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High-Performance Photodetector based on ReSe₂/PtSe₂ van der Waals Heterojunction

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1 ABSTRACT: In recent years, heterojunction photodetectors constructed of two-dimensional
2 materials with no overhanging bonds and lattice mismatches have attracted wide attention due to
3 the advantages of integrating different materials. Here we report a high-performance photodetector
4 with tunable dual auroral responses based on semiconductor (ReSe₂) and semimetal (PtSe₂)
5 heterojunction, with a high reverse commutation ratio of 6.2×10^4 at room temperature. Under
6 visible light irradiation, the photodetector with an open circuit voltage of 0.2 V and a short circuit
7 current of 19 pA, and a high on/off ratio of 5.5×10^4 , exhibits a clear photovoltaic effect. At the
8 same time, the photodetector shows the photoelectric responsivity and detectivity of 153 mA/W
9 and 7.72×10^{11} Jones, respectively. The photodetector also shows a large wideband optical
10 detection capability between ultraviolet and near infrared light. Our work provides a model to
11 develop future electronic and optoelectronic multifunctional devices based on 2D semiconductor
12 and semimetal van der Waals heterojunction.

1 ■ INTRODUCTION

2 Photodetector plays an important role in many scenes in rapid developed science and
3 technology. Because of widely applications in different fields such as medical imaging,¹ target
4 recognition and tracking,² environmental monitoring,³ chemistry,⁴ machine vision systems,⁵ and
5 robot,⁶ photodetector has aroused even more research interest. Traditional photodetectors are
6 usually based on group IV or III-V semiconductors (such as silicon, germanium, etc.). Due to the
7 limitations of these single materials, it is generally hard to meet the increasingly complex
8 application needs in reality. Since the discovery of graphene, more and more high-performance
9 photodetectors made of two-dimensional (2D) layered materials, with high responsivity and high
10 detectivity have been reported.⁷⁻¹⁹ For practical applications, a good photodetector should have
11 high signal-to-noise ratio, short response time, high responsivity, and low or no practical
12 application power dissipation.²⁰ Van der Waals heterostructures (vdWHs) made of 2D materials
13 provide an innovative approach to solve above problems.²¹ With no overhanging bonds and lattice
14 mismatches, vdW heterojunctions can be formed by various 2D materials through vertical
15 superposition, which provides an idea for the construction of vdWHs with unique properties
16 through infinite combination of different two-dimensional materials.²²⁻²⁷

17 Transition metal dichalcogenides (TMDs) exist in the form of MX_2 in the 2D layered
18 materials family. TMDs have the fascinating properties that the band structure varies with its
19 thickness, and the band gap adjusts with its number of layers, and thus attract much attention.²⁸
20 ReSe_2 nanosheets with a twisted triclinic structure with ambipolar transport properties are a
21 promising 2D material with outstanding optical and electrical properties.²⁹ As a typical
22 representative of Group 6 TMDs, ReSe_2 is a layered and highly anisotropic semiconductor with a
23 weak layer-dependent band gap between 1.24 eV and 1.09 eV.^{30, 31} Nevertheless, the wide

depletion barrier of electron-hole recombination at the junction interface is hindered,^{32, 33} and the performance of the photodetector based on 2D ReSe₂ is also limited due to the unbalanced concentrations between electrons and holes. How to solve this problem is undoubtedly a great challenge. Based on literatures, if we combine ambipolar ReSe₂ with semimetal 2D materials, such as PtSe₂, to form semiconductor-semimetal vdWH might solve this problem. As a representative member of the tenth group of TMDs, 2D semiconductor PtSe₂ has a band gap of 1.2 eV and 0.21 eV of monolayer and bilayer, respectively, but it shows semimetal behavior when the layer reaches three and above.³⁴⁻³⁸ At room temperature, the carrier mobility of PtSe₂ can theoretically be as high as 4000 cm² V⁻¹ s⁻¹.³⁹ This makes PtSe₂ an extremely promising channel material for ultra-thin, flexible electronic devices. Selective controlling as either a semiconductor or a semimetal demonstrates the potential of 2D ReSe₂ in integration as a circuit component in two-dimensional circuits, used as both electrode and semiconductor channel, facilitating the formation of uniform semimetal-semiconductor contacts. But up to now, the Van der Waals heterojunction photodetectors combined ReSe₂ with semimetal materials are rarely involved and lack in-depth development in literatures.

In this work, a gate-voltage-regulated heterojunction photodiode combined 2D semiconductor ReSe₂ with 2D semimetal PtSe₂ is demonstrated. The semimetal PtSe₂ has high electron concentration and high electron mobility, which can be used as carrier selective contact,⁴⁰ and the semiconductor ReSe₂ is used as photoactive material. The photodiodes achieved a broadband optical response from ultraviolet (UV) to near-infrared (NIR) regions at room temperature. ReSe₂/PtSe₂ heterojunction diodes exhibit gate-adjustable bidirectional diode behavior. Under the control of gate voltage, the current rectification ratio is more than 6.2×10^4 . The open-circuit voltage is as high as 0.2 V, and the short-circuit current is 19 pA, achieving the

high-light current to dark current ratio of 5.5×10^4 . At the same time, under the laser irradiation at 635 nm, the heterojunction has an excellent responsivity and a high detectivity of 153 mA/W and 7.72×10^{11} Jones, respectively without bias voltage. The EQE is up to 30%, indicating the efficient separation of photogenerated electron-hole pairs.

■ RESULTS AND DISCUSSION

Figs. 1a and b respectively show the schematic diagram of ReSe₂/PtSe₂ heterojunction structures superimposed on SiO₂/Si substrate and optical microscope image of the heterojunction device. The resulted ReSe₂/PtSe₂ vdWH device consists of mechanical stripping and polydimethylsiloxane assisted dry transfer (see the supporting information for the detailed manufacturing process of the device). The ReSe₂/PtSe₂ heterojunction is constructed by a few layers of ReSe₂ and PtSe₂ mechanically separated from the corresponding bulk material, as shown in Fig. 1a, performing form an exact transfer on the substrate via a micro nano transfer platform. As can be seen from the optical microscope images, Fig. 1b, a few layers of ReSe₂ film was superimposed on the PtSe₂ film with an overlapping area of about 17 μm^2 , and then Au/Pt electrodes deposited on the ReSe₂ and PtSe₂ films. As ReSe₂ has a wider band gap than PtSe₂, the ReSe₂ film is stacked on top of PtSe₂ film for more efficient light absorption. The SiO₂/Si substrate with a thickness of 300 nm acts as the doped back gate of electrons or holes in ReSe₂ and PtSe₂ films. Notably, the performance of the device can be easily tested by depositing electrodes in the area near the junction. The results of measuring the heterojunction region by atomic force microscopy (AFM) are shown in Fig. 1d. The thickness information of the ReSe₂ and PtSe₂ films is displayed in the linear scanning contour of AFM image (Fig S1), and from which one can see that ReSe₂ film is about 6.3 nm thick, and thus its number of the layers is approximately 9,⁴¹ while that of the PtSe₂ film is 5.9 nm and its number of layers is about 12.^{42, 43} Raman scattering

measurements were performed on the device to ensure heterogeneous structure and material formation, and the results are shown in Fig. 1c. In the thin ReSe₂ region, the Raman signals caused by the in-plane vibration (E_g) is located at $\approx 118/123\text{ cm}^{-1}$ and the Raman signals at $\approx 158/172\text{ cm}^{-1}$ and $\approx 284/294\text{ cm}^{-1}$ are caused by the out-of-plane vibration (A_g) (Fig. 1c, blue line). These results are consistent with previous report for ReSe₂ film.⁴⁴ In the PtSe₂ region, the peak corresponding to the E_g phonon mode and the A_{1g} mode were respectively observed at 175 cm^{-1} and 205 cm^{-1} (Fig. 1c, red line), and the inconspicuous feature at $\sim 230\text{ cm}^{-1}$ is attributed to an overlap between the E_u and A_{2u} modes, which are longitudinal optical (LO) modes of in-plane the and out-of-plane motions of platinum and selenium atoms, respectively. This is consistent with previous report for PtSe₂ film.⁴⁵ From Fig. 1c one can see all the Raman signals for both ReSe₂ and PtSe₂ appear on the heterojunction region (Fig. 1c, black line), and compared with the corresponding single material, there is no obvious peak shift, demonstrating that the two films forming the heterojunction region have high quality. These results also prove that the heterostructural region consists of the ReSe₂ and PtSe₂.

Subsequently, the electrical properties of individual ReSe₂ and PtSe₂ vdWH devices and ReSe₂/PtSe₂ vdW heterojunction devices are measured. Fig. 2a is a schematic diagram of a ReSe₂/PtSe₂ vdW heterojunction device used to measure electrical properties. As mentioned above, the ReSe₂ film with a larger band gap is placed on the top of PtSe₂ film for better photo absorption. In order to adjust the carrier density of each film, Si substrate acts as the back gate. The metal electrode deposited on the side of ReSe₂ and PtSe₂ are defined as the drain electrode and source electrode, respectively. In addition, two electrodes are attached to each ReSe₂ and PtSe₂ film, which makes it easier to study the properties of individual film of ReSe₂ and PtSe₂ and the ReSe₂/PtSe₂ heterojunction. The measured electrical characteristics of single ReSe₂ and PtSe₂

field-effect transistors (FETs) are shown in Figs. S2 and 3 in supporting information. The transfer curve of ReSe₂ FET exhibits that there is a large conduction current and ambipolar conduction under the control of gate voltage between -40 V and 40 V (Fig. S2c), where N-type conductance behavior is slightly stronger than P-type, which indicates that the Fermi level is close to the minimum conduction value of ReSe₂, and the electron density is higher than the hole density. The linear mode of I-V curve of the PtSe₂ FET (Fig. S3b) indicates that there is ohmic contact between PtSe₂ and the metal electrode, showing semimetal conductivity behavior of PtSe₂ film, which is consistent with previous report.⁴⁶ Fig. 2b shows the linear I_{ds} - V_{ds} curve of the ReSe₂/PtSe₂ vdWH device over the $V_{ds} = \pm 2$ V bias voltage range in a logarithmic scale. Extremely large reverse current and very low forward current can be observed, forming a clear reverse diode. The rectification ratio of the device can be obtained by calculating the ratio of reverse current to forward current. At room temperature, the rectifier ratio reached 4.7×10^2 without gate voltage. Fig. 2c shows the output characteristics of the ReSe₂/PtSe₂ vdWH device in the $V_g = \pm 30$ V gate voltage range. It can be found that the higher the gate voltage is, the better the rectifier performance of the device, regardless of whether the gate voltage is positive or negative. The highest rectifying ratio of 6.2×10^4 is achieved at $V_g = 30$ V, an increase of two orders of magnitude compared to that without gate voltage. Comparing the output characteristics of the ReSe₂/PtSe₂ vdWH device with that of the isolated ReSe₂ vdWH device, one can see that the ReSe₂ film determines the conductivity of the ReSe₂/PtSe₂ vdWH device. The I_{ds} - V_g curve of the ReSe₂/PtSe₂ vdWH devices were measured to study the conductivity behavior of diodes, as shown in Fig. 2d. The ambipolar FET behavior of ReSe₂/PtSe₂ vdWH device shows that P-type and N-type doping can be obtained under the control of gate voltage, and the heterojunction diode can switch between pure P-type diode and pure N-type diode extremely well. The integrated transmission mechanism of

1 ReSe₂/PtSe₂ vdWH will be explained in detail by describing the band arrangement of ReSe₂ and
2 PtSe₂ later.

3 The energy band information of ReSe₂ and PtSe₂ was obtained to study the transmission
4 mechanism so that the behavior of the back-gate regulated rectifier of ReSe₂/PtSe₂ vdWH reverse
5 diode will be further understood. First, the surface potential of ReSe₂ and PtSe₂ was measured by
6 Kelvin Probe force microscopy (KPFM), upon which to determine their Fermi level difference and
7 the result is shown in Fig. 3a. The KPFM measurement diagram is shown in Fig. S4. The KPFM
8 measurement was performed using the RH-coated KPFM tip ($\Phi_{tip} = 4.98$ eV) and the straddle-
9 alignment was estimated. The average ΔV_{CPD} values of ReSe₂ and PtSe₂ films were obtained at
10 190 mV and 160 mV, respectively. The ΔV_{CPD} is defined as the difference between the KPFM tips
11 and the work function of the sample. The functional values of ReSe₂ and PtSe₂ can be obtained by
12 the following formula

$$\Phi_s = \Phi_{tip} - \Delta V_{CPD} \quad (1)$$

14 where Φ_{tip} and Φ_s are the KPFM tips and work function of the sample (ReSe₂ and PtSe₂),
15 respectively. Thus, the work function of ReSe₂ and PtSe₂ films can be estimated, and the results
16 are about 4.79 eV and 4.82 eV, respectively (Fig. 3b), and the difference of Fermi level between
17 ReSe₂ and PtSe₂ is about 30 meV (0.03 eV). Figs. 3b and c show the band distribution before
18 ReSe₂ and PtSe₂ contact and the band arrangement after contact to return to the equilibrium state.
19 According to the KPFM measurement results, the electrons of ReSe₂ will drift into PtSe₂ and reach
20 the equilibrium state when ReSe₂ and PtSe₂ are stacked vertically together. At this time, the
21 electron density in ReSe₂ decreases, resulting in the formation of an obvious band bending electron
22 depletion region on the side of ReSe₂, while the electrons in PtSe₂ will increase. This also means

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1 that an internal electric field from ReSe₂ to PtSe₂ will be formed at the ReSe₂/PtSe₂ vdWH
2 interface. Figure 3d-g shows the energy band arrangement of ReSe₂/PtSe₂ vdWH device under
3 different conditions. For the obvious rectification behavior, ReSe₂ is modulated to heavy N type
4 with the rise of Fermi level when the gate voltage is positive. Fig. 3d shows that, holes flow from
5 ReSe₂ to PtSe₂ under positive bias voltage. As a result of the positive gate voltage, the holes in
6 ReSe₂ are a few charge carriers, so a minor current is generated. Fig. 3e shows that when the bias
7 voltage is negative, the internal electric field is weakened, the potential barrier in the heterojunction
8 region is reduced, and electrons are transferred from the high-level ReSe₂ to the low-level PtSe₂,
9 thus generating a larger current. According to these results, it is clear to find the minor forward
10 current and large reverse current in the I-V characteristic curve of the back gate control of
11 ReSe₂/PtSe₂ vdWH device. This leads to heavily doped reverse diode with rectifier ratio of more
12 than 10⁴. As the positive V_g increases, the Fermi level of ReSe₂ gradually rises, which leads to
13 more bending of the energy band, resulting in an increase in both forward and reverse current. The
14 reverse current increases by a larger margin due to the positive gate voltage, so the rectification
15 ratio increases with increasing gate voltage. ReSe₂ is modulated to heavy P type conductive
16 behavior with the decrease of Fermi level due to the negative gate voltage. Fig. 3f shows that when
17 the bias voltage is positive, the internal electric field is weakened, the potential barrier in the
18 junction area is lowered, and holes flow from ReSe₂ to PtSe₂, thus generating a large current. As
19 shown in Fig. 3g, under negative bias, electrons are transferred from high energy ReSe₂ to low
20 energy PtSe₂. As a result of the negative gate voltage, the electrons in ReSe₂ are minority carriers,
21 so a small current is generated. With the increase of negative V_g, the Fermi level of ReSe₂ is
22 reduced, which leads to more bending of the energy band. However, the density of minority
23 carriers in ReSe₂ gradually decreases, so the reverse current decreases and the forward current

1 increases. According to the above results, it is clear to find the small reverse current and large
2 forward current in the back gate related I-V curve of ReSe₂/PtSe₂ vdWH device.

3 Next, in order to the explore its potential applications, the photoelectric characteristics of the
4 ReSe₂/PtSe₂ vdWH diode are systematically studied. Fig. 4a shows the photodetector irradiated by
5 a 635 nm laser through schematic diagram. It is worth noting that all the photoelectric
6 characteristics are measured without applying the gate voltage ($V_g = 0$ V), unless specially
7 emphasized. The photoresponse range of the ReSe₂/PtSe₂ vdWH device was tested by irradiating
8 the heterostructure with the same incident laser power of 3.6 mW with lasers with the wavelength
9 of 405, 635, 780 and 940 nm. The I-V curves of the heterojunction in the dark are shown in Fig.
10 4d, and an obvious wavelength dependence of the photoresponse can be observed. For the tested
11 four wavelengths of laser, the best photoreponse of the ReSe₂/PtSe₂ vdWH device is under the
12 irradiation of $\lambda = 635$ nm laser. Thus, the 635 nm laser was selected for the subsequent tests. The
13 photoreponse curves of the ReSe₂/PtSe₂ vdWH photodiode under 635 nm laser irradiation with
14 different laser power densities and dark state when $V_g = 0$ V was shown in Fig. 4b. The
15 photoresponse characteristics of the device to laser irradiation of the other three wavelengths are
16 shown in Fig. S5. Under laser irradiation, no matter in the positive bias region or negative bias
17 region, a significant increase in photocurrent can be observed, and positive bias region shows a
18 higher sensitivity to laser power. The light on/off ratio of the device is about 10^3 at zero bias
19 voltage when the laser power density is 7.29 mW/cm². The laser irradiation passes from top ReSe₂
20 film to bottom PtSe₂ film. According to the photoelectric performance of the PtSe₂ film (Fig. S3,
21 supporting information), the difference between light and dark currents is negligible. It can be
22 inferred that only the channels in the top ReSe₂ film produce electron-hole pairs. The I-V
23 characteristics of the ReSe₂/PtSe₂ vdWH device in the small range bias voltage under laser

1 irradiation with different laser power densities are shown in Fig. 4c. It is clear to found that short-
2 circuit current is positive and open-circuit voltage is negative, and under the influence of the
3 increased laser power density, the short-circuit current increases. For high-performance
4 photodetectors, the response speed is an extremely important parameter. The I-T curves of the
5 photodetector under 635 nm laser irradiation with different laser power densities without bias
6 voltage is shown in Fig. S6a, from which one can see under the laser irradiation of periodic switch,
7 the photocurrent can be stabilized in a short time. The one-period I-T curve extracted from Fig.
8 S6a is shown in Fig. S6b. The response speed can be measured by the rise time (defined as the
9 time required for the photocurrent to rise from 10% to 90%) or the decay time (the time required
10 for the photocurrent to fall from 90% to 10%). From Fig. S6b, one can see obtained that the rise
11 time of the $\text{ReSe}_2/\text{PtSe}_2$ vdWH device is about 0.8 s and the decay time is about 0.7 s. A great
12 improvement in response speed is achieved for the $\text{ReSe}_2/\text{PtSe}_2$ vdWH device as compared with
13 that for the single ReSe_2 device⁴¹. The photoresponse at $V_g = -40$ V was measured so that the
14 influence of the gate voltage on the photoelectric performance of the $\text{ReSe}_2/\text{PtSe}_2$ vdWH devices
15 can be further explored. As Fig. 4e shown, the logarithmic form of output characteristic curves of
16 the $\text{ReSe}_2/\text{PtSe}_2$ vdWH device under 635 nm laser irradiation with different laser power densities
17 and at dark state when -40 V gate voltage is applied. Different from $V_g = 0$ V, when -40 V gated
18 voltage is applied, the increase of photocurrent is mainly shown in the region where $V_{ds} < 0$ V,
19 and a high on/off ratio of more than 5.5×10^4 at negative bias is achieved. The I-V characteristics
20 of the $\text{ReSe}_2/\text{PtSe}_2$ vdWH device in the small range bias voltage under laser irradiation with
21 different laser power densities when -40 V gate voltage is applied are shown in Fig. 4f. Under the
22 influence of -40 V gate voltage, the conductive behavior of ReSe_2 changes to heavy P-type with
23 the decrease of Fermi level. At this time, due to the internal electric field, the electrons drift to the

PtSe₂ side and enter the source electrode, while the holes drift to the ReSe₂ side and enter the drain electrode. Therefore, negative I_{sc} and positive V_{oc} can be found in Fig. 4f. When the laser power density is 7.29 mW/cm², the short circuit current of 19 pA and open circuit voltage of 0.2 V are obtained.

The net photocurrent I_{ph} under different laser power densities was tested when $V_g = 0V$ and $V_g = -40 V$ to explore the mechanism of photocurrent generation, and the results are shown in Figs. 5a and d, respectively.

$$I_{ph} = I_{ds} - I_{dark} \quad (2)$$

where I_{ds} and I_{dark} is the current of the device under light and dark state, respectively. In Figs 5a and d, the black point is the data measured in the experiment, and the red line is the curve obtained by fitting the power-law formula

$$I_{ph} = \alpha P^\theta \quad (3)$$

In the formula α is a constant, P is the laser power density of the incident laser, and θ is the power-law index. The value of θ calculated according to the fitting results is about 0.47 and 0.85 with $V_g = 0 V$ and $-40 V$, respectively. The photocurrent is dictated by the photogating effect referring to trap states, and by the photoconductive effect, respectively, when $\theta = 0$ and $\theta = 1$.⁴⁷ Above results indicate that these the photocurrent of the ReSe₂/PtSe₂ vdWH device is determined by these two effects. The photoelectric mechanism of the ReSe₂/PtSe₂ vdWH device can be understood through the energy band diagram shown in Figs. 5c and f. Without gate voltage, the top ReSe₂ film mainly produces photogenerated electron hole pairs. Fig. 4c shows that due to large built-in electric field in the single-side consumption area of ReSe₂, the electrons drift to the ReSe₂ side and are gathered by the drain electrode, while the holes drift to the PtSe₂ side and are gathered by the source

electrode, thus generating positive I_{sc} and negative V_{oc} . Generally when a positive bias voltage is applied, electrons flow from PtSe₂ to ReSe₂. However, according to the photoelectric performance of the PtSe₂, it can be known that the PtSe₂ film is not sensitive to laser irradiation, and the photogenerated electron hole pairs are almost negligible. Therefore, in the positive bias area of Fig. 4e, it can be observed that the photoelectric current generated by the ReSe₂/PtSe₂ vdWH device under laser irradiation is not much higher than the dark current generated in the dark state. When a negative bias voltage is applied, ReSe₂ is excited to generate electron hole pairs, and the electrons flow to PtSe₂, thus generating a larger photocurrent compared to the dark state, as shown in Fig. 5f. Responsivity (R) and detectivity (D^*) play an important role in the quantitative evaluation of high-performance photodetectors, and can be expressed as

$$R = \frac{I_{ph}}{P_{in}} \cdot A \quad (4)$$

$$D^* = \frac{R \cdot \sqrt{A}}{\sqrt{2e \cdot I_{dark}}} \quad (5)$$

where I_{ph} is the net photo current described above, A is the effective illumination area of the photodetector, P_{in} is the power density of the irradiated laser, and e is the unit charge. Figs. 5b and e show the functions of R and D^* to laser power densities when the gate voltage is 0 V and -40 V, respectively, without bias voltage under 635 nm laser irradiation. The increase of laser power density leads to the decrease of R and D^* , indicating that trap states capture the photogenerated electrons and reduce the possibility of photogenerated electrons recombination. From these two figures one can obtained that the maximum R is about 153 mA/W and the maximum D^* is about 7.72×10^{11} Jones, which are better than the value in some previous reported literatures (Table S1), demonstrating excellent performance of the ReSe₂/PtSe₂ vdWH device. External quantum

1 efficiency (EQE) and power conversion efficiency (PCE) are also very important parameters. PCE
2 can be obtained by the formula

$$3 \quad \text{PCE} = \frac{P_{\max}}{P_{\text{in}}} \quad (6)$$

4 where P_{\max} is the maximum electrical power generated by the photodetector under laser irradiation
5 and P_{in} is the effective power of the laser irradiation.⁴⁸ The EQE can be interpreted as the ratio of
6 the number of photons emitted in the plane per unit time to the electron-hole pairs injected in the
7 plane per unit time, which can be obtained by formula

$$8 \quad \text{EQE} = \frac{Rhc}{e\lambda} \quad (7)$$

9 where h is the Planck constant, the value of c is the speed of light and λ is the wavelength of the
10 laser.²⁶ From the calculated results of the PCE and EQE of the ReSe₂/PtSe₂ vdWH device when
11 the gate voltage is 0 V and -40 V under 635 nm laser irradiation without bias voltage shown in
12 Fig. S7, one can see the maximum values of PCE and EQE were $1.5 \times 10^{-3}\%$ and 30%, respectively,
13 indicating efficient separation of photogenerated electron-hole pairs in the ReSe₂/PtSe₂ vdWH
14 device.

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■ CONCLUSIONS

In summary, for the first time a high-performance photodetector based on 2D semiconductor ReSe₂ and 2D semimetal PtSe₂ van der Waals heterojunction was proposed and successfully constructed. The ReSe₂ serves as photoactive material, while PtSe₂ serves as carriers selective contact. The polarity of the photodetector can be adjusted by the gate voltage V_g . A high reverse rectifier ratio of 6.2×10^4 can be realized at room temperature. The short-circuit current and open-circuit voltage are 19 pA and 0.2 V, respectively, and the light on/off ratio is over 5.5×10^4 under proper condition. The photodetector has a responsivity of 153 mA/W and a detectivity of 7.72×10^{11} Jones. The photodetector also shows a wideband optical detection from ultraviolet to near infrared light. Our results demonstrated that 2D semiconductors and 2D semimetals have great potential in the development of high-performance photodetectors.

■ EXPERIMENTAL SECTION

Device Fabrication. The method of constructing ReSe₂/PtSe₂ vdWH device was the dry transfer method with PDMS as the carrier and the mechanical stripping method. First, the corresponding films were prepared from the bulk of ReSe₂ and PtSe₂ by mechanical stripping with adhesive tape, and then the ReSe₂ and PtSe₂ films were transferred to the Si substrate covered with 300 nm SiO₂ using PDMS as the carrier. After that, the electrode pattern is drawn by electron beam lithography (EBL, eLINE Plus, Raith), and then Ti (thickness of 15 nm) and Au (thickness of 60 nm) are deposited on the substrate by electron beam evaporation and thermal evaporation. Finally, the electroplating device is processed in acetone under the standard stripping process.

Device Characterization and Testing. The optical microscope (WMJ-9688) was used to study ReSe₂/PtSe₂ vdWH device. The Raman spectrum were measured using a laser source with a wavelength of 532 nm and a Raman/photoluminescence (PL) system (FST2-Ahdx-DZ) equipped with a monochromator. The film thickness and surface potential of the ReSe₂ and PtSe₂ films were measured by KPFM (Hitachi AFM5500M). The electrical performance of ReSe₂ and PtSe₂ vdWH devices was measured by using a semiconductor device parameter analyzer (Keithley 4200A-SCS) in the detection station (HFS600E-PB4). In the process of optical response measurement, 635 nm laser was always irradiated on the effective area of the device. The above measurement results were obtained at room temperature.

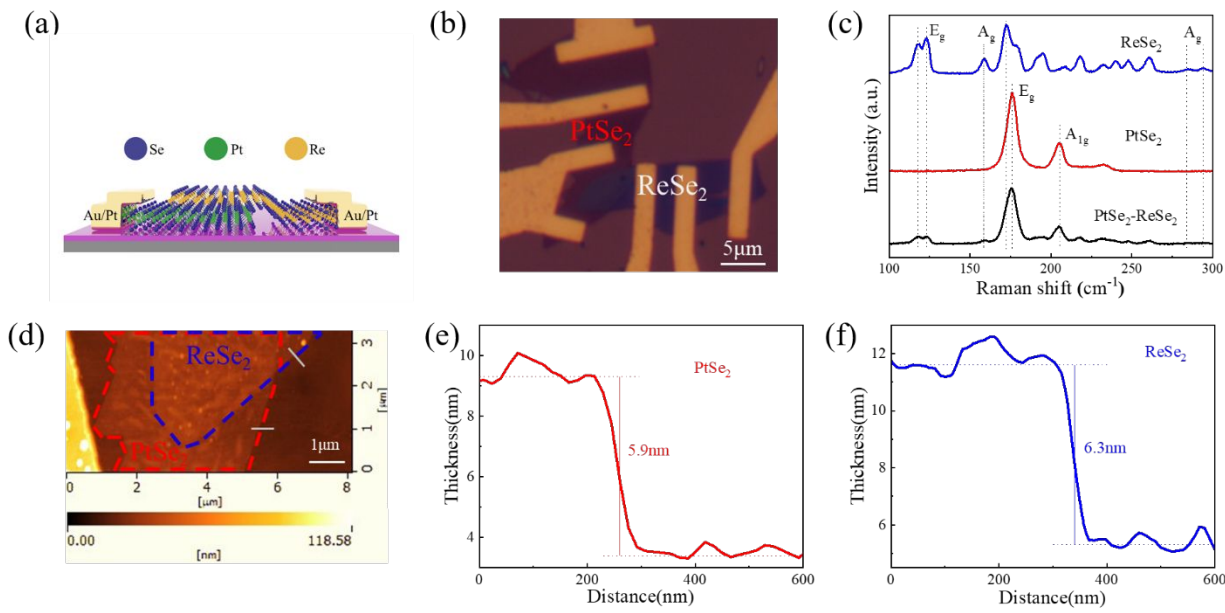


Figure 1. Optical characterizations and AFM measurements of the ReSe₂/PtSe₂ vdWH heterojunction. (a) Schematic diagram of ReSe₂/PtSe₂ heterostructures superimposed on SiO₂/Si substrates, (b) optical microscope image of the heterojunction. (c) Raman spectra of ReSe₂ and PtSe₂ films, and ReSe₂/PtSe₂ vdWH heterojunction. (d) AFM measurement results of ReSe₂/PtSe₂ vdWH. Thickness information of ReSe₂ film (e) and PtSe₂ film (f).

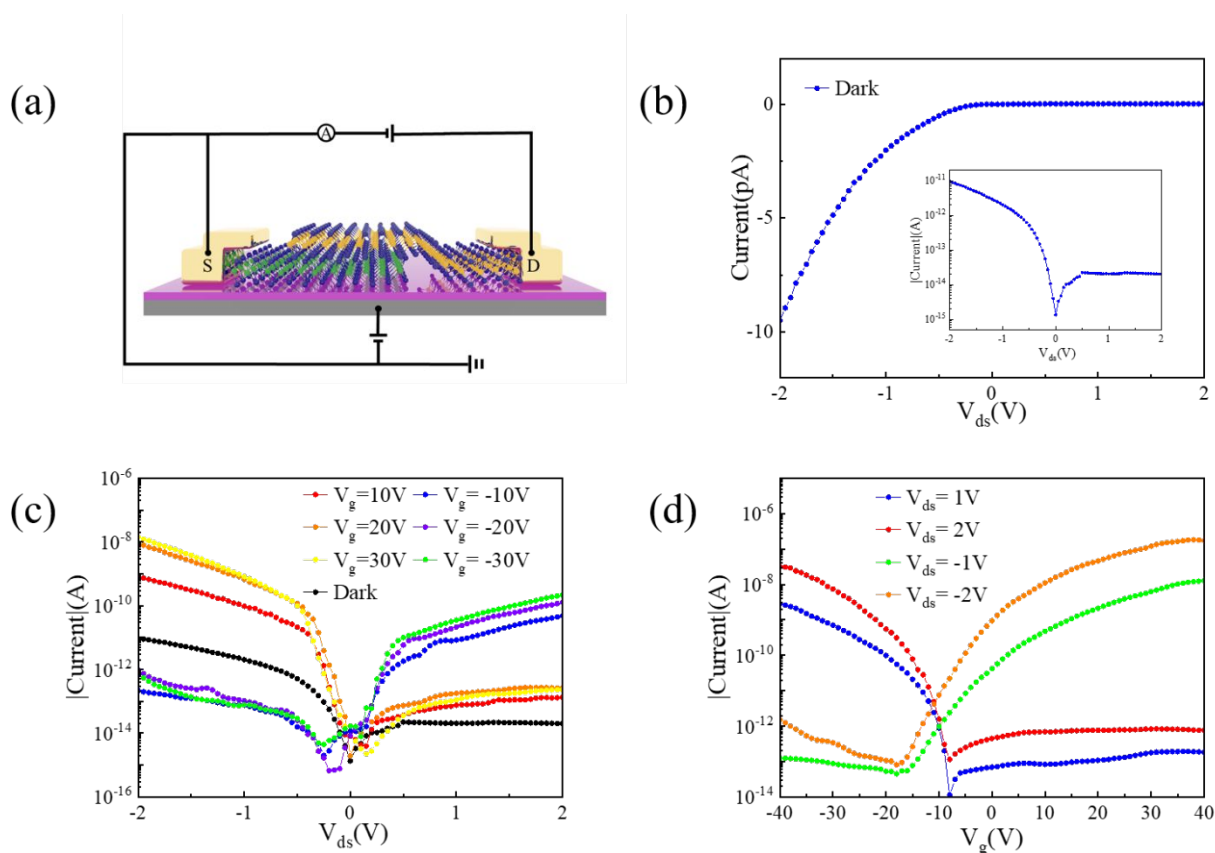


Figure 2. Electrical properties of the ReSe₂/PtSe₂ vdWH device. (a) Schematic diagram of a ReSe₂/PtSe₂ vdWH device used to measure electrical properties. (b) Linear I_{ds} - V_{ds} characteristics of ReSe₂/PtSe₂ vdWH device in the range $V_{ds} = \pm 2$ V. The I_{ds} - V_{ds} characteristics is on logarithmic scale. (c) Output characteristics of ReSe₂/PtSe₂ vdWH devices on logarithmic scales in the $V_g = \pm 30$ V range. (d) I_{ds} - V_g characteristics of ReSe₂/PtSe₂ vdWH device at different bias voltages.

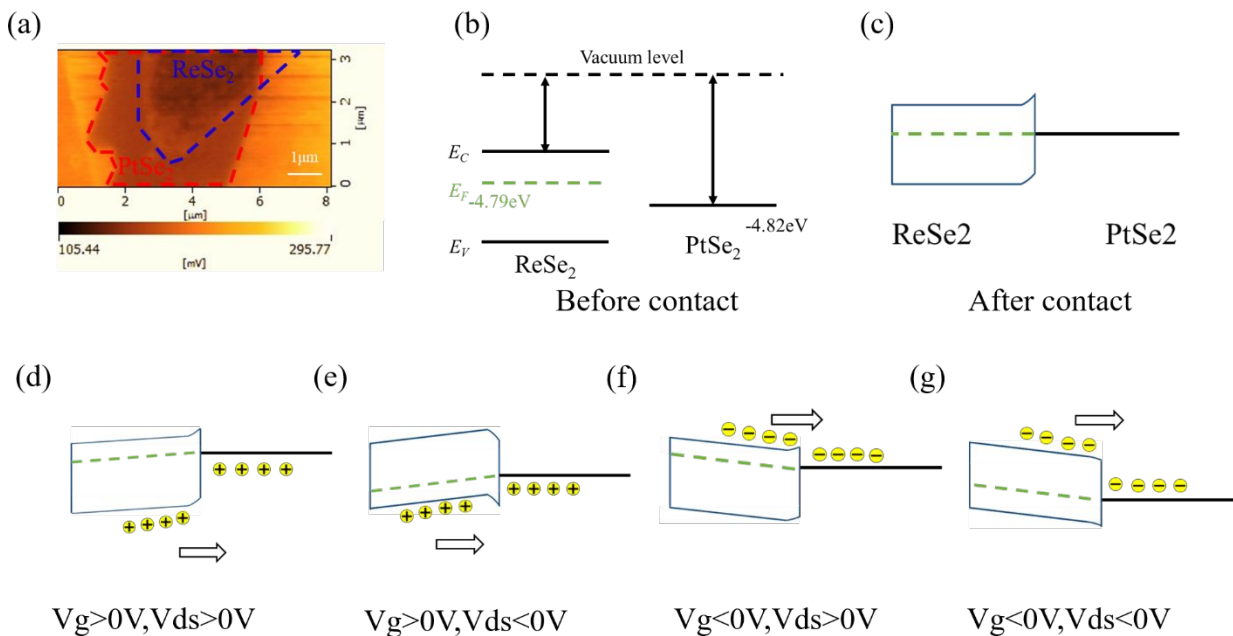


Figure 3. KPFM characterization and schematic energy band diagrams of the ReSe₂/PtSe₂ vdWH device. (a) KPFM image of the ReSe₂/PtSe₂ vdWH. (b) (c) Band structure of ReSe₂/PtSe₂ vdWH before and after contact. (d)-(g) Energy band diagrams of the ReSe₂/PtSe₂ vdWH device under different bias voltages V_{ds} and gate voltages V_g .

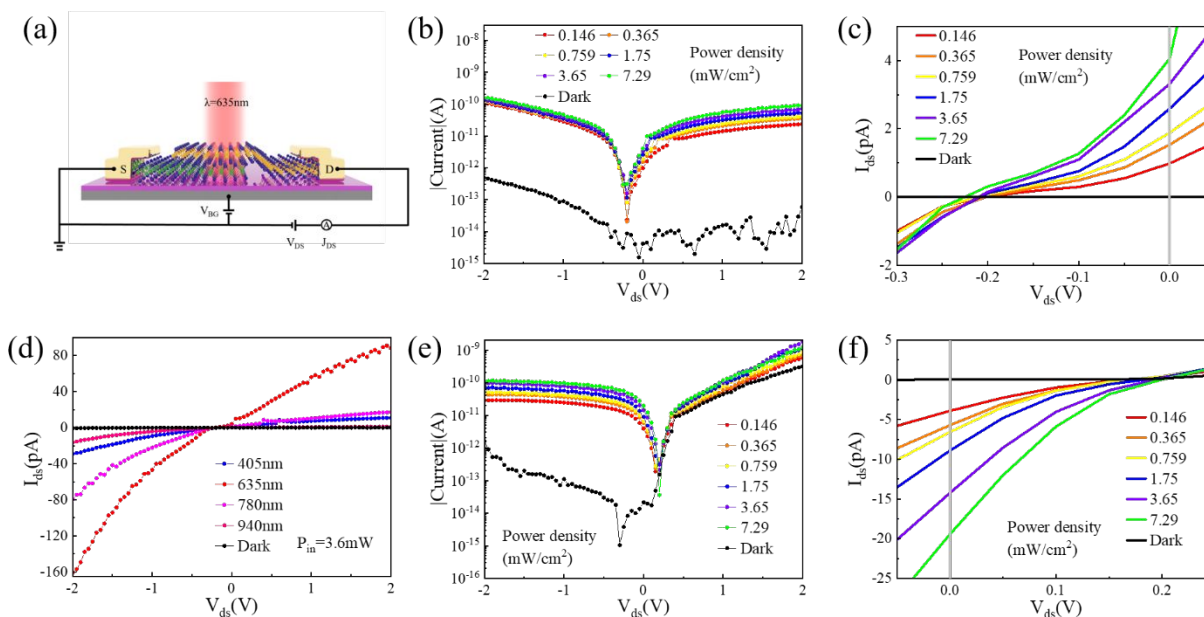


Figure 4. Photoresponse characteristics of the ReSe₂/PtSe₂ vdWH device. (a) Schematic diagram of a heterojunction irradiated by a 635 nm laser. (b) (c) When $V_g = 0$ V, I_{ds} - V_{ds} curve of the device under 635 nm laser irradiation with different power densities and I_{ds} - V_{ds} curve under small bias voltage. (d) I_{ds} - V_{ds} characteristics of the ReSe₂/PtSe₂ vdWH device measured in darkness and under illumination at different wavelengths with 3.6 mW incident laser power. (e) and (f) When $V_g = -40$ V, I_{ds} - V_{ds} curve of the device under 635 nm laser irradiation with different power densities and the I_{ds} - V_{ds} curve under small bias voltage.

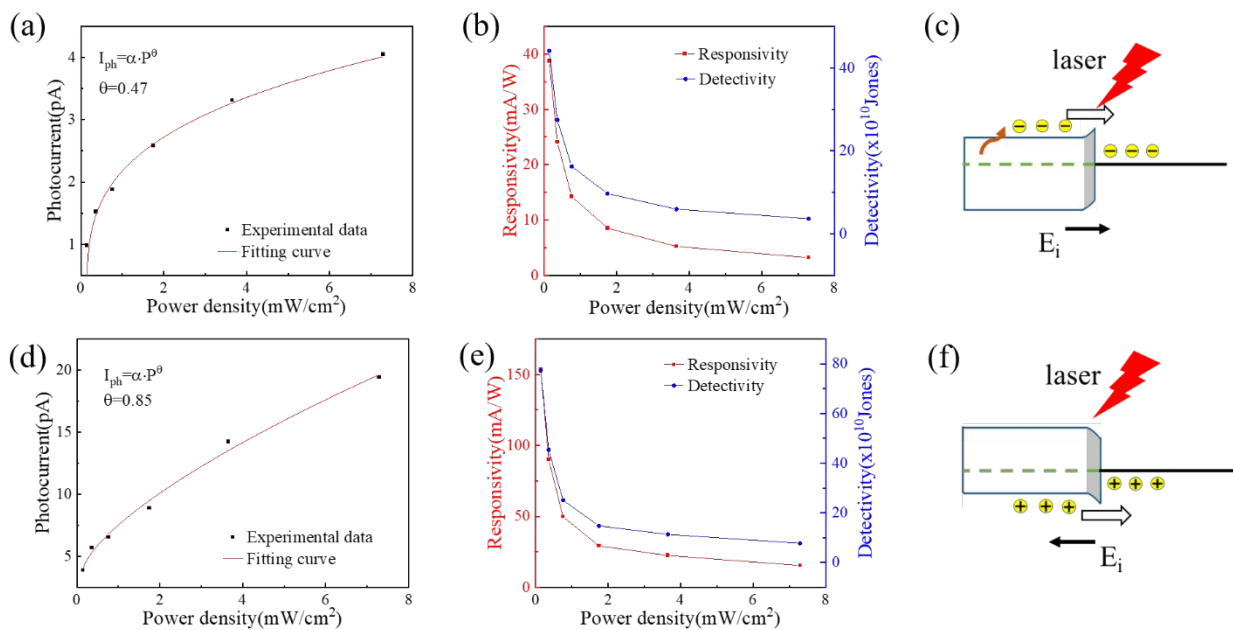


Figure 5. Input light power density dependence of the photocurrent I_{ph} of the ReSe₂/PtSe₂ vdWH device under incident laser ($\lambda = 635$ nm) (black dots), the red lines show the best-fitted results at (a) $V_g = 0$ V and (d) $V_g = -40$ V. Laser power density ($\lambda = 635$ nm) dependent responsivity and detectivity at (b) $V_g = 0$ V and (e) $V_g = -40$ V. Photogenerated carrier band diagram of the heterojunction without bias voltage at (c) $V_g = 0$ V and (d) $V_g = -40$ V

1 ASSOCIATED CONTENT

2 Supporting Information

3 The detailed experimental process is shown in the supporting information.

4 Experimental section

5 **Figure S1.** AFM test results of ReSe₂ and PtSe₂.

6 **Figure S2.** Electrical properties of a single ReSe₂ FET.

7 **Figure S3.** Electrical properties of a single PtSe₂ FET.

8 **Figure S4.** Schematic of the KPFM measurement.

9 **Figure S5.** Photoresponse characteristics of ReSe₂/PtSe₂ vdWH devices.

10 **Table S1.** Comprehensive comparison of the critical parameters of fabricated ReSe₂/PtSe₂
11 vdWHs with previously reported ReSe₂ based photodetectors.

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16 Author Contributions

17 L.L. and G.H.L. supervised the project. Y.S and L.L. proposed the project. Y.S, L.L., and J.W.C.
18 designed the experiments. F.X.Z. prepared bulk materials. Z.Y.D exfoliated 2D materials and

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1 stacked heterostructures. Under supervision of Y.C.D, Q.G.Q. made electrode patterns using EBL.
2 Q.G.Q. made electrodes by metal evaporation coating. Under supervision of Y.S., X.F.M.
3 performed Raman spectra measurement. Under supervision of M.L. performed AFM/KPFM
4 measurement. Y.S. performed the electrical characterization supervised by J.W.C.. L.L., J.W.C.,
5 Y.S., M.L., Z.Y.D. and Y.C.D. analyzed the results. Y.S and Z.Y.D. drew schematic diagrams. All
6 authors discussed the results and provided constructive comments on the manuscript.

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14 Notes

15 The authors declare no competing financial interest.

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