Large Lateral Photovoltaic Effect in MoS$_2$/GaAs Heterojunction

Lanzhong Hao 1,2*, Yunjie Liu 1, Zhide Han 1, Zhijie Xu 1 and Jun Zhu 2

Abstract
Molybdenum disulfide (MoS$_2$) nanoscaled films are deposited on GaAs substrates via magnetron sputtering technique, and MoS$_2$/GaAs heterojunctions are fabricated. The lateral photovoltaic effect (LPE) of the fabricated MoS$_2$/GaAs heterojunctions is investigated. The results show that a large LPE can be obtained in the MoS$_2$/n-GaAs heterojunction. The LPE exhibits a linear dependence on the position of the laser illumination and the considerably high sensitivity of 416.4 mV mm$^{-1}$. This sensitivity is much larger than the values in other reported MoS$_2$-based devices. Comparatively, the LPE in the MoS$_2$/p-GaAs heterojunction is much weaker. The mechanisms to the LPE are unveiled by constructing the energy-band alignment of the MoS$_2$/GaAs heterojunctions. The excellent LPE characteristics make MoS$_2$ films combined with GaAs semiconductors promising candidates for the application of high-performance position-sensitive detectors.

Keywords: MoS$_2$, GaAs, Photovoltaic, Heterojunction, Interface

Background
Due to its excellent properties, molybdenum disulfide (MoS$_2$) is being investigated as one typical kind of two-dimensional materials to develop next-generation microelectronic devices and optoelectronic devices [1–5]. Unlike graphene, MoS$_2$ has obvious band gap and its band gap decreases with increasing layer numbers [6]. The presence of the obvious band gap allows the fabrication of the MoS$_2$ transistors with an on/off ratio exceeding $10^8$ and the photodetectors with high responsivity [7, 8]. Recently, MoS$_2$ combined with other semiconductors has attracted much interest, such as GaAs, Si, and GaN [9–13]. These designed heterostructures supply feasible technical route for MoS$_2$-based materials to develop practically applicable optoelectronic devices. Among all these bulk semiconductors, GaAs has a suitable direct band gap of $\sim 1.42$ eV and high electron mobility ($\sim 8000$ cm$^2$ V$^{-1}$ s$^{-1}$). Lin et al. fabricated MoS$_2$/GaAs solar cells with a power conversion efficiency over 9.03% [9]. Further, Xu et al. reported a MoS$_2$/GaAs self-driven photodetector with the extremely high detectivity of $3.5 \times 10^{13}$ Jones [10]. In previous reports, the studies on MoS$_2$/GaAs heterostructures have been mainly focused on the application in the area of solar cells and photodetectors. However, the MoS$_2$/GaAs as a position-sensitive detector (PSD) based on the lateral photovoltaic effect (LPE) has been reported rarely. Different from the ordinary longitudinal photovoltaic effect, the LPE originates from the lateral diffusion and recombination of the photon-generating carriers in the inversion layer at the interface [14–18]. In the LPE effect, a lateral photovoltage (LPV) can be obtained and it changes linearly with the laser spot position on the active region of the device surface. These characteristics make LPE very useful in developing high-performance PSDs and have been studied widely in the area of robotics, biomedical applications, process control, position information systems, and so on.

In this work, MoS$_2$ thin films with different thickness are deposited on the surface of n-p-GaAs substrates via magnetron sputtering technique. A large LPE is observed in the fabricated MoS$_2$/n-GaAs heterojunction, and the sensitivity reached 416.4 mV mm$^{-1}$. Our results further show that the LPE exhibits obvious dependence on the carrier types of the GaAs substrates and the thickness of the MoS$_2$ films. Through the construction...
of the energy-band alignment at the interface, the mechanisms to the LPE in the devices are proposed.

**Methods**

MoS$_2$ thin films were deposited on (100)-oriented GaAs substrates using the DC magnetron sputtering technique. The MoS$_2$ powders (purity, ~99%) were cold-pressed into a disk under the pressure of 20.0 MPa. The as-fabricated disk ($\Phi 60.0 \text{ mm} \times 4.5 \text{ mm}$) was used as the target during sputtering. The $n$-$p$-GaAs substrates were used in our experiments, respectively. Before the deposition, the substrates were ultrasonically cleaned in sequence by alcohol, acetone, and de-ionized water. Subsequently, MoS$_2$ thin films with different thickness ($d_{\text{MoS}_2} = \sim 10, 30, 50, 90 \text{ nm}$) were grown on the GaAs substrates at the temperature of 400 $^\circ$C, respectively. During the deposition, the working pressure and power were kept at 1.0 Pa and 10.0 W, respectively. As a reference, MoS$_2$ thin films were also deposited on intrinsic GaAs ($\text{i-GaAs}$) substrates under the same condition. Finally, about 300-$\mu$m In pads with a diameter of 0.5 mm as electrodes were pressed on the MoS$_2$ film.

The MoS$_2$ films were characterized using Raman spectroscopy (HORIBA, HR800) with the excitation wavelength of 488 nm. The surface of the sample was scanned by an atomic force microscope (AFM). X-ray photoemission spectroscopy (XPS) was performed by a Kratos Axis ULTRA spectrometer with a monochromatic Al K$_\alpha$ X-ray source. The deposition rate was obtained by the thickness from the cross-sectional scanning electron microscope (SEM) (Additional file 1: Figure S1) and the deposition time, then each film thickness was determined by the deposition rate and each deposition time. The transmission spectra were measured by Shimadzu UV-3150 spectrophotometer. Ultraviolet photoelectron spectroscopy (UPS) was performed using an unfiltered He-I (21.22 eV) gas discharge lamp. LPVs were measured using a Keithley 2000 voltmeter and three-dimensional electric motorized stage with a laser of 650-nm wavelength as the illumination source. The current-voltage ($I$-$V$) curves were measured with a Keithley 2400 SourceMeter.

**Results and Discussion**

Figure 1 shows the Raman spectrum of the MoS$_2$ film on the GaAs substrate. Besides the peak of the GaAs substrate at $\sim 287.1 \text{ cm}^{-1}$, two characteristic MoS$_2$ Raman peaks can be seen, the $A_{1g}$ mode at $\sim 406.7 \text{ cm}^{-1}$ and $E_{12g}$ mode at $\sim 378.9 \text{ cm}^{-1}$. The right two insets show the illustration of the atomic vibration in MoS$_2$. The $A_{1g}$ mode corresponds to the S atoms oscillating in antiphase along the out-of-plane direction, and the $E_{12g}$ mode corresponds to the S and Mo atoms oscillating in antiphase parallel to the crystal plane. As shown in the figure, the Raman peak corresponding to the $A_{1g}$ mode is preferentially excited for the film. According to our measurements, the intensity ratio of $A_{1g}/E_{12g}$ is about 2.1. These Raman characteristics are similar with other reported results about MoS$_2$ thin films [19]. The $A_{1g}$ mode corresponds to the S atoms oscillating in antiphase along the out-of-plane direction, and the $E_{12g}$ mode corresponds to the S and Mo atoms oscillating in antiphase parallel to the crystal plane. As shown in the figure, the Raman peak corresponding to the $A_{1g}$ mode is preferentially excited for the film. According to our measurements, the intensity ratio of $A_{1g}/E_{12g}$ is about 2.1. These Raman characteristics are similar with other reported results about MoS$_2$ thin films [19]. The left inset shows an AFM topographic image of the 40-nm MoS$_2$ film grown on the GaAs substrate. From the figure, we can see that the surface of the film is composed of dense cone-like grains. According to the measurements, the root-mean-square (RMS) roughness of...
the film is about 1.7 nm, and the average size of grains is about 76.3 nm in diameter. These grains on the surface could decrease the surface reflection to the external light and enhance the light absorption of the fabricated device.

Figure 2 shows the XPS spectra of the MoS$_2$ film. As shown in Fig. 2a, the peaks at 229.3 and 232.5 eV are related to the Mo 3d$_{5/2}$ and Mo 3d$_{3/2}$ orbitals, respectively. As shown in Fig. 2b, S 2p$_{3/2}$ and S 2p$_{1/2}$ orbitals of divalent sulfide ions (S$^{2-}$) are observed at 162.2 and 163.3 eV, respectively. The results are in good agreement with the reported values for the MoS$_2$ crystal [17, 18].

Figure 3a shows the longitudinal $I$-$V$ curve of the fabricated MoS$_2$/n-GaAs heterojunctions. Two In electrodes with the diameter of about 0.5 mm were pressed on the surface of the MoS$_2$ film and the backside of the GaAs, respectively. The inset shows the schematic circuit for the longitudinal measurements. The forward voltage is defined as a positive bias voltage applied on the top In electrode. As shown in the figure, the fabricated MoS$_2$/n-GaAs heterojunction shows obvious rectifying behaviour. The rectifying ratio ($I_+/I_-$) measured at ± 1.0 V is about 520. In our experiments, both In/MoS$_2$ and In/GaAs belong to ohmic contacts and their $I$-$V$ curves are almost linear. Thus, the rectifying $I$-$V$ characteristic in the heterojunction is mainly originated from the MoS$_2$/GaAs contact. Figure 3b shows the transverse $I$-$V$ curve of the fabricated MoS$_2$/n-GaAs heterojunctions. Two In electrodes with the diameter of about 0.5 mm were pressed on the surface of the MoS$_2$ film. The top inset shows the schematic circuit for the transverse measurements. From the figure, the $I$-$V$ curve shows slightly nonlinear increase of the currents with increasing voltages. This indicates that an inversion layer at the MoS$_2$/n-GaAs interface is formed [18]. The bottom inset shows the $I$-$V$ curves of the single MoS$_2$ films on the intrinsic GaAs substrate. From the figure, an almost linear $I$-$V$ curve can be seen, further indicating the ohmic nature of the In/MoS$_2$ contact. At the voltage of +0.5 V, the current of the single MoS$_2$ is about 3.1 $\times$ 10$^{-2}$ $\mu$A, much smaller than the value in the MoS$_2$/n-GaAs, about 2.3 $\mu$A. Thus, compared to the MoS$_2$ film, the inversion layer at the MoS$_2$/n-GaAs interface supplies a path with a much lower resistivity for carrier transport during the transverse measurements of the MoS$_2$/n-GaAs heterojunction.

Figure 4a shows the schematic circuit for the measurement of the LPE of the fabricated MoS$_2$/GaAs heterojunction. Two In electrodes with the diameter of 0.5 mm are pressed on the surface of the MoS$_2$ film to perform the measurements of the LPE. The distance (2L) between the electrodes is ~ 1.0 mm. During our measurements, electrodes A and B were connected to the positive and negative probes of a Keithley 2000 voltmeter, respectively. Figure 4b shows the LPE curves of the MoS$_2$/n-GaAs and MoS$_2$/p-GaAs heterojunctions, respectively. The thickness of the MoS$_2$ films is ~ 30.0 nm. When the surface of the MoS$_2$ film is partially illuminated by a laser spot with the diameter of about 0.1 mm, a large LPE can be observed in the MoS$_2$/n-GaAs heterojunction. As shown in the figure, the LPE shows an approximately linear dependence on the laser spot position when the laser spot moves between electrodes A and B on the MoS$_2$ surface. From the figure, we can see that the LPV depends on the position of the laser spot. This can be fitted with the diffusion theory [16],

$$LPV = K_0 \left[ \exp\left(-\frac{|L-x|}{d}\right) - \exp\left(-\frac{|L+x|}{d}\right) \right]$$

where $K_0$, $2L$, $d$, and $x$ represent a proportionality coefficient, the distance between two electrodes, the carrier diffusion length, and the laser spot position, respectively. The well-fitted results in the figure clearly suggest that the LPE in the MoS$_2$/n-GaAs heterojunction arises from the lateral diffuse flow and recombination of the excited carriers away from the laser position. As shown in the figure, the LPV value is zero when the light spot is at the centre between two electrodes, which can be attributed to the diffusion symmetry of the carriers. When the light
position is close to the A electrode, the LPV is positive and vice versa. This indicates that the LPE in the MoS$_2$/n-GaAs heterojunction is caused by the hole-type photoexcited carriers. The maximum LPV is obtained when the laser illumination is closest to the electrodes. According to our measurements, the maximum lateral photovoltage (LPV$_\text{max}$) is about 208.2 mV in the linear region of the MoS$_2$/n-GaAs heterojunction. Comparatively, the LPV of the MoS$_2$/p-GaAs heterojunction is much smaller and its LPV$_\text{max}$ is only 7.3 mV, as shown in the figure. From the figure, we can see that the LPE of the MoS$_2$/p-GaAs heterojunction is determined by the electron-type photoexcited carriers. Additionally, nonlinear LPE characteristics of the MoS$_2$/p-GaAs heterojunction can be seen from the figure when the laser spot moves between the A and B electrodes.

Figure 5 shows the LPE sensitivity of the MoS$_2$/n-GaAs heterojunction as a function of the laser power and the thickness ($d_{\text{MoS}_2}$) of the MoS$_2$ film. The sensitivity is defined by $S = \text{LPV}_{\text{max}}/L$. Obviously, the $S$ increases drastically with increasing laser power initially but then slowly saturates when the power further increases. This saturation could be caused by the rapidly increasing recombination rate of the photoexcited holes with increasing laser intensity in the illuminated region [20]. As shown in the figure, an obvious LPE and a high sensitivity can be obtained even under the weak laser illumination of 100.0 μW. From the figure, a significant dependence of the sensitivity on the thickness of the MoS$_2$ films can be seen. When $d_{\text{MoS}_2} = \sim 10.0$ nm, $S = 165.4$ mV mm$^{-1}$ under the laser illumination of 100.0 μW. With increasing film thickness, $S$ increases gradually. When $d_{\text{MoS}_2} = 30.0$ nm, $S$ reaches 416.4 mV mm$^{-1}$. This sensitivity is much larger than the reported MoS$_2$/Si devices [17, 18]. After $d_{\text{MoS}_2} > 30.0$ nm, $S$ decreases with further increasing MoS$_2$ thickness. When $d_{\text{MoS}_2} = 90.0$ nm, $S = 283.3$ mV mm$^{-1}$. Thus, to obtain the largest LPE and sensitivity, there is an optimum thickness of the MoS$_2$ film in the fabricated MoS$_2$/n-GaAs, about 30.0 nm.

Figure 6a presents the UPS spectra of the MoS$_2$ film on the Si substrate. The work function ($W$) of the film
can be calculated from the difference between the cutoff of the highest binding energy and the photon energy of the exciting radiation. From the figure, \( W = 5.24 \text{ eV} \) can be obtained. The distance \( \Delta E \) between the valence band \( (E_V) \) and the Fermi level \( (E_F) \) of MoS\(_2\) film can be extracted from the onset energy, as shown in the inset. The \( \Delta E \) for the MoS\(_2\) film is about 0.51 eV. Using the data from the transmittance spectrum of the MoS\(_2\) film on quartz substrate, \( (\alpha h \nu)^2 \) is plotted as a function of photon energy \( h \nu \), wherein \( h \) is the Planck constant and \( \nu \) is the photon frequency. The \( \alpha \) is the absorption coefficient, calculated by \( \alpha d = \ln(1/T) \) \[21\], wherein \( d \) and \( T \) are the thickness and transmittance of the film, respectively. The band gap \( (E_g) \) of the film can be determined.
from the intercept of the line on the $hv$ axis, $E_g = 1.54 \text{ eV}$, as shown in Fig. 6b. Based on these energy-band parameters, the $p$-type behaviour of the MoS$_2$ film can be determined, which can be further proved by Hall measurements. The Hall results show that the concentration of the hole-type carrier and the mobility are about $3.8 \times 10^{15} \text{ cm}^{-3}$ and $11.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively.

In order to clarify the mechanisms of the LPE in the MoS$_2$/GaAs heterojunctions, the energy-band diagrams at the interface are constructed based on the results in Fig. 6. Here, $E_g = 1.42 \text{ eV}$ and $E_F = 4.17 \text{ eV}$ for $n$-GaAs are taken into account to construct the band structure [22]. When the MoS$_2$ film is deposited onto the GaAs substrate, the electrons flow from the GaAs into the MoS$_2$ film at the interface due to the higher $E_F$ of the GaAs. The flowing process stops when Fermi levels are equal and the MoS$_2$/GaAs $p$-$n$ junction is fabricated, as shown in Fig. 7a. Consequently, a built-in field ($E_{bi}$) is formed at the interface of the MoS$_2$/GaAs heterojunction. $E_{bi} = [E_F(n\text{-GaAs}) - E_F(MoS_2)]/e = 1.07 \text{ V}$ and it points from the GaAs to the MoS$_2$ film. Thus, the asymmetric longitudinal $I$-$V$ curve is shown in Fig. 3a. Due to the existence of a strong $E_{bi}$, large quantities of hole-type carriers are accumulated near the interface and an inversion layer is formed in the $n$-GaAs substrate near the interface, as shown in Fig. 7b. Similar with the two-dimensional hole gas (2DHG) [23], the inversion layer could exhibit the feature of high conduction due to the high sheet concentration of the holes ($p^+$). From Fig. 3b, we can see that the conduction of the inversion layer is much larger than the MoS$_2$ film. Thus, the conduction between two electrodes on the same side of the MoS$_2$ film is dominated by the inversion layer rather than the MoS$_2$ film. When the junction is partially illuminated by the laser, the light is absorbed and the electron-hole
pairs in the MoS2 film and GaAs can be induced, respectively, as shown in Fig. 7c. However, it can be expected that most of the laser is absorbed by the GaAs substrates due to its much larger thickness and smaller band gap. Under the laser illumination, the electron-hole pairs can only be excited in the illuminated region and spatially separated by the $E_{bi}$. Due to the orientation of the $E_{bi}$ pointing from GaAs to MoS2, the photoexcited holes flow towards the interface and enter into the inversion layer in the GaAs, as shown in Fig. 7c. The photoexcited holes in the inversion layer diffuse laterally away from the illuminated spot to the two electrodes. The concentration of the excited holes collected by the two electrodes is different for different distances from the illuminated spot. Thus, a large LPV is formed between the electrodes, and the LPE is observed in the heterojunction. This is in accord with the fitted results from Fig. 3b, and the LPE induced by the diffusion of the photoexcited electrons is obtained in the MoS2/n-GaAs heterojunction, as shown in Fig. 4. However, the concentration of the accumulated carrier in the inversion layer might be lower due to the weak $V_{bi}$ of only 0.08 V in the p-p heterojunction compared to the p-MoS2/n-GaAs junction. This increases the difficulties of the transport of the photoexcited electrons in the inversion layer. Seriously, the Schottky barriers can be formed between the n-type inversion layer and the p-MoS2 film, as shown in Fig. 7e. These characteristics of the p-MoS2/p-GaAs junction suppress the collection of the photoexcited electrons at the electrodes. As a result, the LPE could be reduced largely. As shown in Fig. 4b, the LPV$_{max}$ for the p-p junction is only 7.3 mV while it reaches 208.2 mV in the p-n junction.

It usually happens in the reported heterojunction-type PSDs that the LPE can be tuned by changing the thickness of the cap layers [20]. This can be well understood by considering the recombination of the photoexcited carriers in the film and the evolution of the built-in field. In the MoS2/GaAs p-n junction, the recombination of photon-generated carriers can be enhanced due to the long transporting path in the thick MoS2 film before near the interface of the heterojunction and the inversion layer is formed. Thus, the LPE induced by the diffusion of the photoexcited electrons is obtained in the MoS2/GaAs p-p heterojunction, as shown in Fig. 4. However, the concentration of the accumulated carrier in the inversion layer might be lower due to the weak $V_{bi}$ of only 0.08 V in the p-p heterojunction compared to the p-MoS2/n-GaAs junction. This increases the difficulties of the transport of the photoexcited electrons in the inversion layer. Seriously, the Schottky barriers can be formed between the n-type inversion layer and the p-MoS2 film, as shown in Fig. 7e. These characteristics of the p-MoS2/p-GaAs junction suppress the collection of the photoexcited electrons at the electrodes. As a result, the LPE could be reduced largely. As shown in Fig. 4b, the LPV$_{max}$ for the p-p junction is only 7.3 mV while it reaches 208.2 mV in the p-n junction.

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they are collected by the electrodes. This reduces the LPE of the heterojunctions. Reversely, a thinner film can greatly decrease the recombination, which causes the increase of the LPE. However, when the MoS$_2$ thickness is smaller than the critical value, the $E_{\text{bi}}$ at the interface decreases with further decreasing MoS$_2$ thickness [24]. This can reduce the separation of photon-generated electron-hole pairs, and the LPE decreases. Thus, there is an optimum thickness of the MoS$_2$ film for obtaining the highest LPE, about 30 nm.

Conclusions
In summary, MoS$_2$ thin films were deposited on the surface of the GaAs substrate via magnetron sputtering technique. A large LPE was obtained in the fabricated MoS$_2$/n-GaAs heterojunction, and the dependence of the LPV on the position of the laser illumination showed good linearly. Due to the formation of the strong built-in field at the interface, the MoS$_2$/n-GaAs heterojunction exhibited a high sensitivity of 416.4 mV mm$^{-1}$, while the value was only 7.3 mV mm$^{-1}$ for the MoS$_2$/p-GaAs. Our results further showed that the LPE exhibited obvious dependence on the thickness of the MoS$_2$ films and about 30.0 nm was the optimum thickness of the MoS$_2$ film to acquire the highest LPE in the fabricated MoS$_2$/n-GaAs heterojunctions. The mechanisms to the LPE in the MoS$_2$/GaAs devices were clarified based on the energy-band alignment at the interface.

Additional file

Additional file 1: Large lateral photovoltaic effect in MoS$_2$/GaAs heterojunction. Figure S1. Cross-sectional SEM images of the as-grown MoS$_2$ film. (DOCX 731 kb)

Abbreviations
ΔE: Distance between $E_c$ and $E_v$; $d_{\text{MoS}_2}$: Thickness of the MoS$_2$ film; $E_{\text{bi}}$: Built-in field; $E_c$: Conduction band level; $E_v$: Fermi energy level; $E_g$: Energy-band gap; $E_F$: Valence band level; $I$: Current-voltage; LPE: Lateral photovoltaic effect; LPV: Lateral photovoltage; LPV$_\text{max}$: Maximum lateral photovoltage; MoS$_2$: Molybdenum disulfide; PSD: Position-sensitive detector; UPS: Ultraviolet photoelectron spectroscopy; $W$: Work function

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Authors’ Contributions
LH participated in the fabrication of MoS$_2$/GaAs heterojunctions, analysed the data, and wrote the manuscript. YL interpreted the data for Raman spectrum. ZH performed the electrical measurements. ZX participated in the construction of the energy-band diagram. JZ discussed the mechanisms of the LPE. All authors read and approved the final manuscript.

Authors’ Information
LH is an associate professor in materials physics and PhD degree holder specializing in electronic thin films and integrated devices. YL is an associate professor and PhD degree holder in the growth of thin films. ZH is a lecturer studying on optical-electronic materials and optoelectronic devices. ZY is a lecturer studying on composites materials. JZ is a professor and PhD degree holder specializing in physical vapour deposition technique.

Ethics Approval and Consent to Participate
Not applicable.

Consent for Publication
Not applicable.

Competing Interests
The authors declare that they have no competing interests.

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