Article

Low-Powered Photodetector Based on Two-Dimensional $\text{InS}_{0.3}\text{Se}_{0.7}/\text{WS}_2$ Heterostructure

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Abstract: Photodetectors based on two-dimensional (2D) materials have great potential applications in the field of new energy, such as fuel cells, solar cells, and other fields. Van der Waals (vdW) heterojunction photodiodes are expected to be one of the promising applications of two-dimensional materials due to the photoelectric properties without consideration of lattice mismatch. High-efficiency photoelectric sensors based on two-dimensional materials have great significance to reducing the energy consumption of devices. Here, we build a complex vdW heterostructure by combining $\text{InS}_{0.3}\text{Se}_{0.7}$ with another suitable 2D material WS_2 . Few-layer graphite was used as electrodes to enhance the optoelectronic performance of indium monochalcogenides. Evident photocurrent is observed in the $\text{InS}_{0.3}\text{Se}_{0.7}/\text{WS}_2$ vdW heterostructure device arising from the formed p–n junction at the interface. The uniformity and photoresponse of the $\text{InS}_{0.3}\text{Se}_{0.7}/\text{WS}_2$ vdW heterostructure has been further investigated by the photocurrent mapping. It shows that the entire photovoltaic current was originated from the $\text{InS}_{0.3}\text{Se}_{0.7}/\text{WS}_2$ vdW heterojunction by scanning photocurrent microscope images. Furthermore, the response speed is enhanced at small bias voltage. The transient photoresponse can be well reproduced in almost 100 cycles, indicating the good repeatable optoelectronic performance. Our study indicates that the as-prepared $\text{InS}_{0.3}\text{Se}_{0.7}/\text{WS}_2$ vdW heterostructures are attractive building blocks for photodetectors application. Our findings will open up a new way to further develop high-performance, low-power, and energy-efficient photodetectors based on indium monochalcogenides.

Keywords: 2D materials; photodetector; indium monochalcogenides; van der Waals heterostructure

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1. Introduction

In recent years, the development of advanced semiconductors based on two-dimensional (2D) materials is expected to solve the energy problem, which is caused by the huge growth of the number of sensors bringing by high integration and smaller size of electronic devices [1–4]. 2D semiconductor devices with low power consumption and high performance can be widely used in new energy fields, such as photocatalysis, fuel cells, solar cells, and others [3,5–11]. Photodetectors are expected to be one of the promising applications of materials due to the photoelectric properties produced in single- or multilayer 2D materials. Especially, the low power photodetector with good visible light response, high stability, and fast photoresponse is very crucial for low-power applications, such as future internet of things, wearable electronics, and flexible electronics [12–16].

The complex heterostructures composed of different 2D layered materials open up a new platform for electronics and optoelectronics because of these novel and extraordinary properties, such as tunable bandgaps, passivated surfaces, and high mobility [12–14]. 2D materials usually possess different carrier polarities; when combining with different 2D materials to form the vdW heterostructures, the heterostructures can generate a pronounced photovoltaic effect [10,17,18]. In the past decade, various photodetectors based on 2D materials have been proven to have superior performance [19–22]. More recently, indium

monochalcogenides InX (InS, InSe, or InS_xSe_{1-x}) have attracted much attention due to their excellent stability, high carrier mobility, and moderate band gap, showing promising applications in storage devices, optical sensors, thermoelectric devices, and low-cost and environmentally friendly solar cells [23–30]. Previous research on InS_xSe_{1-x} alloys shows that the bandgap shifts from ~1.27 to ~1.42 eV as S composition increases from 0 to 0.3 without changing crystal structure [31]. Interestingly, when InSe is fabricated from single-layer to multilayer with thickness larger than seven layers, its band structure changes from indirect gap to direct gap [23,28]. However, the photoelectric properties are significantly hindered by the little absorption of light induced by indirect band gap. On the other hand, tungsten disulfide (WS₂) has inspired great efforts in optoelectronics because of their suitable and direct band gap [32–37]. Therefore, WS₂ can act as one active absorber to construct many kinds of heterojunctions. Typically, the semiconductor heterojunctions possess three kinds of band arrangement, i.e., Type I—straddling, Type II—staggered, and Type III—broken-gap [20]. Type II possesses alternating band gap alignment between conduction bands and valence bands of the heterojunctions [20,37,38], and thus brings the decrease of the band gap and significantly increases carrier recombination rate because of the built-in electric field, leading to a wider detection wavelength range and faster response time. When the InSe layer and WS₂ layer are stacked together, a Type-II band alignment will be formed [21,32,33]. This construction of vdW heterostructures can enhance the optoelectronic performance.

In this work, the photoelectric properties of a type II vdW heterostructure based on InS_{0.3}Se_{0.7}/WS₂ were investigated. Graphene was used as electrodes by placing on the top of mechanically exfoliated few-layers InS_{0.3}Se_{0.7} flakes and a single-layer WS₂ flake. Graphene-on-semiconductor has been proven to be a quality heterojunction with efficient photoelectric conversion. Based on this hybrid heterostructure, we analyzed the performance of a photodetector. It can be responsive to the light, and the photoresponse region coincides with the overlapping area of InS_{0.3}Se_{0.7} layers and WS₂ single layer, which means all photovoltaic currents were originated from the InS_{0.3}Se_{0.7}/WS₂ heterojunction. The photocurrent waveform with a chopping frequency of 5 Hz indicated a good repeatable optoelectronic performance.

2. Results and Discussion

From the Raman spectra of the InS_{0.3}Se_{0.7} flakes, as shown in Figure S1 in Supporting Information, four typical Raman active modes at about 41 cm⁻¹, 119 cm⁻¹, 180 cm⁻¹, and 220 cm⁻¹, respectively, are observed in both areas, corresponding to the *E*, *A*_{1g}¹, *E*_{2g}¹, and *A*_{1g}² modes, which is also consistent with the previous reports [31]. The scanning electron microscopy (SEM) image (Figure S2a) of the as-synthesized InS_{0.3}Se_{0.7} crystals are of layered structures, energy-dispersive X-ray spectroscopy (EDS) (Figure S2b–e) shows the elements of In, S, and Se evenly spread and the atomic ratio of In, S, and Se elements is estimated to be about 1:0.3:0.7. The photoluminescence spectra (PL) at different temperatures are also shown in Figure S3, indicating that the PL peak gradually shifts to a long wavelength for both WS₂ flakes and InS_{0.3}Se_{0.7} flakes with increasing temperature. Figure 1a shows the optical image of the InS_{0.3}Se_{0.7}/WS₂ vdW heterostructure. The *n*-type monolayer of WS₂ flakes was mechanically exfoliated from bulk crystal onto the SiO₂ substrates (marked by the green line in Figure 1a). The *p*-type InS_{0.3}Se_{0.7} flakes with the thickness of several tens of nanometers (marked by the orange line in Figure 1a) were mechanically exfoliated onto the PDMS-based gel supplier and then were transferred on the selected areas of *n*-type monolayer WS₂ flakes using a micromanipulator and digital camera to form the vdW *p*–*n* junction [38,39]. Afterwards, two pieces of few-layer graphite were transferred on InS_{0.3}Se_{0.7} flake and WS₂ flake, respectively, using the same processes, as marked by the black line in Figure 1a [40]. The Cr/Au electrodes were patterned on the few-layer graphite using the standard electron-beam lithography process in order to increase the response speed of the InS_{0.3}Se_{0.7} and WS₂ with high photoresponsivity, by using the transparent electrodes to modify the difference work function between the electrodes and the InS_{0.3}Se_{0.7}

and WS₂ [22]. For example, for InSe/graphene, it can increase about 40 times faster than that of InSe/metal device [25,29]. This design of crystal/graphene electrical contacts could be important for high-performance optoelectronic devices.

Figure 1. (a) Optical image of InS_{0.3}Se_{0.7}/WS₂ heterostructure devices. The graphite flakes, WS₂ flakes, and InS_{0.3}Se_{0.7} flakes are marked by black line, green line, and orange line, respectively. (b) Schematic diagram of the device. (c) Current–voltage (*I*–*V*) characteristics of the InS_{0.3}Se_{0.7} and WS₂. (d) Current–voltage (*I*–*V*) characteristics of the InS_{0.3}Se_{0.7}/WS₂ heterostructure.

Figure 1b presents a schematic of the InS_{0.3}Se_{0.7}/WS₂ vdW heterostructure device. The electrodes on the thin WS₂ flake and the InS_{0.3}Se_{0.7} flakes and SiO₂/Si substrate are defined as drain, source, and back gate, respectively. A schematic drawing of the band diagram of the InS_{0.3}Se_{0.7}/WS₂ heterostructures is shown in Figure S4. Figure 1c show the current–voltage (*I*–*V*) characteristics of the graphite on the InS_{0.3}Se_{0.7} flakes and WS₂ flake, respectively. Those *I*–*V* curves are linear, the resistance of graphite on InS_{0.3}Se_{0.7} flakes is about 1600 Ω, and it is about 750 Ω for graphite on WS₂. Whereas, the output curves of InS_{0.3}Se_{0.7}/WS₂ heterostructure show clearly non-linear asymmetrical *I*–*V* characteristics with a contact resistant, as show in Figure 1d. The output curve of the InS_{0.3}Se_{0.7}/WS₂ vdW heterostructure is measured in the bias potential range of ±1 V in the dark condition and at zero gate voltage. The InS_{0.3}Se_{0.7}/WS₂ vdW heterostructures features a very low dark current down to 1 pA, which is originated from the depletion layer and the energy barrier of the *p*–*n* junction at the interface in the fabricated InS_{0.3}Se_{0.7}/WS₂ vdW heterostructure devices, as shown in the illustration in Figure 1b.

When the laser is illuminated on the *p*–*n* junction, the photogenerated electron–hole pairs can be effectively separated: electrons and holes reside preferably in the InS_{0.3}Se_{0.7} and WS₂ layers, respectively [32,33], and finally result in the observation of open-circuit voltage (*V*_{oc}) and short-circuit current (*I*_{sc}). To explore the optoelectronic properties of the as-prepared InS_{0.3}Se_{0.7}/WS₂ vdW heterostructure devices, an argon green laser at 532 nm with different power density was adopted to irradiate the device. Figure 2a shows the *I*–*V* characteristics of the InS_{0.3}Se_{0.7}/WS₂ vdW heterostructure devices under the dark

condition and different laser power densities ($P = 1.5, 2.5$, and 3 mW). As can be seen, the magnitude of the V_{oc} and I_{sc} increases gradually with increasing laser power, as indicated by blue triangles and red triangles in Figure 2b. Such increment of the V_{oc} and I_{sc} is believed to originate from the increased number of photo-generated carriers under laser irradiation with increased power. Under 15.3 mW/cm^2 illumination with the conditions of $V_{ds} = -0.6 \text{ V}$, $V_g = 0 \text{ V}$, $\lambda = 532 \text{ nm}$, the values of responsivity, detectivity were 0.25 A/W , $1.8 \times 10^{10} \text{ Jones}$, respectively.

Figure 2. (a) I - V characteristics of the InS_{0.3}Se_{0.7}/WS₂ heterostructure under dark condition and different laser power ($p = 1.5, 2.5$, and 3 mW). (b) The magnitude of the photocurrent (I_{sc}) and open-circuit voltage (V_{oc}) as a function of laser power.

To further study the optoelectronic properties of the InS_{0.3}Se_{0.7}/WS₂ vdW heterostructure devices, the photocurrent mapping is measured at zero bias potential and room temperature. The 2D plot of Figure 3 shows the intensity of photocurrent as a function of X and Y axis. By comparing with the optical image in Figure 3, it can be seen that the photocurrent is mainly generated at the overlap areas of the InS_{0.3}Se_{0.7}/WS₂ vdW heterostructure, as indicated by the blue dashed line. It is worth mentioning that no measurable photocurrent was observed in non-overlapping regions, with only the InS_{0.3}Se_{0.7} layers or single-layer WS₂, demonstrating that the two graphene electrodes do enhance the photoresponse by contributing efficient carriers. This phenomenon further demonstrates that the measured photocurrent in our InS_{0.3}Se_{0.7}/WS₂ vdW heterostructure devices is originated from the p - n junction formed at the interface, in which the photo-generated electron-hole pairs are effectively separated by the built-in field of the p - n junction. As shown in Figure S4, the depletion zone (as indicated by the green area) is formed at the interface of the InS_{0.3}Se_{0.7} flakes and WS₂ flakes. When the light is irradiated on the InS_{0.3}Se_{0.7}/WS₂ heterostructures, the photo-induced electron-hole will be separated at the interface. Electrons are driven to WS₂ side, while holes are driven to InS_{0.3}Se_{0.7} side. Finally, the electrons and holes were collected by the drain and the source, respectively, resulting in negative I_{sc} and positive V_{os} , as shown in Figure 2. In addition, the intensity of the photocurrent in the overlap area is almost the same, indicating the good uniformity of the as-prepared InS_{0.3}Se_{0.7}/WS₂ vdW heterostructure devices. Therefore, the whole photovoltaic current was originated from the heterojunction. This can eliminate the contingency of photoresponse and further determine the position of the photocurrent.

The transient photoresponse characteristics were evaluated employing a 532 nm constant wave laser with optical power of 3 mW , and a frequency-programmable chopper. The photocurrent waveform with a chopping frequency of 5 Hz is presented in Figure 4. When the laser is irradiated, the resolved transient photoresponse cycles are clearly observed. The transient photoresponse can be well reproduced in almost 100 cycles, indicating the good repeatable optoelectronic performance of the InS_{0.3}Se_{0.7}/WS₂ vdW heterostructure devices.

Figure 3. Photocurrent mapping of the $\text{InS}_{0.3}\text{Se}_{0.7}/\text{WS}_2$ vdW heterostructure device under 532 nm laser irradiation at zero bias potential. A photoresponse was observed in the heterostructure region. The blue dotted line indicates the reference line of the heterostructures center and the strongest part of the photocurrent.

Figure 4. The photocurrent waveform with a chopping frequency of 5 Hz.

3. Conclusions

The optoelectronic properties of $\text{InS}_{0.3}\text{Se}_{0.7}/\text{WS}_2$ vdW heterostructure devices was systemically investigated. The optoelectronic properties originate from the p - n junction formed at the $\text{InS}_{0.3}\text{Se}_{0.7}/\text{WS}_2$ vdW heterostructure. Furthermore, the response speed is enhanced at a small bias voltage. The uniformity and the repeatable photoresponse features at the interface make the $\text{InS}_{0.3}\text{Se}_{0.7}/\text{WS}_2$ vdW heterostructure device a promising candidate for the potential photodetector devices application. Our study indicates that the

InS_{0.3}Se_{0.7}/WS₂ vdW heterostructures are covered by graphene as electrodes are attractive building blocks in low-power photodetectors application, which could play an important role in the sustainable development of energy.

Supplementary Materials: The following are available online at <https://www.mdpi.com/article/10.3390/su13126883/s1>, Figure S1: Raman spectra of the InS_{0.3}Se_{0.7} flakes, Figure S2: Scanning electron microscopy (SEM) image and Energy-dispersive X-ray spectroscopy (EDS) of the as-synthesized InS_{0.3}Se_{0.7} crystals, Figure S3: Photoluminescence (PL) spectra of the InS_{0.3}Se_{0.7}/WS₂ heterostructures measured at indicated temperatures, Figure S4: Band diagram of the InS_{0.3}Se_{0.7}/WS₂ heterostructures.

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