Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Electronic Supplementary Information

High-performance, self-powered flexible MoS_2 photodetectors with asymmetric van der Waals gaps

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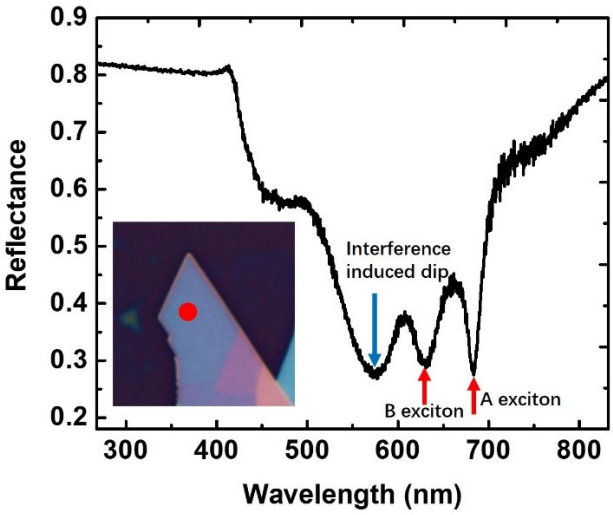

Fig. S1. The reflectance spectrum of a MoS_2 flake on SiO_2(300 nm)/Si substrate obtained from the sample highlighted by a red dot whose optical image is inset in this figure. The thickness of the measured MoS_2 flake is ~70 nm as obtained from the AFM.

Due to the small size of the mechanically exfoliated sample (~micrometer) as well as the untransparent supported SiO_2(300 nm)/Si substrate, the reflectance measurement is carried out to investigate the absorption properties of the MoS_2 flakes in the UV-visible light range. The reflection spectrum is collected by using a confocal WITEC alpha 300 spectrometer with LED white light source Kohler lighting. A 100× objective lens (NA = 0.9) with a spot size of 0.72 μm was used in the reflection experiments. Fig. S1 shows the reflectance spectrum of MoS_2 flake on SiO_2(300 nm)/Si substrate, the thickness of the measured MoS_2 flake is ~70 nm as obtained from atomic force microscopy (AFM). The reflectance spectrum is obtained by measuring reflection from the surface of MoS_2 and further divided by the white light used. In the UV-visible light range of 300 nm to 800 nm, broad reflection dips are observed. The first reflection dip is located at 576.1 nm, which is originated from the interference induced reflection dip. Here, SiO_2 works as the Fabry-Perot interference cavity. The second and the third reflection dips located at 630.0 nm and 681.7 nm are originated from B exciton and A exciton absorptions, individually. A and B exciton absorptions are originated from transitions from two spin-orbit coupling induced split valence bands to the conduction band, individually.[1] Thus, the 450 nm light has enough energy to excite electrons from the valence band to the conduction band.

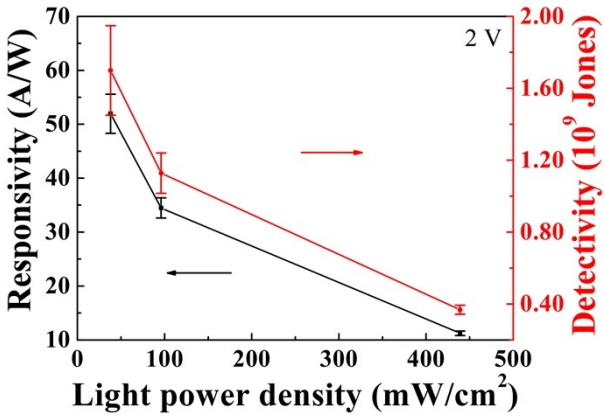

Fig. S2. Responsivity and detectivity of the MoS_2 photodetector (Device 1) under the illumination of 450 nm light with power densities of 38 mW/cm^2, 96 mW/cm^2, and 439 mW/cm^2 at a bias of 2 V.

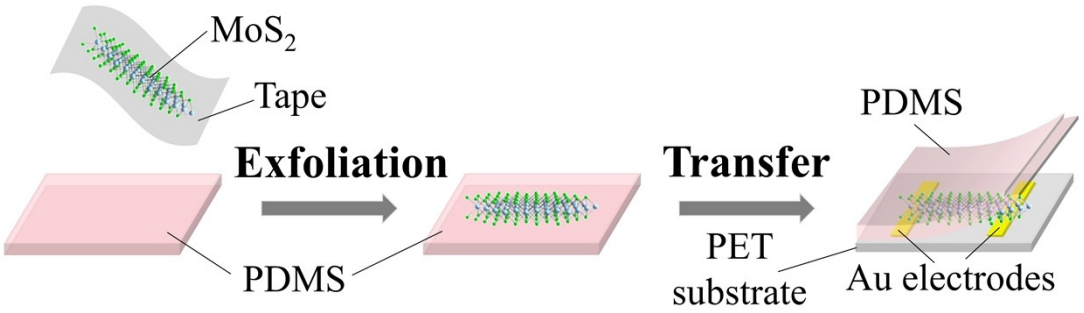

Fig. S3. Fabrication process flow for making MoS_2 photodetectors by the dry transfer method.

The dry transfer method is adopted to make another MoS_2 photodetector to explore whether the transfer method influences the self-powered detection ability. Fig. S3 shows the dry transfer fabrication process. MoS_2 flakes are peeled off from bulk crystal by scotch tape. The tape with MoS_2 flakes is brought into contact with the polydimethylsiloxane (PDMS) film and removed from the PDMS film. Some MoS_2 flakes are transferred onto the PDMS film via van der Waals force. The MoS_2 flake suitable for the fabrication of photodetector is found by the optical microscope. Then, this flake is transferred onto Au electrodes by the position-controllable movement of the PDMS film. Finally, the PDMS film is stripped from the PET substrate, the flake falls onto Au electrodes due to its weak adhesion with the PDMS film, and some typical MoS_2 photodetectors can be obtained by this dry transfer method. This method does not use any solution to avoid its influence on the performance of MoS_2 photodetectors.

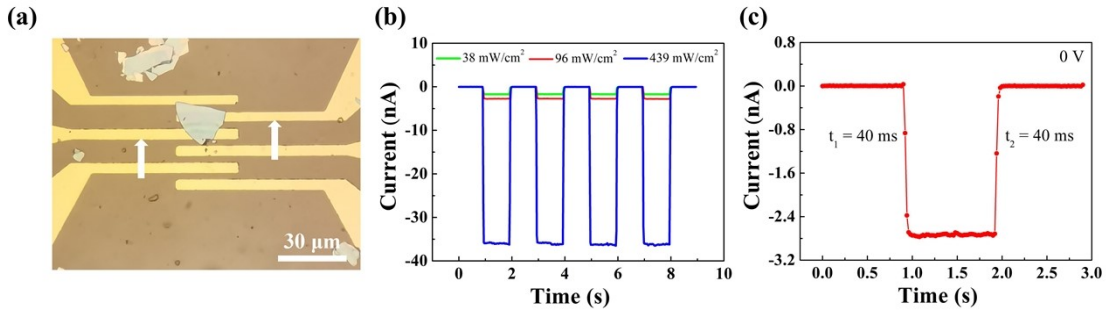

Fig. S4. (a) OM image of the MoS_2 photodetector (Device 2) fabricated by the dry transfer method. (b) The responses of the MoS_2 photodetector to the 450 nm light with different light power densities at 0 V. (c) Response time and recovery time of the MoS_2 photodetector.

Fig. S4a demonstrates the OM image of another typical MoS_2 photodetector (Device 2) fabricated by the dry transfer method. Two Au electrodes marked by white arrows are connected to the source meter to measure the performance of Device 2. Fig. S4b presents the responses of Device 2 to the on-off 450 nm light with a power density of 38 mW/cm^2, 96 mW/cm^2, and 439 mW/cm^2 at 0 V. It can be seen that Device 2 can also detect the 450 nm light repeatedly and stably under the self-powered mode. Under the illumination of the 439 mW/cm^2 450 nm light, the photocurrent of Device 2 is ~36 nA, which is on the same order of magnitude as that of Device 1. The response time and recovery time of Device 2 are both 40 ms (Fig. S4c), which are the same as those of Device 1. It can be concluded that the transfer process has no significant influence on the self-powered detection ability.

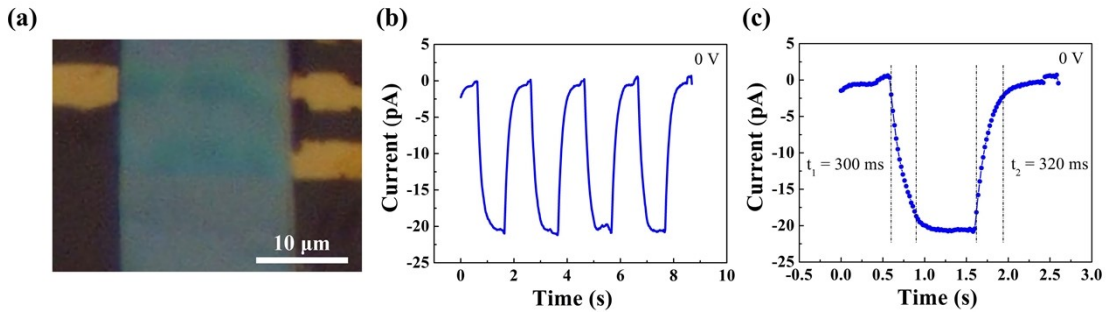

Fig. S5. (a) OM image of another MoS_2 photodetector (Device 3). (b) The response of Device 3 to the 450 nm light with a power density of 685 μW/cm^2 at 0 V. (c) Response time and recovery time of Device 3 to the 450 nm light with a power density of 685 μW/cm^2 at 0 V.

Another MoS_2 photodetector (Device 3) is also fabricated by the dry transfer method, as shown in Fig. S5a. It is worth noting that the MoS_2 flake has uniform thickness and the same contact area with both electrodes. Fig. S5b illustrates that Device 3 can also detect the 450 nm light repeatedly and stably at 0 V, indicating the self-powered detection ability does not result from the uneven thickness of the MoS_2 flake and the contact area difference between the MoS_2 flake and two Au electrodes. The response time and recovery time of Device 3 to the 450 nm light with a power density of 685 μW/cm^2 at 0 V are 300 ms and 320 ms, respectively (Fig. S5c). It can be concluded that the response speed of the photodetector slows down with the decrease of incident light power density, which is also attributed to the trapping effect.

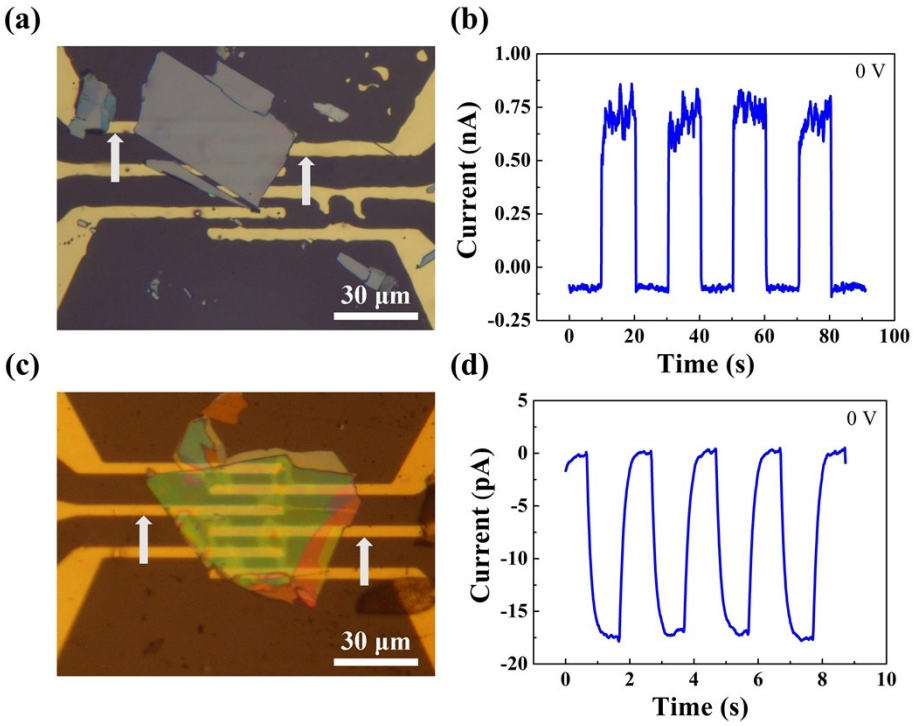

Fig. S6. (a) OM image of a typical ReS_2 photodetector (Device 4). (b) The response of the ReS_2 photodetector to the 450 nm light at 0 V. (c) OM image of a typical GaSe photodetector (Device 5). (d) The response of the GaSe photodetector to the 450 nm light at 0 V.

The method for the fabrication of the self-powered flexible MoS_2 photodetector also is appropriate to fabricate self-powered flexible photodetectors based on other two-dimensional materials, such as ReS_2 and GaSe, as shown in Fig. S6. The ReS_2 photodetector and the GaSe photodetector also can detect the 450 nm light repeatedly and stably under the self-powered mode.

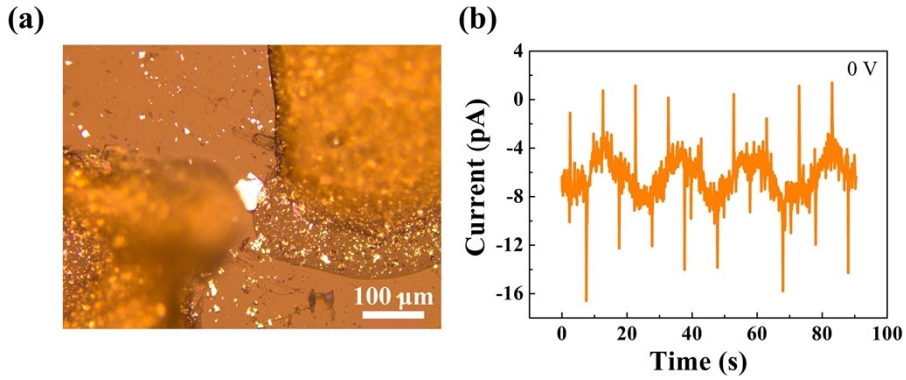

Fig. S7. (a) OM image of a typical MoS_2 photodetector (Device 6). The contacts between the MoS_2 flake with Au electrodes are a combination of surface contact and edge contact. (b) The response of Device 6 to the 450 nm light (38 mW/cm^2) with an on-off period of 10 s at 0 V.

For Devices 1-5, Au electrodes are in contact only with the surface of 2D materials. This kind of contact between electrodes and 2D materials is defined as surface contact. Devices 1-5 can detect 450 nm light repeatedly and stably at 0 V, indicating that they can operate as self-powered photodetectors. The optoelectronic devices based on 2D materials are significantly affected by the electrical contacts.[2] To explore the influence of contact on the self-powered detection ability, a typical MoS_2 photodetector (Device 6) with a combination of surface contact and edge contact is fabricated, as shown in Fig. S7a. Two metal electrodes are in contact with the surface and edge of a MoS_2 flake. Edge contacts can lead to a shorter bonding distance with stronger hybridization than surface contacts. The edge contact can suppress the influence of vdW gaps on the electrical property.[2] Fig. S7b shows the response of Device 6 to 450 nm light with an on-off period of 10 s at 0 V. Device 6 does not have a significant response to 450 nm light, indicating that the driving force of the self-powered photodetectors in this work originates from surface contact.

References

1. K. P. Dhakal, D. L. Duong, J. Lee, H. Nam, M. Kim, M. Kan, Y. H. Lee and J. Kim, *Nanoscale*, **2014**, *6*, 13028-13035.

2. A. Allain, J. Kang, K. Banerjee and A. Kis, *Nat. Mater.*, **2015,** *14*, 1195-1205.