdWH FET for highly sensitive and broadband photo-detecting performance. A large rectification ratio (RR) of 6.3 × 10⁵ is achieved through the formation of Ohmic contact. Further, the photocurrent properties of the MoTe₂/PdSe₂ FET were studied under incident laser light with different input powers in a broad VNIR region (λ = 405–2000 nm). The key parameters of the photodetectors, in this case R, D*, EQE, and the response times, which can be tuned by applying back-gate voltage (Vbg) and explained based on the optical transition in terms of not only the intra-TMD band-to-band transition but also inter-TMD charge-transfer (CT) transition, were also evaluated. Such high gate-modulated photo-detecting characteristics with the rapid response times of the p-MoTe₂/n-PdSe₂ vdWH FET devices are clear evidence of the excellent potential of optoelectronics and will likely be essential when developing highly efficient photodetectors in the broad VNIR region.

2. Results and Discussion

Initially, prior to the investigation of the p-MoTe₂/n-PdSe₂ vdWH FET, we undertook electrical measurements of a p-type MoTe₂ FET and an n-type PdSe₂ FET separately with various metal electrodes in the dark and under incident light. Figure 1a shows a schematic illustration of a p-MoTe₂ FET with metal electrodes (drain and source) under incident light. In order to investigate the roles of the metal electrodes on the device performance of the p-MoTe₂ FET, three different metals, here Pd, Ni, and Cr, having different work functions (φₖ) were deposited as electrodes with a thickness of 6 nm onto p-MoTe₂, followed by 60-nm-thick Au capping layers. The work functions of the Cr and Ni metals in this case are lower as compared to those of Pd (Φₚd (4.5 eV) < ΦₚNi (5.0 eV) < ΦₚPd (5.3 eV)). The inset of Figure 1b shows an
optical image of completed FETs with a p-MoTe₂ flake, with the thickness determined to be 13 nm (see Figure S1a, Supporting Information).

From MoTe₂ FETs, we first investigated the device performance in the dark (see Figures S1–S3, Supporting Information). Observed characteristics of the devices include the p-type nature of MoTe₂ with a low threshold voltage of \( \approx 8 \) V and a high ON/OFF current ratio \( (I_{on}/I_{off}) \) up to \( 10^4 \) (Figure S1b, Supporting Information), as well as high hole mobility \( (\mu_H) \) of \( 120 \) cm² V⁻¹ s⁻¹ with the Pd electrodes. Such high hole mobility and the low threshold voltage of the p-MoTe₂ FET with the Pd electrodes, as compared to those of other TMD-based FETs on SiO₂ substrates,\(^{[9a]} \) may be due to the high work function of the Pd metal, possibly resulting in a low Schottky barrier height \( (\varphi_B \approx 28 \text{ meV}) \)\(^{[29]} \) at the Pd-MoTe₂ junction (Figure S2, Supporting Information). Moreover, the linear behaviors of the current–voltage \( (I_{ds}–V_{ds}) \) curves of the p-MoTe₂ FET with Pd verify the Ohmic contact (Figure S3, Supporting Information). The observed key parameters of the p-MoTe₂ FETs are summarized in Table 1.

Subsequently, optoelectrical measurements of p-MoTe₂ FETs were taken under incident laser light with different wavelengths \( (\lambda_s) \) in the VNIR region for the three different metal electrodes (Pd, Ni, and Cr). Figure 1b shows the \( I_{ds}–V_{ds} \) curves of the MoTe₂ FET with Pd electrodes under incident laser light with different

Table 1. Comparison of the key parameters of p-MoTe₂ and n-PdSe₂ FETs and p-MoTe₂/n-PdSe₂ vdWH FETs.

| Structure          | Electrode | \( \mu_H \) [cm² V⁻¹ s⁻¹] | \( \varphi_B \) [meV] | \( \tau_r/\tau_d \) [ns] | \( R \) [A W⁻¹] | \( D^* \) [Jones] | EQE [%] |
|--------------------|-----------|---------------------------|------------------------|--------------------------|----------------|----------------|------|
| p-MoTe₂ FET        | Pd        | 120                       | 28                     | 0.5/0.9                  | 5.3 \times 10^4 | 4.2 \times 10^6 | 1.6 \times 10³ |
|                    | Ni        | 98                        | 45                     | 3/6                      | 2.9 \times 10^2 | 1.5 \times 10^5 | 9.0 \times 10^2 |
|                    | Cr        | 56                        | 90                     | 9/11                     | 1.5 \times 10^2 | 9.2 \times 10^4 | 4.6 \times 10²  |
| n-PdSe₂ FET        | Sc        | 162                       | 16                     | 0.09/0.07                | 1.2 \times 10³ | 2.1 \times 10^1 | 3.6 \times 10^3 |
|                    | Al        | 102                       | 38                     | 0.1/0.3                  | 3.7 \times 10² | 6.4 \times 10^1 | 1.1 \times 10^3 |
|                    | Ti        | 98                        | 75                     | 0.8/0.9                  | 2.4 \times 10² | 9.5 \times 10^4 | 7.5 \times 10²  |
| MoTe₂/PdSe₂ vdWH FET | Pd and Sc | –                         | –                      | 0.01/0.03                | 1.2 \times 10³ | 2.4 \times 10^4 | 3.5 \times 10^6 |
wavelengths in the VNIR region. The drain current $I_{ds}$ was increased from 10 µA in the dark to 31 µA at the bias voltage $V_{ds}$ of 2.0 V under the illumination of incident visible laser light ($\lambda = 532$ nm) with an input power of 20 nW. In contrast, under the illumination of incident NIR laser light ($\lambda = 1310$ nm), the increase in the $I_{ds}$ response is quite small due to the relatively large intrinsic bandgap ($\approx 1.1$ eV) of MoTe$_2$. From these $I_{ds}$ data, the photoreponsivity $R$, a pivotal parameter of photodetectors, was also estimated using the relationship $R = I_{ds\text{light}} / PA$[40] where $A$, $I_{ds\text{light}}$, and $P$ are the junction area, the photocurrent current ($I_{ds\text{light}} = I_{ds\text{light}} - I_{ds\text{dark}}$), and the input power of incident light, respectively. The estimated maximum value of $R$ for the MoTe$_2$ FETs with Pd is 532 A W$^{-1}$ in the visible region ($\lambda = 405$ nm) at zero back gate voltage ($V_{bg} = 0$ V) (Figure 1c). As expected, in the NIR region, however, the responses of the devices are very close to zero, as shown in the figure. Further detailed descriptions of the electrode effect on $R$ and other characteristics of the MoTe$_2$ FETs with Pd are described in Figure S4a–c, Supporting Information.

Next, we took electrical measurements of n-type PdSe$_2$ FETs with various metal electrodes in the dark and under incident light. Figure 1d shows a schematic illustration of an n-PdSe$_2$ FET with Pd described in Figure S4a–c, Supporting Information. The drain current $I_{ds}$ with Pd is 532 A W$^{-1}$ in the visible region ($\lambda = 532$ nm) with an input power of 20 nW. In contrast, under the illumination of incident NIR laser light ($\lambda = 1310$ nm), the drain current $I_{ds}$ was in-creased from 10 µA in the dark to 31 µA at the bias voltage $V_{ds}$ of 2.0 V. Given these observations, we also estimated the $R$ values of the PdSe$_2$ FETs as a function of the wavelength of incident light. The estimated maximum value of $R$ was found to be in the range of $1.2 \times 10^5$ A W$^{-1}$, as shown in Figure 1f. Such high $R$ values of PdSe$_2$ may be caused by the photo-doping and/or photo-gating effect.[16,32] To confirm this effect, the transfer curves of the FET with Sc were measured under incident light with different $\lambda$ and compared to those measured in the dark (Figure S8a, Supporting Information), exhibiting illumination-induced shifts towards positive gate voltages. Such shifts are a clear confirmation of the photo-gating effect via the increased number of trapping holes at the interface, resulting in additional electron flows in PdSe$_2$ under light illumination.[16,32] Detailed descriptions of the electrode effect on $R$ and other characteristics of the n-PdSe$_2$ FETs are described in Figure S8b–d, Supporting Information.

At this point, we focus our attention on the vdWHs of MoTe$_2$ and PdSe$_2$ (MoTe$_2$/PdSe$_2$). We fabricated an FET with MoTe$_2$ (10 nm)/PdSe$_2$ (13 nm) vdWHs and then measured its electrical characteristics in the dark and under incident laser light. Figure 2a shows a schematic illustration of the MoTe$_2$/PdSe$_2$ vdWH FET. Figure 2b shows an optical image of a completed MoTe$_2$/PdSe$_2$ vdWH FET with Pd and Sc electrodes for the optimization of the Ohmic contact for the vdWHs. In order to investigate the structural fingerprints of p-MoTe$_2$ and n-PdSe$_2$ in the fabricated vdWHs, the Raman spectra of each TMD material and the vertically stacked materials with the heterostructure were measured, as presented in Figure 2c. From the PdSe$_2$ flake of the FET, we observed the first three Raman peaks at low wavenumbers ($A_1^g \approx 144$ cm$^{-1}$, $A_2^g \approx 209$ cm$^{-1}$, and $B_2^g \approx 223$ cm$^{-1}$) and the fourth peak at a high wavenumber ($A_1^g \approx 256.5$ cm$^{-1}$) (upper curve). These peaks correspond to the vibrational modes of Se atoms and the relative vibration mode between the Pd atom and Se atoms.[33] For the p-MoTe$_2$ flake, three Raman peaks were observed at $A_{1g} \approx 173.6$ cm$^{-1}$, $E_{2g} \approx 235.1$ cm$^{-1}$, and $B_{1g} \approx 290.9$ cm$^{-1}$, which belong to the out-of-plane, in-plane, and bulk-inactive modes, respectively (lower curve). The main Raman peak $E_{2g}^1$ of MoTe$_2$ is related to the in-plane vibrations between the Mo and Te atoms.[80] Further, in the MoTe$_2$/PdSe$_2$ vdWH heterostructure, the observed Raman spectra clearly confirm that the basic structural characteristics of both TMD materials are clearly conserved, as shown in the figure (middle curve).

Subsequently, we investigated the device performance of the MoTe$_2$/PdSe$_2$ vdWH FET. In the dark, we found and verified excellent electric characteristics of the heterojunction device (see Figure S9, Supporting Information). Examples include an efficient gate-voltage-controlled effect of the rectification ratio $RR$ with a corresponding maximum value of $\approx 0.63 \times 10^6$ due to the considerable suppression of the reverse/leakage current (Figure S9a,b, Supporting Information) via the electric switching characteristic of the TMDs between the semiconducting and semi-insulating phases,[34] and an efficient gate-voltage-modulated ideality factor ($\eta$) with a lower value of 1.1 (Figure S9c,d, Supporting Information), among others. It should be noted that the maximum $RR$ value of the MoTe$_2$/PdSe$_2$ vdWH FET in this study is significantly high in comparison with those of the various TMD devices reported previously (Table S1, Supporting Information).[35] Thus, it is clear that effective electrostatic control of the charge transport in MoTe$_2$/PdSe$_2$ vdWHs can provide a way to design various TMD-based electronic devices. In addition, it should be noted that when the layer thickness of MoTe$_2$ or PdSe$_2$ decreased, the device performance began to deteriorate (Figure S10, Supporting Information).

Next, we explored the optoelectronic properties of the MoTe$_2$/PdSe$_2$ vdWH FETs. Figure 2d shows the $I_{ds} - I_{ds}$ characteristics as measured under the illumination of incident light ($\lambda = 532$ nm) with various input powers at $V_{bg} = 0$ V. From the $I_{ds} - I_{ds}$ curves, it was found that the forward and
reverse photocurrents $I_{ph}$ increase continuously as the input power of the incident laser light increases. Such increases in the reverse and forward photocurrents of the MoTe$_2$/PdSe$_2$ vdWH heterojunction device are mainly due to the efficient generation of photo-excited electron–hole (e–h) pairs and the effective separation of excess e–h pairs caused by the built-in electric field at the interface of the heterojunction. Figure 2e shows the photocurrent $I_{ph}$ as a function of the input power of incident light ($\lambda = 532$ nm). In this figure, the photocurrent shows sub-linear dependence on the input power of incident light, expressed as $I_{ph} = cP^{\theta}$, where $c$ is a proportional constant, $P$ is the input power of the incident light, and $\theta$ denotes the power-law index.[36] From the best linear fit to the experimental data, the obtained value of the power-law index $\theta$ is 0.94, which is very close to 1.0 for an ideal photodetector having a low trap state junction.[36] Thus, this result clearly indicates that the MoTe$_2$/PdSe$_2$ vdWHs have a small number of trap states at the sharp heterojunction interface, which is crucial when attempting to realize highly sensitive photodetectors.

In addition, we studied the dependence of the photoresponse of the MoTe$_2$/PdSe$_2$ vdWH FET on the wavelength $\lambda$ of incident light in the VNIR range. Figure 2f shows the $I_{ds} - V_{ds}$ curves of the MoTe$_2$/PdSe$_2$ vdWH FET for several different $\lambda$ values of incident light. Interestingly, as indicated in the figure, the current $I_{ds}$ of the heterojunction FET decreases slightly as $\lambda$ of the incident light increases. For example, the observed values of $I_{ds}$ were 6.45 and 2.85 $\mu$A for $\lambda = 405$ and 2000 nm, respectively. Note that in contrast to the significant decrement of $I_{ds}$ of the single TMD-based FETs with MoTe$_2$ or PdSe$_2$ in the NIR region (see Figures 1b and 1e), $I_{ds}$ of the MoTe$_2$/PdSe$_2$ vdWH FET decreased slightly as $\lambda$ increased. Such a small decrement of $I_{ds}$ of the vdWH FET in the NIR region cannot be explained solely in terms of the intrinsic optical absorption of each TMD material used (MoTe$_2$ and PdSe$_2$), as discussed below.

Next, we evaluated the temporal photoresponse characteristics of the MoTe$_2$/PdSe$_2$ vdWH FET while turning the incident light on and off. The temporal photoresponses of the device were measured as a function of time for various input powers ($P$s) of incident light ($\lambda = 1064$ nm) (Figure 3a) and for various $\lambda$s of incident light in the VNIR range at a fixed $P$ of 100 nW (Figure 3b). From these responses, we estimated the response times, that is, the rise ($\tau_r$) and decay ($\tau_d$) times, using the simple relationship of $I_{tot} = I_{dark} + c \exp \left[ -t/\tau_{rd} \right]$, where $I_{tot}$ is the temporal photocurrent, $I_{dark}$ is the dark current, $c$ is a proportional constant, and $t$ is the time. By fitting the data, we attained remarkably fast rise and decay times of the vdWH FET, that is, $\tau_r = 16.1$ $\mu$s and $\tau_d = 31.1$ $\mu$s for NIR incident light with a wavelength $\lambda$ of 2000 nm. The obtained response times of the MoTe$_2$/PdSe$_2$ vdWH FET for several $\lambda$s are presented in Figure 3c. In comparison, the temporal photoresponses of single TMD FETs with MoTe$_2$ or PdSe$_2$ were also measured for several different electrodes (Figure S11, Supporting Information). The
Figure 3. Temporal photocurrents of the MoTe$_2$/PdSe$_2$ vdWH FET as a function of time under incident light a) with several different input power levels at a fixed $\lambda$ (1064 nm) and b) with several different $\lambda$s in the VNIR region ($\lambda$ = 405–2000 nm) at a fixed $P$ (100 nW) while turning the incident light on and off. c) Rise ($\tau_r$) and decay ($\tau_d$) times of the MoTe$_2$/PdSe$_2$ vdWH FET at several $\lambda$s of incident light with $V_{bg}$ equal to $-5$ V and $V_{ds}$ set to $-20$ V. d) Energy band diagram of the MoTe$_2$/PdSe$_2$ vdWH FET under incident light illumination. e) Energy band diagrams of the MoTe$_2$/PdSe$_2$ FET under negative ($V_{bg} < 0$ V, left panel) and positive ($V_{bg} > 0$ V, right panel) back gate voltages. $E_C$: conduction band, $E_V$: valence band, and $E_F$: Fermi level.

estimated $\tau_r/\tau_d$ values of the MoTe$_2$ FETs were 0.5/0.9, 3.0/6.0, and 9.0/11.0 ms for the Pd, Ni, and Cr electrodes, respectively, while the estimated $\tau_r/\tau_d$ values of the PdSe$_2$ FETs were 0.1/0.3, and 0.7/0.9 ms for the Sc, Al, and Ti electrodes, respectively. Thus, both the MoTe$_2$ and PdSe$_2$ FETs exhibited relatively slow rise and decay times, with their operating wavelength ranges restricted to the visible wavelength region due to their limited optical absorption ranges ($\lambda < 980$ nm) via the electronic transition from the valence band to the conduction band, that is, intra-TMD band-to-band transitions of each individual TMD material. Therefore, $\Delta E$ (in the case of MoTe$_2$ and PdSe$_2$) is small enough ($\Delta E < 0.6$ eV) to be feasible for use in the NIR region ($\lambda > 980$ nm) up to $\lambda = 2000$ nm.

Moreover, it is noteworthy that such an inter-TMD CT transition in the MoTe$_2$/PdSe$_2$ heterostructure device results in significantly fast photoresponses, especially for the NIR region, as shown in Figure 3c. The remarkably short response times related to the inter-TMD CT transition in the long-wavelength region ($\lambda > 980$ nm) can be ascribed to the quick separation into photo-carriers and the rapid charge carrier transfer at the interface of the MoTe$_2$/PdSe$_2$ vdWHs. It should be noted that the observed rise and decay times of the MoTe$_2$/PdSe$_2$ vdWH FET are one order of magnitude faster than those of other TMD-based heterostructure devices reported previously.[11,23a,37]

Interestingly, it was also found that the response times of the MoTe$_2$/PdSe$_2$ vdWH FET studied here are strongly affected by the back gate voltage $V_{bg}$. In order to explain this behavior, energy-level band diagrams of the MoTe$_2$/PdSe$_2$ FET under negative and positive $V_{bg}$ levels are presented in Figure 3e. Under negative back gate voltage ($V_{bg} < 0$ V), the energy level of the bottom MoTe$_2$ TMD shifted up, which enhanced the strength of the electric field ($E$) at the interface between the MoTe$_2$ and PdSe$_2$ layers in the vdWHs, subsequently facilitating the rapid separation and collection of numerous photo-carriers via inter-TMD CT transitions. Thus, the photosresponse times, $\tau_r$ and $\tau_d$, decreased significantly to 16.1 and 31.1 $\mu$s (at $V_{bg} = -20$ V and $V_{bias} = -5$ V), respectively, even for incident light in the NIR region ($\lambda = 2000$ nm) (Figures 3c and 3d). In contrast, under positive back gate voltage ($V_{bg} > 0$ V), the energy level of MoTe$_2$ moved downward and thus the strength of $E$ and the CT transition decreased, resulting in reductions of the separation and collection of photo-carriers with slow response times ($\tau_r/\tau_d$ $\approx$ 65.6/92.8 $\mu$s).

Next, we analyzed the characteristic parameters, in this case the photo-switching ratio ($I_{ph}/I_{dark}$) and the responsivity $R$ of the
MoTe$_2$/PdSe$_2$ vdWH FET. From the data shown in Figure 3a,b, we estimated the photo-switching ratio $I_{ph}/I_{dark}$ of the vdWH FET and found that the $I_{ph}/I_{dark}$ ratio is extremely high, reaching nearly $\approx 10^4$ mainly due to the enhanced photocurrent $I_{ph}$ as well as the suppressed dark current $I_{dark}$ in the vdWH FET (Figure S12, Supporting Information). The responsivity $R$ values of the vdWH FET were then extracted as a function of the input power of incident light with several different wavelengths (Figure 4a). As shown in the figure, high $R$ values were obtained due to the small bandgap with low Schottky contact barriers in the VNIR region. The responsivity $R$ is significantly higher over the broad spectral range due to the intensification and separation of photo-carriers in the vdWH FET device via the intra-TMD band-to-band transitions together with the inter-TMD CT transitions under the internal electric field at the interface between the PdSe$_2$ and MoTe$_2$, as mentioned above. Thus, a high value of $R$ can be achieved in the MoTe$_2$/PdSe$_2$ vdWH FET over a broad VNIR spectral range.

The responsivity $R$ values of the MoTe$_2$/PdSe$_2$ vdWH FET were also observed as a function of $V_{bg}$ for several wavelengths of incident light in the VNIR range (Figure 4b). For incident light at a short wavelength $\lambda$ (visible range), as the gate voltages decrease ($V_{bg} < 0$), the $R$ value also decreases continuously. This is caused by the reduced collection of photo-carriers generated via the intra-TMD band-to-band transitions at a negative $V_{bg}$ owing to the increased effective $\phi_F$ at the interface between the MoTe$_2$, TMD and Pd electrode. In contrast, for incident light with a long wavelength $\lambda$ (NIR range), the value of $R$ increases with a decrease in $V_{bg}$. In this NIR range, a large amount of photocurrent can be generated due to the inter-TMD CT transition between MoTe$_2$ and PdSe$_2$ at $V_{bg} < 0$. It is therefore clear that not only the intra-TMD band-to-band transition but also the inter-TMD CT transition for the MoTe$_2$/PdSe$_2$ vdWH FET can be efficiently controlled by the back gate voltage, leading to a high value of $R$ ($\approx 10^6$) in the broad VNIR region. We also investigated the effects of the metal contacts on the $R$ values. As shown in Figure S13a, Supporting Information, the Ohmic contact with Pd and Sc electrodes for the MoTe$_2$/PdSe$_2$ vdWHs (Pd/MoTe$_2$/PdSe$_2$/Sc) show the best results among the metal contacts studied here, mainly due to the small values of $\phi_F$ for the Pd and Sc metal contacts. The maximum value of $R$ for the contact-optimized MoTe$_2$/PdSe$_2$ vdWH FET reached $1.2 \times 10^5$ AW$^{-1}$, which is significantly higher than previously reported values (Figure S13b, Supporting Information).

Furthermore, the specific detectivity ($D^*$) of MoTe$_2$/PdSe$_2$ vdWH FETs with Pd and Sc contact electrodes were also estimated using the relationship $D^* = \sqrt{\Delta f / \langle i_n^2 \rangle} R$, where $\Delta f$ and $i_n$ represent the bandwidth and noise current, respectively.
respectively. The mean square noise current ($i_n^2$) is calculated according to ($i_n^2 = 4k_B T A f / R_n$), where $A_f$ and $R_n$ correspondingly denote the bandwidth and device resistance. The estimated mean square noise current is $1.8 \times 10^{-25}$ A$^2$ Hz$^{-1}$. A remarkably high value of $D^*$ of $2.4 \times 10^{14}$ Jones is achieved, which is nearly two orders of magnitude higher than those previously reported,[12a,19a,40,41] as shown in Figure 4c. Even at room temperature, the MoTe$_2$/PdSe$_2$ vdWH FET exhibits a significantly wide spectral response from 400 to 2000 nm, with $D^*$ greater than $10^{14}$–$10^{12}$ Jones at wavelengths from 400 to 1064 nm, greater than $10^{11}$ Jones from 1064 to 1550 nm, and greater than $10^9$ Jones from 1550 to 2000 nm. Such high detectivity with a broad response of the MoTe$_2$/PdSe$_2$ vdWH FET is comparable to or even better than those from conventional Si and GaAs photodetectors.[41b,e,f] Moreover, the $EQE$s of the MoTe$_2$/PdSe$_2$ vdWH FET were also extracted using the relationship $EQE = R_{\text{DS}} \times \omega_k$ (Figure S13c, Supporting Information). As shown in the figure, the obtained $EQE$ values were considerably high, reaching $EQE \approx 3.5 \times 10^6$ at $\lambda = 405$ nm and $EQE \approx 1.3 \times 10^5$ at $\lambda = 2000$ nm, much greater as compared to previous values, as shown in the figure, especially for the NIR region.[13b,19a,c,d] In addition, the long-term storage stability of the MoTe$_2$/PdSe$_2$ vdWH FET was also studied, as presented in Figure S14, Supporting Information.

Finally, in Figure 4d, we show the responsivity outcomes and response times of several important photodetector technologies, including the MoTe$_2$/PdSe$_2$ vdWH FET studied here. It is clear from the figure that the response time of the MoTe$_2$/PdSe$_2$ vdWH FET is considerably faster as compared to previous values, especially for the VNIR region, without sacrificing other key figures of merit such as the responsivity and $EQE$. Thus, the MoTe$_2$/PdSe$_2$ vdWHs studied here hold great promise for fast and high photo-detectivity over a wide spectral range in the VNIR region. Such significantly improved values of $R$, $D^*$, and $EQE$ in the broad VNIR region with fast response times mainly stem from the excellent Ohmic contact with low Schottky barriers, the sharp interface, and the inter-TMD CT transition within the MoTe$_2$/PdSe$_2$ vdWHs. Hence, the MoTe$_2$/PdSe$_2$ vdWHs studied here can be a useful for improving the photocurrent and photo-gating characteristics, with possible applications in various optoelectronics in the VNIR region.

Overall, the findings above show that the photoresponse characteristics of FETs with the TMD heterostructure, such as MoTe$_2$/PdSe$_2$ vdWHs, are mainly influenced by the metal-TMD contacts and the CT transition between MoTe$_2$/PdSe$_2$ vdWHs, which will provide a key platform for further improvements in the performance of VNIR photodetectors coupled with TMD materials.

3. Conclusion

In summary, we reported a novel TMD heterostructure assembly consisting of MoTe$_2$ and PdSe$_2$. Effective tuning of the rectification ratio $RR$ of $6.3 \times 10^3$ at $V_{BG} = -40$ V with a low $\phi_S$ value and sharp interface is realized for the MoTe$_2$/PdSe$_2$ vdWH FET with Pd and Sc electrodes. This $RR$ value is much higher than those of other TMD vdWHs reported previously. Excellent phototresponse performance capabilities of the MoTe$_2$/PdSe$_2$ vdWH FET were noted in the VNIR region ($\lambda = 405 - 2000$ nm). Rapid rise (16.1 $\mu$s) and decay (31.1 $\mu$s) times were also obtained with NIR light ($\lambda = 2000$ nm). Moreover, extraordinary $R = 1.2 \times 10^5$ A $W^{-1}$, $D^* = (2.4 \times 10^{14})$ Jones, and $EQE = (3.5 \times 10^6)$ outcomes were obtained in the MoTe$_2$/PdSe$_2$ vdWH FET. These significantly improved values of $R$, $D^*$, and $EQE$ mainly stem from the excellent Ohmic contact with low Schottky barriers, the sharp interface, and the inter-TMD charge-transfer transition together with intra-TMD band-to-band transitions in the MoTe$_2$/PdSe$_2$ vdWHs. Therefore, the TMD-based vdWHs of MoTe$_2$/PdSe$_2$ studied here represent a new opportunity to develop state-of-the-art photonic devices in optoelectronics, such as effective VNIR optical sensors, waveguide-integrated photodetectors, and/or nanophotodetectors.

4. Experimental Section

A multilayer MoTe$_2$ or PdSe$_2$ nanoflake was exfoliated and transferred onto a p-type Si/SiO$_2$ (300 nm) substrate, acting as a back gate, with the help of the standard mechanical exfoliation method using Scotch tape.[17a] The thickness of the transferred flake (MoTe$_2$ or PdSe$_2$) on the substrates was identified by the interference effect.[14c] Further, Raman spectroscopy and atomic force microscopy were utilized to confirm the thicknesses of the flakes. Only those flakes that had a uniform thickness and clean surfaces were used for the electric and optoelectric measurements.

In order to fabricate the MoTe$_2$/PdSe$_2$ vdWHs, a multilayer PdSe$_2$ nanoflake but also exfoliated and transferred onto the top of a polydimethylsiloxane (PDMS) stamp by the mechanical exfoliation method. Next, with a micro-aligner stage, the nanoflake with multilayer PdSe$_2$ on the PDMS stamp was transferred onto a multilayer MoTe$_2$ flake prepared on a Si/SiO$_2$ substrate. Subsequently, the MoTe$_2$/PdSe$_2$ vdWHs were placed in a furnace for thermal annealing at 200 °C under an Ar/H$_2$ (97.5%/2.5%) gas flow. After annealing, metal electrodes on the flakes were formed and patterned by electron-beam deposition and lithography with poly(methyl methacrylate) (PMMA). For the metal electrodes, thin layers ($\approx 6$ nm) of different metals having high work functions (Pd, Ni, and Cr for MoTe$_2$) or low work functions (Sc, Al, and Ti for PdSe$_2$) were deposited. This step was followed by the subsequent deposition of 60-nm Au capping layers by electron-beam evaporation. After the deposition of the metal electrodes, the samples were placed in acetone for the lift-off process. The final sample FET devices were then placed into a vacuum box to measure their electrical performance capabilities.

The fabricated TMD FETs were analyzed according to their transfer curves ($I_{DS}-V_{BG}$) at a constant $V_{DS} = 1.0$ V using a Keithley source voltmeter (K-2400) and a picometer (K-6485). The back-gate voltage ($V_{BG}$) was swept between $V_{BG} = \pm 40$ V and the drain current ($I_{DS}$) was determined using the picometer. For optical characterization, the sample FET and heterostructure devices were illuminated under incident laser light with different input powers and wavelengths.

Supporting Information

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

Acknowledgements

This research was supported by the Basic Science Research Program through the National Foundation of Korea (NRF), funded by the Korea Government (MEST) (2020R1A2B5B03097060) and Kwangwoon University (2021).

Conflict of Interest

The authors declare no conflict of interest.
Data Availability Statement

Research data are not shared.

Keywords

charge-transfer transition, field-effect transistors, photoresponsivity, specific detectivity, transition-metal dichalcogenides

Received: October 1, 2020
Revised: February 10, 2021
Published online: March 16, 2021
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