NIR self-powered photodetection and gate tunable rectification behavior in 2D GeSe/MoSe₂ heterojunction diode

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Two-dimensional (2D) heterostructure with atomically sharp interface holds promise for future electronics and optoelectronics because of their multi-functionalities. Here we demonstrate gate-tunable rectifying behavior and self-powered photovoltaic characteristics of novel p-GeSe/n-MoSe₂ van der waal heterojunction (vdW HJ). A substantial increase in rectification behavior was observed when the devices were subjected to gate bias. The highest rectification of ~1 × 10⁴ was obtained at V₉ = −40 V. Remarkable rectification behavior of the p-n diode is solely attributed to the sharp interface between metal and GeSe/MoSe₂. The device exhibits a high photoresponsivity of 465 mAW⁻¹, an excellent EQE of 670%, a fast rise time of 180 ms, and a decay time of 360 ms were obtained. Furthermore, the diode exhibits detectivity (D) of 7.3 × 10⁹ Jones, the normalized photocurrent to the dark current ratio (NPDR) of 1.9 × 10¹⁰ W⁻¹, and the noise equivalent power (NEP) of 1.22 × 10⁻¹³ WHz⁻¹/². The strong light-matter interaction stipulates that the GeSe/MoSe₂ diode may open new realms in multi-functional electronics and optoelectronics applications.

Beyond the great success of graphene and their derivatives, the analogs of the 2D materials, such as the transition metal dichalcogenide (TMD) and the transition metal carbide (TMC) nanostructures have strikingly increased interest in science. Compared to graphene, which has high carrier mobility but zero bandgap limited its device application, and the transition-metal dichalcogenides (TMDs) that have the formula MX₂ (M = Mo, W; Ge; X = S, Se, or Te), a class of 2D semiconductors have recently attracted remarkable scientific and technological interest for innovative devices. They exhibit a wide range of material properties, such as high carrier mobility for both electrons and holes, have a relatively large bandgap of 1.5–2.5 eV, have a tunable direct bandgap that ranges from 0.3 to 1.5 eV, have an ideal sub-threshold swing of ~60 mV/dec, an I₉/I₉ ratio of 10⁻³⁻¹⁰⁻⁴, and high carrier mobility (200 cm² V⁻¹ s⁻¹@ room temperature mobility for a single-layer MoS₂ transistor with high-K dielectric and 1000 cm² V⁻¹ s⁻¹@ 3 K temperature)¹⁻³. Due to the weak van der Waals interface forces in graphene and TMDs materials, the p–n diode or the Schottky barriers (SBs) at the metal/TMDs interfaces have played an important role in electronic devices⁴. Besides, the electrical properties are hindered by the contact resistances rather than the intrinsic TMDs properties. The development of the TMDs with different bandgaps and work functions allow for the bandgap engineering of heterostructures that may possess new physical and electrical properties. Among TMDs materials, which include MoSe₂, GeS, and GeSe, have been proposed as alternative 2D systems, and they have exhibited better performance in photodetectors. GeSe belongs to the layered IV–VI nanostructures since the p-type semiconductors with narrow bandgap (1.1–1.2 eV) are potential alternatives to the lead chalcogenides and open an avenue to fabricate highly efficient electronic and optoelectronic devices⁵⁻⁶. The MoSe₂ from indirect (bulk crystal) to direct (monolayer) results in the bandgap increasing from 1.1 to 1.5 eV, which become direct in single atomic layers, and it makes them promising candidates in field-effect transistors (FETs), photovoltaic cells, light-emitting diodes (LEDs), and photodetectors. With the lower light response in photodetection/sensing, the bandgap tunability value for electrons or hole transfers between bulk TMDs materials down to the monolayer and slow electron transfer pathways is still greatly impeded. Consequently,

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The optical image of the p-GeSe/n-MoSe₂ heterostructure is shown in Fig. 1b. The atomically thin flakes of the materials using a scotch tape mechanical exfoliation technique, which is similar to the technique that is employed for work function of the Cr depicted in Fig. 2d. The field-effect carrier mobility (\( \mu_{FE} \)) of the p-GeSe and the n-MoSe₂ heterojunction region was formed. To clean the surface and optimize the charge carrier, the flakes of the p-GeSe and the n-MoSe₂ were identified using an optical microscope, and the multilayer n-MoSe₂ was directly stacked on the top of the p-GeSe flake. Raman spectroscopy and atomic force microscopy (AFM) were also conducted. Electron beam lithography was used for the metal deposition of palladium/gold (Pd/Au:10/20 nm) and Cr/Au:10/20 nm onto the p-GeSe and the n-MoSe₂, respectively. The lift-off processes were conducted to form electrodes on the multilayer p-GeSe and n-MoSe₂ flakes. The electrical characterization at room temperature were exhibited using a Keithly 4200A-SCS parameter analyzer. The photovoltaic characteristics of the p-GeSe/n-MoSe₂ heterojunction photodetector was performed using a continuous wave laser beam from a diode NIR laser (850 nm) that was directly illuminated onto the device.

Experimentation

We prepared all the p-GeSe and the n-MoSe₂ atomically thin flakes by peeling them from their parent bulk crystals using a scotch tape mechanical exfoliation technique, which is similar to the technique that is employed for the exfoliation of graphene10,11 and we transferred it onto a Si/SiO₂ (300 nm) substrate using a transparent poly(dimethylsiloxane) (PDMS) stamp using an aligned dry transfer12,13. The multilayer p-GeSe and the n-MoSe₂ flakes were identified using an optical microscope, and the multilayer n-MoSe₂ was directly stacked on the top of the p-GeSe flake. Raman spectroscopy and atomic force microscopy (AFM) were also conducted. Electron beam lithography was used for the metal deposition of palladium/gold (Pd/Au:10/20 nm) and Cr/Au:10/20 nm onto the p-GeSe and the n-MoSe₂, respectively. The lift-off processes were conducted to form electrodes on the multilayer p-GeSe and n-MoSe₂ flakes. The electrical characterization at room temperature were exhibited using a Keithly 4200A-SCS parameter analyzer. The photovoltaic characteristics of the p-GeSe/n-MoSe₂ heterojunction photodetector was performed using a continuous wave laser beam from a diode NIR laser (850 nm) that was directly illuminated onto the device.

Result and discussions

A schematic illustration of the demonstrated p-GeSe/n-MoSe₂ heterostructure device is depicted in Fig. 1a. The optical image of the p-GeSe/n-MoSe₂ heterostructure is shown in Fig. 1b. The atomically thin flakes of the p-GeSe and the n-MoSe₂ were peeled from their parent bulk crystals using a scotch tape mechanical exfoliation technique, which was transferred on a 300 nm SiO₂/Si substrate27. A few-layers of the n-MoSe₂ were directly illuminated onto the device.  

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\[
\mu_{FE} = \frac{L}{W} \left( \frac{dV_{ds}}{dV_{bg}} \right) \frac{1}{C_{bg} V_{ds}}
\]

where W is the channel width, L is the channel length, \( C_{bg} \) is the gate capacitance (~ 115 aF/µm²) for the SiO₂ substrate, and \( \left( \frac{dV_{ds}}{dV_{bg}} \right) \) is the slope of the transfer curve. The mobilities of the p-GeSe and the n-MoSe₂ were estimated to be 110 cm²V⁻¹s⁻¹ and 85 cm²V⁻¹s⁻¹, respectively.

The gate-tunable electrical characteristics were also investigated. Figure 3a exhibits the gate dependent output characteristics of the p-GeSe/n-MoSe₂ heterostructure diode, and Fig. 3b shows the same output curves in a corresponding logarithmic plot. It revealed that the rectifying behavior of the device is tuned by the electrostatic consideration will still be given to matching the band alignment for electron or hole transfers between bulk materials for the development of photodetector creation.
gate-voltage. The forward bias rectifying current increases as the gate voltages ($V_g$) increased from $-V_g$ to $+V_g$, the electrons are attracted to the interface between GeSe and SiO$_2$ to form accumulation layer results the Fermi level of GeSe moves towards the conduction band and lowering potential barrier height results in decreasing rectification current attributed to electrostatic doping of electrons. Moreover, we investigated the rectification ratio, which is defined as the ratio of the forward current over the reverse current, $I_f/I_r$, up to $1.4 \times 10^4$ at $V_g = -40$ V.

We found that at a positive gate voltage of $V_g = +40$ V, both the reverse and forward currents increase concurrently, which suppress the rectification as a result. In anticipation of the negative gate voltage, the reverse current is constrained to increase the rectification in the p-GeSe/n-MoSe$_2$ heterostructure diode, which is depicted in Fig. 3c. Additionally, we estimated the ideality factor to confirm the performance of the rectifying behavior of the p-GeSe/n-MoSe$_2$ heterojunction diode using the thermionic emission theory.$^{4,35}$

$$I_D = I_S \left[ \exp \left( \frac{qV}{nK_B T} \right) - 1 \right]$$

(2)

where $I_S$ is the reverse bias saturation current, $n$ is the ideality factor, $q$ is the elementary charge, $T$ is the temperature, and $K_B$ is Boltzmann’s constant. After the interpretation above, the equation becomes
The ideality factor (n) can be obtained via the following equation.

\[
\ln(I_D) = \ln(I_S) + \left(\frac{q}{n k_B T}\right) V
\]  

(3)

The ideality factor (n) can be obtained via the following equation.

\[
n = \left(\frac{q}{k_B T}\right) \left|\frac{dV}{d\ln I_D}\right|
\]  

(4)

Figure 3d illustrates the ideality factor function of the gate voltage, the ideality factor of 1.1 is obtained at \( V_g = -40 \) V, which is close to the ideal diode value (\( n = 1 \)). The relative degrading of the gate tunable ideality factor is attributed to the surface carrier recombination at the interface of the p-GeSe/n-MoSe2 diode, which results in a decrease in the electric field. The variation in the electron affinity and the bandgap between the monolayers generates an atomically sharp hetero-interface, and the interface band alignment of the p-GeSe/n-MoSe2 heterostructure is predicted to be a type II band alignment, which is shown in Fig. 4.

Furthermore, we investigated the self-powered photovoltaic characteristics of a p-GeSe/n-MoSe2 heterostructure device. The self-powered photodetectors are devices that can separate photoexcited carriers by the built-in electrical field at the junctions without any external power source. On this principle, the p-n junctions can be established for the photovoltaics. We used an NIR (850 nm) laser with various illumination power intensities (53.3, 98.5, 123, and 139 mW/cm²) to measure the photocurrent generated from the photodiode that was based on the p-GeSe/n-MoSe2 heterojunction. A strong photoresponse was observed in the p-GeSe/n-MoSe2 junction region, which showed that a continuous charge separation occurred at the junction. Figure 5a presents the \( I_{ds}-V_{ds} \) curves of the p-GeSe/n-MoSe2 heterojunction in dark and under photon irradiation with wavelength of 850 nm at zero bias with a constant gate voltage (\( V_g = 0 \)). The \( I_{ds}-V_{ds} \) curves are shifted down under the irradiation of light, which revealed that the device can be developed for self-powered photovoltaic energy conversion under the action of open-circuit voltage (\( V_{oc} \)). We investigated an open-circuit voltage (\( V_{oc} \)) of 0.349 V and a short-circuit current (\( I_{sc} \)) of 14.5 nA for the 139 mW cm⁻² light intensity. The external quantum efficiency (EQE) was investigated by using the following formula.

\[
EQE = \frac{hc}{\epsilon \lambda R}
\]

(5)

where \( \lambda \) is the incident light wavelength, \( h \) is the plank’s constant, and \( c \) is the velocity of light. We obtained a value for EQE of 670% in the p-GeSe/n-MoSe2 diode. The power intensity-dependent EQE is depicted in Fig. 5b.
Additionally, we also characterized the transient photoresponse of the device. The dynamic photoresponse rise and fall time of the p-GeSe/n-MoSe₂ diode was observed under an NIR laser light irradiation with a wavelength (λ) of 850 nm at various power intensities, which is shown in Fig. 5c. The rise time is the τ_r, the time it takes by the device to reach 90% from 10% and the fall time is τ_f, the time it takes by the device to decay from 90 to 10%³⁶,³⁸,³⁹. We found a rise time of 180 ms and a fall time of 360 ms, which are shown in Fig. 5d. The response time of the device is not as fast as we expected, which may be due to the charge carrier trapping and the longer charge dissociation time⁴⁰–⁴².

Moreover, in order to evaluate the performance of the device, several important figures of merits were calculated. For example, responsivity (R), detectivity (D), the normalized photocurrent to a dark current ratio (NPDR), and the noise equivalent power (NEP) with variation of incident light power intensities were calculated. The
responsivity ($R = J_p/P_{in}$), where $J_p$ is the photocurrent density and $P_{in}$ is input power per area, and the detectivity ($D = R/\sqrt{2qJ_d}$), where $q$ is the elementary charge and $J_d$ is the dark current density, are significant facets of the photodetector36,38,39, which is shown in Fig. 6a. The greater value of responsivity is attributed to the higher photocurrent43. Similarly, the device that has a lower dark current provides a higher detectivity. Thus, the greater values of both $R$ and $D$ are important aspects of an efficient photodetector37,43,44. We obtained a high responsivity of $R = 465$ mAW$^{-1}$ and detectivity of $D = 7.3 \times 10^9$ Jones.

Figure 5. (a) The $I–V$ characteristics of p-GeSe/n-MoSe$_2$ heterojunction diode under dark and variable intensities. (b) The external quantum efficiency EQE function of incident power. (c) The time-dependent photoresponse of p-GeSe/n-MoSe$_2$, heterojunction diode under illuminations with different laser light (@850 nm) intensity at $V_{ds} = 0$ V. (d) The rise time and decay time.

Figure 6. (a) The responsivity, $R$ (mA W$^{-1}$), and the detectivity, $D$ (Jones) function of power intensities (b) the normalized photocurrent to dark current ratio NPDR (W$^{-1}$) and the noise equivalent power NEP (W Hz$^{-1/2}$) function of power intensities.
In summary, we demonstrate a p-GeSe/MoSe2 based multifunctional HJ p–n diode. The diode explicitly exhibits gate tunable high rectification of $\sim 1 \times 10^4$ at negative gate bias ($V_g = -40$ V). The introduction of the ohmic contacts reveals that the rectification behavior of a p–n diode is solely attributed to the sharp interface between metal contacts and GeSe/MoSe2. Our device shows high photoresponse at an NIR (850 nm). The high responsivity of 465 mAW$^{-1}$, the excellent EQE (670%), the fast rise time of 180 ms, and the decay time of 360 ms were obtained. The device also shows detectivity D of 7.3 x 10$^{13}$ Jones, a normalized photocurrent to dark current ratio NPDR of 1.9 x 10$^{10}$ W$^{-1}$, and a noise equivalent power NEP of 1.22 x 10$^{-13}$ WHz$^{-1/2}$. The NEP revealed that the photodetector, which is based on the p-GeSe/n-MoSe2 heterojunction, has the capability of detecting power as low as 10$^{-13}$. Additionally, we characterized the spectral responsivity of the p-GeSe/n-MoSe2 heterojunction. The device was subjected to constant illuminating power of 53 mW cm$^{-2}$ with wavelength ranging from 220 to 850 nm. Figure S1c shows a sharp increase of the spectral response on the short wavelength side is reasonably due to more photon energy absorbed by the device, attributed to more electrons and holes generation under larger photons energy. Table S1 in supplementary information shows the comparative investigated photoresponse and sensitivity of our device based on p-GeSe/n-MoSe2 heterojunction, which is much higher than the previously reported values. The strong light-matter interaction in the device explicitly suggests that the p-GeSe/n-MoSe2 based heterojunction p–n diode is a promising candidate for optoelectronics technologies.

Conclusions

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Author contributions
M.H., S.H.A.J., & A.A.: Design, carried out experiments analyzed data and co-wrote paper. D.N.C., J.J., & Y.S.: Performed AFM experiments and analyzed data. M.R., & S.Ab.: Co-wrote paper.

Competing interests
The authors declare no competing interests.

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