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Visible to near-infrared photodetectors based on MoS$_2$ vertical Schottky junctions

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Abstract
Over the past few years, two-dimensional (2D) nanomaterials, such as MoS$_2$, have been widely considered as the promising channel materials for next-generation high-performance phototransistors. However, their device performances still mostly suffer from slow photoresponse (e.g. with the time constant in the order of milliseconds) due to the relatively long channel length and the substantial surface defect induced carrier trapping, as well as the insufficient detectivity owing to the relatively large dark current. In this work, a simple multilayer MoS$_2$ based photodetector employing vertical Schottky junctions of Au-MoS$_2$-ITO is demonstrated. This unique device structure can significantly suppress the dark current down to $10^{-12}$ A and enable the fast photoresponse of 64 $\mu$s, together with the stable responsivity of $\sim$1 A W$^{-1}$ and the high photocurrent to dark current ratio of $\sim$10$^6$ at room temperature. This vertical-Schottky photodetector can also exhibit a wide detection range from visible to 1000 nm. All these results demonstrate clearly that the vertical Schottky structure is an effective configuration for achieving high-performance optoelectronic devices based on 2D materials.

Supplementary material for this article is available online

Keywords: MoS$_2$, vertical Schottky junction, dark current, photodetectors

(Some figures may appear in colour only in the online journal)
much larger bandgap of ∼1.8 eV. Generally, when MoS2 is thinned down to the 2D monolayer, the broken inversion symmetry and indirect-to-direct bandgap transition are correspondingly observed as a result of quantum confinement [2–4]. Also, the mechanical strength of MoS2 membrane is 30 times higher than that of steel while the stability can be maintained up to 1100 °C in an inert environment [5]. When fabricated into field-effect transistors, these monolayer or few-layer based devices exhibit a high ON/OFF current ratio of 107, an impressive drain current density of 240 μA μm−1 and a peak field-effect mobility of 184 cm2 V−1 s−1 [6]. In this regard, MoS2 has become an ideal candidate of channel materials among many 2D materials for various applications.

In specific, Yin et al reported monolayer MoS2 based photodetectors, demonstrating a photoresponsivity of 7.5 mA W−1 when the illumination power, bias and gate voltages are 80 μW, 1 V and 50 V, respectively [7]. Although these photodetectors can outperform the single-layer graphene based photodetectors in terms of the responsivity by almost ten times [1, 8], the response time (i.e. 50 ms) of MoS2 based detectors is not quite as fast as the one of graphene counterparts [7]. Recently, high-performance MoS2 photodetectors have been achieved utilizing unique device structures and/or special designs. The highest responsivity of MoS2 photodetectors can reach 880 A W−1 under an ultralow illumination power of 150 pW (i.e. 24 W cm−2) when a substantially high back-gate voltage of 70 V and a large drain bias of 8 V is applied [9]. Despite the high responsivity, this type of photodetectors still display an insufficient response with the long rise time (trise) and inefficient decay time (tdecay) of 4 s and 9 s, accordingly. This relatively slow response is attributed to the trap states existed on the MoS2 device channel surface and/or located at the interface between MoS2 channel and underlying SiO2 dielectric layer, which capture photo-generated electrons and then result in the long lifetimes of photo-generated holes. Hence, these devices sacrifice their photoresponse speed in order to obtain the high photoresponse gain [10]. In addition, the responsivity of monolayer MoS2 based photodetectors can be further increased by decreasing the illumination laser power, in which this enhancement can be partly owing to the absorption saturation in MoS2 and partly in consequence of the screening of a built-in electric field caused by the photoexcited electrons in the conduction band of MoS2 [11]. At the same time, the MoS2 device can be integrated with a ferroelectric polymer film to exhibit a maximum responsivity of 2570 A W−1 under a low illumination power of 1 nW at a zero gate voltage and a Vds of 5 V [12]. It is noted that this ferroelectric polymer film plays a crucial role to strongly suppress the dark current and help to realize a highly sensitive photodetector. More importantly, the trise and tdecay of such device are observed to be 1.8 ms and 2.0 ms, respectively, which are much faster than those of state-of-the-art MoS2 based photoconductive devices. In any case, to the best of our knowledge, the photoresponse time of most MoS2 devices is yet limited in the order of milliseconds.

With the purpose to further optimize these figures of merit, in this work, we design and fabricate multilayer MoS2 based photodetectors employing simple vertical Schottky junctions of gold-MoS2-indium tin oxide (Au-MoS2-ITO). Impressively, the obtained device exhibits the excellent detector performance with a fast response time of 64 μs, a stable responsivity of ∼1 A W−1, a high photocurrent to dark current ratio of 106 and an ultralow dark current of 10−12 A at a zero bias in ambient environment. This superior photoresponse is ascribed to the extremely short channel length configured in the vertical manner for the fast transit time as well as the built-in electric field associated to accelerate the separation of photogenerated electron–hole pairs. All these results evidently indicate the technological potency of vertical Schottky structure as an effective configuration for achieving high-performance optoelectronic devices.

2. Experiment

2.1. Fabrication of photodetectors

The multilayer MoS2 was mechanically exfoliated from commercially available crystals (purchased from SPI supplies). The layers were distinguished by the observation under optical microscopy and Raman spectroscopy on a 300 nm thick SiO2 dielectric layer thermally grown on the highly doped p-type Si substrate. EBL (JEOL 6510 with Nanometer Pattern Generation System (NPGS)) was first employed to define the bottom electrode. The 100 nm thick ITO was achieved by magnetron sputtering (JSD300-II Magnetron) at a temperature of 100 °C as the top electrode. The image in figure 1(c) was obtained by a Bruker Multimode 8 with Scan Assist-Air probe under the peak force mode in ambient condition. The electrical and optoelectronic characterization were performed in ambient condition at room temperature using a Lake Shore probe station. The photoresponse to laser excitation measurements employed a focused laser beam (λ = 637 nm) from a Single Mode Fiber-Pigtailed LP637-SF70.

3. Results and discussion

3.1. Device structure and electrical characterization of MoS2

Figure 1(a) shows the illustrative device schematic of the fabricated photodetector with vertical Schottky junctions of Au-MoS2-ITO, while figure 1(b) displays the corresponding top-view optical microscope image of the device. It is clear that the device is constructed with multilayer MoS2 sandwiched between the top ITO electrode and the bottom Au electrode pre-deposited on a highly p-doped Si/SiO2 substrate, in which the composition and crystallinity of MoS2 can be identified with Raman spectroscopy (supporting information figure S1 is available online at stacks.iop.org/NANO/28/484002/mmedia) [13]. Further investigation by atomic force microscope as displayed in figure 1(c) confirms that the device channel is 90 nm in the thickness, being the multilayer MoS2. During the device fabrication, electron-beam lithography (EBL) is first used to define the bottom electrode, followed by the deposition of bottom electrode (Cr/Au, 15/50 nm). After the lift-off process, the multilayer MoS2 is transferred on top of the bottom Cr/Au electrode. The
The mechanical transfer process of MoS$_2$ onto the device substrate is performed within a glove box as described in supporting information figure S2. The top electrode of 100 nm thick ITO is then deposited by magnetron sputtering at a temperature of 375 K. The transmission spectrum of the deposited ITO film is also depicted in supporting information figure S3, which demonstrates an acceptable transmittance (above 80%) ranging from 500 to 1000 nm. Importantly, figure 1(d) shows the $I$–$V$ characteristics of the photodetector measured in dark at room temperature. The diode-like behavior with a considerable current rectification (i.e. forward/reverse current ratio, $I_F/I_R > 10^3$ at $|V| = 0.5$ V) demonstrates that there exists a decent Schottky barrier. Since an ultralow reverse dark current of $\sim 10^{-12}$ A is achieved at zero bias voltage here, it can significantly improve the detectivity by distinguishing the photocurrent from the dark current. In addition, the $I$–$V$ characteristics can be fitted well with an ideal diode equation of $I = I_s (e^{qV/kT} - 1)$, where $I$ is the diode current, $I_s$ is the reverse saturation current, $V$ is the voltage bias, $q$ is the electron charge, $k$ is the Boltzmann constant, $T$ is the temperature and $n$ is the ideality factor of the diode. By fitting the experimental data, an ideality factor of 2.1 is obtained, which is slightly higher than the ideal value of 2, suggesting few impurities at the interface or in the multilayer MoS$_2$ channel, even though these impurities are often observed in many photovoltaic materials [14]. The figure 1(d) inset also displays the logarithmic plot of $I$–$V$ characteristics of the device, revealing the importance of contact barriers established between electrodes and MoS$_2$ for suppressing the dark current of the photodetectors achieved in this work.

To shed light on the operation mechanism of the vertical Schottky junction based photodetector, simplified energy band diagrams are employed to describe the photocurrent generation process taken place in dark and under illumination as depicted in figure 2. In general, for the Schottky-barrier structure, there are three distinct operational states, which are known as the equilibrium condition (i.e. $V = 0$), forward bias condition (i.e. $V > 0$) and reverse bias condition (i.e. $V < 0$). These three states can be independently activated with a suitable bias voltage such that the surface barrier defined at the MoS$_2$/Au interface can be precisely modulated. Explicitly, figure 2(a) illustrates the equilibrium energy band diagram of the fabricated Schottky-barrier structure with a stack of Au-MoS$_2$-ITO at $V = 0$. Because the work function of Au metal (5.1 eV) is higher than that of MoS$_2$ (4.6 eV), the electrons in MoS$_2$ would be injected into the Au electrode in order to maintain the equilibrium of Fermi energy when they are contacted with each other. This way, an ideal built-in electric field of 0.5 eV could be formed in principle which can reduce the dark current at zero bias voltage [15]. Moreover, in

![Figure 1](image1.png)

**Figure 1.** (a) Three-dimensional schematic view of the fabricated device. (b) The typical optical image of the device. The scale bar is 10 $\mu$m. (c) The AFM image of the multilayer MoS$_2$ transferred onto the substrate with the height profile measured for the deposited film thickness. (d) $I$–$V$ characteristics of the fabricated photodetector measured in the dark at room temperature. The inset gives the same $I$–$V$ curve in the logarithmic plot, showing remarkable rectifying $I$–$V$ characteristics.
this particular device configuration, the reflected light coming from the bottom Au electrode can further enhance the detectivity of the photodetector. On the other hand, the top ITO electrode employed is a heavily doped (n-type) transparent conductive oxide with a relative low work function of 4.5 eV; therefore, this ITO would form an almost perfect Ohmic contact with the intrinsically n-type MoS2 channel. Notably, the above discussion ignores the influences...
of any interface state, which may lead to Fermi level pinning and reduce effective work function of both Au and ITO [16]. Also, as compared with the forward and reverse bias conditions, there exhibits the lowest dark current in this equilibrium state due to the large surface barrier. Under illumination, the electrons in the valence band of MoS$_2$ are excited into the conduction band and separated near or in the depletion region owing to the build-in electric field, which forms an effective photovoltage driving the external circuit. The ultralow dark current can then be ignored as compared with the relatively large photocurrent as shown in figure 2(d). Meanwhile, the photocurrent at the forward bias (figure 2(e)) can be even larger than that of the equilibrium state (figure 2(d)) due to the steep downward energy band bending near Au/MoS$_2$ interface, which accelerates the separation of the electron–hole pairs at $V > 0$ within the breakdown voltage. In contrary, as shown in figure 2(c), the surface barrier is manipulated to be much smaller at $V < 0$, resulting in the larger thermionic current and thus decreasing the detectivity of the device [17, 18]. Therefore, the vertical Schottky junction is confirmed to play a crucial role in the photodetection studied in this work, in which this barrier can be modulated precisely by the bias voltage for different operation modes of the detector.

3.2. Performance evaluation of the MoS$_2$ photodetector

In order to verify the above mentioned photodetecting mechanism, the photocurrent generation of the device can be mapped by scanning photocurrent microscopy at $V = 0$, where a focused laser beam is scanned over the entire device and the corresponding photocurrent at each microscopic location is then extracted, as shown in figure 3(a). It is obvious that the spatially resolved photocurrent map reveals prominent photocurrent generation in the overlapping region between Au and MoS$_2$ stacked in the vertical manner, while there is almost no photocurrent generation in the other regions. All these imply that there is an effective Schottky-barrier established between Au and MoS$_2$ whereas an Ohmic contact between ITO and MoS$_2$, which is consistent with the analysis concluded in figure 2. Furthermore, corresponding output characteristics of the device under the 637 nm laser illumination is presented in figure 3(b). An impressively high photocurrent to dark current ratio of $I_{\text{light}}/I_{\text{dark}} \approx 10^6$ is obtained at a power density of 3.38 mW mm$^{-2}$, suggesting an excellent sensitivity to the incident photon with the wavelength.

![Figure 4.](image-url) (a) and (b) present the rising and decaying edge of the detector at $V = 0$ with a laser of 637 nm. The time constants of 64 $\mu$s and 51 $\mu$s are obtained, respectively. (c) Stability test of the device. During 12 000 cycles of the switching operation, the photocurrent and dark current of the device remain unchanged. (d) The photocurrent measured as a function of the incident photon with the wavelength.
free electrons are generated and then driven by the built-in potential, which is independent of the illumination power (if the illumination power is enough), forming larger photocurrents at zero voltage bias. However, when the voltage bias is precisely offset against the open circuit voltage, the photoinduced carriers are unable to establish an effective flow and then contribute to an insignificant current.

At the same time, responsivity \( R \) and specific detectivity \( D^* \) are two critical parameters employed to evaluate the sensitivity of detectors. \( R \) is given by \( \frac{I_p}{P} \), where \( I_p \) designates the photocurrent in the detector and \( P \) represents the power density of illumination. Assuming that short noise is the major factor contributing to the dark current, the detectivity can be expressed as \( D^* = RA^{1/2}/(2eF)^{1/2} \) [19, 20], where \( R \) is the responsivity, \( A \) is the area of the photodetector and \( I \) is the dark current. As demonstrated in figures 3(e) and (f), the responsivity \( R \) and detectivity \( D^* \) of the photodetector are found to be \( \sim 1 \text{ A W}^{-1} \) and \( \sim 10^{10} \text{ Jones} \), respectively, under various illumination power densities ranging from 0.003 38 all the way to 3.38 mW mm\(^{-2} \) at \( V = 0 \), indicating that the vertical junction based device is extremely sensitive to the incident illumination.

Moreover, the temporal photoresponse is another crucial performance parameter for photodetectors, determining the capability of distinguishing a fast signal. To study the response speed, a high-speed digital oscilloscope was used to record the current following the high-frequency modulated laser. Figures 4(a) and (b) show one entire cycle of the time-resolved photoresponse with both rising and decaying edges at an illumination density of 3.38 mW mm\(^{-2} \) (\( \lambda = 637 \text{ nm} \)). The rise time \( t_{\text{rise}} \) and decay time \( t_{\text{decay}} \) of photodetector is defined as the total time required for the output to rise from 10% to 90% of the pulse peak and to fall from 90% to 10%, respectively. The rising and decaying edges can also be perfectly fitted by single exponential functions, which yield the time constants of 64 and 51 \( \mu \text{s} \) for our detectors, being much faster than other MoS\(_2\) based detectors reported in the literature [10–12]. This superb fast photoresponse can be ascribed to two reasons: (1) the shorter channel length configured in the vertical direction can substantially decrease the carrier transit time. (2) The built-in electric field can further accelerate the photogenerated electron–hole pair separation. Furthermore, the signals recorded by the oscilloscope do not exhibit any noticeable change after 12 000 cycles of operation as depicted in figure 4(c), indicating the excellent stability and reliability of the photodetector. Besides, based on the band gap of multilayer MoS\(_2\) (i.e. 1.2 eV) [20–22], the fabricated device should be able to detect photons with the wavelength up to 1030 nm, which is longer than one of 850 nm corresponding to the indirect bandgap transition [12, 23]. Indeed, as shown in figure 4(d), the vertical Schottky junction based detector studied here displays a decent photocurrent and responsivity with the wavelength ranging from visible to 1000 nm (the responsivity as function of the wavelength is shown in figure S6). Such wide spectral photoresponse can as well be attributed to the low dark current of \( 10^{-12} \text{ A} \) and the shorter channel length for the efficient extraction of photocurrent, in addition to the relatively small bandgap of multilayer MoS\(_2\) channel. All these results clearly present an effective route to achieve the excellent and comprehensive optoelectric performance for vertical Schottky-barrier photodetectors based on 2D materials.

4. Conclusion

In summary, a simple MoS\(_2\) based photodetector utilizing vertical Schottky junctions of Au-MoS\(_2\)-ITO is demonstrated. This simple unique device structure can enable the excellent photodetector performance, including an ultralow dark current of \( 10^{-12} \text{ A} \) and a superb fast photoresponse of \( \sim 64 \mu \text{s} \) at zero bias voltage. The responsivity and detectivity can also reach \( \sim 1 \text{ A W}^{-1} \) and \( \sim 10^{10} \text{ Jones} \), respectively. Importantly, the detection spectrum is superior to most recently reported pure MoS\(_2\) photodetectors. These detectors can exhibit a wide detection spectrum with the wavelength ranging from visible to near-infrared (i.e. 0.7–1 \( \mu \text{m} \)), revealing its potential applications in optoelectronic devices. All these results indicate clearly the technological potency of multilayer MoS\(_2\) configured in vertical Schottky junctions with Au electrode for the development of high-performance photodetectors.

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Author Contributions

FG and HF contributed equally to this work. WH and JW conceived and supervised the research. FG fabricated the devices. FG and HF performed the measurements. WH, JW, FG and HF wrote the paper. All authors discussed the results and revised the manuscript.

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