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Integrated high-performance infrared phototransistor arrays composed of nonlayered PbS-MoS₂ heterostructures with edge contacts

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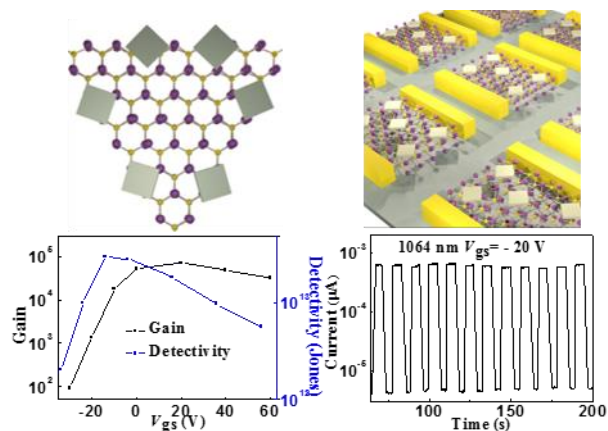
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ABSTRACT: Molybdenum disulfide (MoS₂) has attracted a great deal of attention in optoelectronic applications due to its high mobility, low off-state current and high on/off ratio. However, its intrinsic large bandgap limits its application in infrared detection. Here, we have developed a high-performance infrared photodetector by integrating non-layered PbS and layered MoS₂ nanostructures via van der Waals epitaxy. Density functional theory (DFT) calculations indicate that PbS nanoplates are in contact with MoS₂ edges through strong chemical hybridization, which is expected to offer a fast transmission path for carriers that

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3 enhances the response speed. The phototransistor exhibits a fast response ($\tau_{\text{rising}} = \tau_{\text{decay}} = 7.8$ ms) as well as high
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5 photoresponsivity (4.5×10^4 AW⁻¹) and $I_{\text{light}}/I_{\text{dark}}$ (1.3×10^2) in the near-infrared spectral region at room
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7 temperature. In particular, the detectivity (D^*) is as high as 3×10^{13} Jones, which is even better than that of
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9 commercial Si and InGaAs photodetectors. Furthermore, by controlling the growth and micro-fabrication
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11 patterning, periodic device arrays of PbS-MoS₂ that are capable of infrared detection are achieved on Si/SiO₂
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13 substrates. Our work provides a possible method for the integration of photodetector arrays on Si-based
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15 electronic devices and lays a solid foundation for the practical applications of MoS₂-based devices in the future.
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18 **KEYWORDS:** non-layered PbS-MoS₂ heterostructure, edge contact, integration, infrared phototransistor
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22 Molybdenum disulfide (MoS₂) has shown remarkable potential in optoelectronic devices due to its high
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24 mobility,^{1, 2} low off-state current and high on/off ratio.³⁻¹⁴ Despite such superb performance, its intrinsically
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26 large bandgap limits its application in infrared detection. Recently, a hybrid MoS₂-PbS colloidal quantum dot
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28 (QD) phototransistor achieved an ultrahigh photoresponsivity of 10^3 AW⁻¹ at 1500 nm.¹⁵ PbS exhibits a broad
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30 spectral response from the UV to mid-IR region, which is highly desired for various applications, including
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32 thermal imaging, telecommunications, biological imaging, remote sensing and environmental monitoring.^{16, 17}
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34 Due to its excellent infrared photoconductive properties, the photoelectrical response of MoS₂ extends to the
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36 infrared spectral region. However, hybrid phototransistors have some disadvantages: (1) they suffer from large
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38 dark currents owing to the surface n-type doping from 1,2-ethanedithiol (EDT), which lowers the ratio of the
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40 light to dark current ($I_{\text{light}}/I_{\text{dark}}$) and limits detectivity. (2) They have a modest response speed ($\tau_{\text{decay}} = 0.3-0.4$ s)
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42 due to the presence of crystal boundaries between QDs and the relatively long van der Waals interaction
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44 distance in the interface.^{15, 18} In addition, the vdW gap in the interface increases the tunnel barrier of carriers
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46 from PbS to MoS₂.¹⁹ More recently, a few-layer MoS₂ phototransistor driven by ferroelectrics exhibited a fast
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48 response speed of up to 2 ms and high detectivity of 10^{12} Jones at 635 nm.²⁰ However, it exhibited a very poor
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50 responsivity (< 1 AW⁻¹ at 1500 nm) due to the weak light harvesting ability of the indirect bandgap MoS₂ in
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52 the infrared region.²¹ Despite the tremendous progress in the synthesis and application of MoS₂ for infrared
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54 detection, there remains a huge challenge of simultaneously achieving fast response speed, high
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56 photoresponsivity and detectivity in the infrared spectral region. Furthermore, detector technologies should be
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3 compatible with traditional complementary metal-oxide semiconductor (CMOS) electronics for integration
4 into high-sensitivity sensing and imaging systems.^{18, 22} Periodic arrays have shown superior performance in
5 active matrix displays and sensor arrays, which require minimal cross-talk between neighboring devices.²³⁻²⁹
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7 Therefore, integrating the non-layered PbS-MoS₂ nanostructure and forming a controllable heterostructure on
8 Si chips is a crucial subject for their practical applications.
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13 Here, we demonstrate a heterostructure phototransistor composed of non-layered PbS nanoplates that are
14 edge-contacted with a layered MoS₂ sheet. The PbS nanoplates are in contact with the edges of MoS₂ along the
15 <110> direction. Combined with density functional theory (DFT) calculations, we elucidate that PbS
16 nanoplates are in contact with MoS₂ edges via a strong chemical hybridization, which is expected to offer a
17 fast transmission path for carriers to enhance the response speed. The device presents fast response ($\tau_{\text{rising}} =$
18 $\tau_{\text{decay}} = 7.8$ ms), high responsivity (4.5×10^4 A/W) and $I_{\text{light}}/I_{\text{dark}}$ (1.3×10^2), and an ultrahigh detectivity of $3 \times$
19 10^{13} Jones in the infrared spectral region. Furthermore, by controlling the growth and micro-fabrication
20 patterning, periodic device arrays of MoS₂-PbS that are capable of infrared detection are achieved on Si/SiO₂
21 substrates. Our work paves a new way for the fabrication of high-performance infrared photodetectors and also
22 provides a possible method for the integration of photodetector arrays on Si-based electronic devices.
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34 Figure 1a exhibits the schematic illustration of epitaxial growth of non-layered PbS nanoplates-MoS₂
35 heterostructures, which were prepared by low-pressure chemical vapor deposition (LPCVD). The PbS powder
36 was thermally evaporated into vapor molecules and then self-assembled into single crystalline nanoplates on
37 MoS₂. MoS₂ was synthesized by chemical vapor deposition (CVD) from molybdenum trioxide (MoO₃) and
38 pure sulfur powders (more details are presented in the Supplementary information details). Interestingly, PbS
39 nanoplates selectively grew along the edge of the MoS₂ triangle (as observed in the optical microscope images
40 in Figure 1b). The thickness of MoS₂ is approximately 3 nm. The selected area electron diffraction (SAED)
41 pattern exhibits a perfect hexagonal arrangement, which clearly reveals the high crystallinity of the CVD MoS₂,
42 as shown in supplementary information Figure S1. The corresponding Raman spectra is presented in
43 supplementary information Figure S2 with a distance of 23 cm^{-1} between two vibrating modes at 386 and 409
44 cm^{-1} , which are attributed to CVD MoS₂ E_{2g} and A_{1g} modes, respectively.³⁰ The sharp spectral features indicate
45 the inexistence of substantial disorders in the CVD MoS₂. To verify that PbS nanoplates unambiguously have
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3 edge-contact with MoS₂, scanning electron microscopy (SEM) and atomic force microscopy (AFM) were
4 employed, and the images are depicted in the inset of Figure 1b, Figure 1c and Figure S3. Almost all PbS
5 nanoplates contacted with the MoS₂ triangle edge along the diagonal of the square PbS nanoplates. The
6 selective growth behavior could be explained by the fact that the triangular MoS₂ edge with an unsaturated
7 dangling bond is favorable for initial nucleation. The mechanism behind this can be further explained via
8 density functional theory (DFT) calculations, as shown in Figure 4. The high-resolution transmission electron
9 microscopy (HRTEM) image (Figure 1d) demonstrates that the PbS nanoplate is well-defined single
10 crystalline with clear crystal fringes of (200) planes. As shown in the inset of Figure 1d, the SAED pattern
11 clearly reveals the cubic symmetry of the nanoplates. The standard square geometry confirms the intrinsic
12 cubic crystal structure of PbS nanoplates. Therefore, PbS nanoplates have (002) top and bottom planes and
13 contact with the MoS₂ triangle edges along their <110> direction. Van der Waals epitaxy involves a weak van
14 der Waals interaction between two adjacent layers. Here, MoS₂ is used as the growth substrate because it is a
15 layered material with a highly smooth and chemically passivated surface.³¹⁻³³ The standard square geometry of
16 PbS nanoplates confirms the nature of van der Waals epitaxy as PbS shows a lattice mismatch with MoS₂; this
17 is less likely in conventional epitaxial growth. In addition, as shown in Figure S4 (a-c), the time-dependent
18 growth of PbS-MoS₂ heterostructures exhibits the following features: (1) PbS nuclei are observed at the MoS₂
19 edge (t=30 S), (2) Small PbS particles are formed and parts of the nanoplates tend to coalesce with each other
20 (t=90 S), and (3) square-shaped PbS nanoplatelets are formed (t=180 S). On the basis of the gradual evolution
21 in the morphology, we propose that PbS is formed through a mechanism based on van der Waals epitaxy.
22 Specifically, this formation phenomenon is common for a van der Waals epitaxy-based mechanism.^{29, 34, 35}
23 Figure 1e and Figure 1f exhibit the statistics of the lateral dimension distribution and thickness distribution of
24 the PbS nanoplates on MoS₂. The lateral dimension and thickness distribution of nanoplates range from 150 to
25 750 nm and 20 to 80 nm, respectively. Compared with previous van der Waals epitaxy methods, the PbS
26 nanoplates become thicker under the same reaction parameters. This indicates that the edge contact with MoS₂
27 strongly affects the morphology of PbS nanoplates.

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30 To systematically investigate the optoelectronic properties of PbS-MoS₂ heterostructures, back-gated
31 phototransistors were fabricated on n-doped silicon substrates. Electron beam lithography (EBL) was
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3 employed to pattern source/drain electrodes on as-synthesized PbS-MoS₂ heterostructures. Subsequently, metal
4 electrodes (Cr/Au, 10 nm/ 60 nm) were deposited by thermal evaporation. The schematic of a typical device is
5 shown in Figure 2a. The transfer characteristics (I_{ds} - V_{gs}) were measured under laser illumination (wavelength,
6 $\lambda = 800$ nm) with various power densities at $V_{ds} = 2$ V, $T = 300$ K and the results are depicted in Figure 2b. The
7 channel length and channel width are 3.2 and 1.3 μm , respectively. The electron mobility is estimated as 1.4
8 $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$. For an excitation wavelength of 800 nm, photogenerated carriers mainly originate from PbS
9 nanoplates, which represent the photosensitive component in the heterostructures is PbS nanoplates (bandgap,
10 $E_g = 0.4$ eV)³⁶, rather than MoS₂ ($E_g = 1.2$ eV)³⁷. We compare the optoelectronic performance between MoS₂
11 devices and PbS-MoS₂ heterostructures, as shown in Figure S5 (a-b). Although the power density is as high as
12 604 mW/cm^2 , the multilayer MoS₂ device shows modest photoresponse due to the weak light harvesting ability
13 of the indirect bandgap material, as depicted in Figure S5 a. Compared with MoS₂ devices, the PbS-MoS₂
14 heterostructure exhibits a superior photoresponse when $P = 178$ mW/cm^2 . In Figure S5 b, it is observed that the
15 rise and decay time of the photocurrent after switching exceeds 5 s for the MoS₂ device at $P = 604$ mW/cm^2 ,
16 which is similar to their analogues.²¹ In addition, a steady photoswitching behavior was observed upon
17 excitation by a 1064 nm and 1340 nm laser with power densities of 124 and 145 mW/cm^2 , respectively.
18 However, MoS₂ is not involved in photocarrier generation due to its bandgap (1.2 eV = 1033 nm).³⁷ Therefore,
19 photogenerated carriers mainly originate from PbS nanoplates in the heterostructures. Furthermore, the
20 effective irradiated area includes both the exposed MoS₂ and PbS-MoS₂ heterostructure when responsivity (R)
21 is obtained, as observed in Figure 2d. Therefore, photogenerated electrons are transferred from PbS to MoS₂,
22 which results in a pronounced photoresponse, as observed in the inset of Figure 2b. The linear output
23 characteristic (I_{ds} - V_{ds}) curves at different power densities are displayed in Figure 2c; they indicate ohmic-like
24 contact behavior. Responsivity (R) is one of the most important parameters for photodetectors, and it is defined
25 by the following formula: $R = I_{ph}/PS$, where I_{ph} is the photocurrent ($I_{ph} = I_{light} - I_{dark}$), P is the incident power
26 density and S is the effective illuminated area. The effective irradiated area is approximately 2.98 μm^2 . In
27 Figure 2d, the responsivity of our device is plotted as a function of the applied back-gated voltage at different
28 power densities. We observe that the responsivity does not monotonically increase as the gate voltage
29 increases at a fixed laser power; the responsivity is as high as 4.5×10^4 A/W when $V_{gs} = 20$ V and $P = 0.15$
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3 mW/cm². However, the responsivity decreases when $V_{gs} > 20$ V because of the reduced photocurrent, which is
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5 due to the high dark current at the on-state. The gate-modulated behavior of the responsivity indicate that the
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7 gate voltages effectively control the state of photodetectors, which highlights its potential in video frame rate
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9 imaging applications.³⁸ The specific detectivity (D^*) can be calculated by $D^* = RS^{1/2}/(2qI_d)^{1/2}$, where R , S , q
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11 and I_d are the responsivity, the effective illuminated area, the electron charge, and the dark current,
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13 respectively. Figure 2e shows that the responsivity and detectivity of this device increase linearly with the laser
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15 power, which indicates that photogenerated electron-hole pairs are effectively separated. A similar gate-
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17 tunable behavior is also observed for the photo-gain (G) and detectivity, as shown in Figure 2f. The maximum
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19 G of 7×10^4 is obtained at $V_{gs} = 20$ V and $P = 0.15$ mW/cm². However, we note that the peak value of the
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21 detectivity of 3×10^{13} Jones shifts to $V_{gs} = 0$ V because superior D^* is jointly determined by high responsivity
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23 and low dark current. These impressive results indicate an excellent performance of our photodetectors based
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25 on PbS-MoS₂ heterostructures in the near-infrared region, which is significantly superior to the performance of
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27 their analogues with hybrid structures and even better than the performance of commercial Si- and InGaAs-
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29 based devices. A comparison of critical performance indicators is provided in Table 1.
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32 To study the photoswitching characteristic and the stability of the photodetectors based on PbS-MoS₂
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34 heterostructures, the time-resolved photoresponse in the near-infrared region was measured. In Figure 3a, the
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36 current periodically changes with the alternate on-off switching of the laser, which indicates a highly stable
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38 and reversible response of the phototransistor. Figure 3b shows the plot of a photocurrent on/off switching
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40 cycle in Figure 3a. It is observed that the photodetector exhibits rapid photocurrent switching. Both the rise
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42 and decay times are found to be 7.8 ms, which is comparable to the rise and decay times of state-of-the-art 2D
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44 photodetectors. Similarly, a relatively steady photoswitching behavior is also observed for excitations by the
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46 1064 nm and 1340 nm laser under the same test condition, as displayed in Figure 3c. These phenomena further
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48 confirm that the outstanding photoresponse in the near-infrared region stems from as-synthesized PbS
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50 nanoplates. Moreover, it is worth noting that the photocurrent on-off switching ratio is as high as 10^2 at $V_{gs} = -$
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52 20 V and $P = 124$ mW/cm², which indicates an ultrasensitive ability in the detection of infrared light. Indeed,
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54 the lateral dimension of as-synthesized PbS nanoplates ranges from 150 to 750 nm, which gives rise to the
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56 relatively low surface coverage of PbS nanoplates on the MoS₂. However, a fast response time and high
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3 photoresponsivity could still be realized in our devices for different surface coverage. As shown in Figure S6 a,
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5 the rise and decay times of PbS-MoS₂ heterostructures with a coverage of 9% approach 15.6 and 23.4 ms,
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7 respectively. Similarly, an ultrafast response time of 7.8 ms is also obtained for the same device with coverage
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9 of 37% and 43%, as observed in Figure S6 (b-c). This indicates that the response time is less affected by the
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11 coverage area of PbS. Moreover, we plotted the responsivity as a function of surface coverage of PbS. Figure
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13 S6 d shows that the responsivity exhibits an increased trend as the coverage of PbS increases. We attribute the
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15 fast and sensitive response to the novel edge-contact between MoS₂ and PbS nanoplates. There are three key
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17 factors that determine the high performance of the phototransistor. Firstly, PbS nanoplates contact with MoS₂
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19 edges by a strong chemical hybridization, which is expected to offer a fast transmission path for carriers that
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21 enhances the response speed. Secondly, PbS nanoplates are highly single-crystalline (Fig. 1d), and they
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23 provide a long-range order for the electronic transport path with reduced boundary defect states where
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25 scattering can occur. Finally, the vdW interaction distance in our heterostructures should be smaller than that
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27 in hybrid colloidal PbS quantum dot-MoS₂ structures,^{15, 38} which is beneficial for the transfer of
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29 photogenerated electrons from PbS to MoS₂. Significantly, the edge-contact plays a crucial role in the high
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31 performance of the phototransistor because the seamless contact between PbS and MoS₂ through the chemical
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33 bonding interaction aids carrier transport in the interface. Recently, a low average contact resistance of 30
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35 kΩ.μm has been achieved for the graphene and TMD layer with edge contacts.³⁹ Although the area of the
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37 edge-contact is much smaller than that of the total interface, it can provide a lower contact resistance and
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39 enhance the transfer of photogenerated electrons from PbS to MoS₂. A detailed explanation and theoretical
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41 simulation will be discussed below.
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44 From our experiments, we clearly observe an interesting phenomenon of PbS nanoplates-MoS₂
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46 heterostructures where all of the PbS nanoplates are in contact with the edges of MoS₂ along the <110>
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48 direction. Simultaneously, the abundant unsaturated Mo atoms on the edge of MoS₂ result in a high activity of
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50 the edge area. This fact indicates the possibility that the PbS nanoplates is primarily nucleated at the edge, and
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52 the heterostructures were mainly maintained by the chemical bonds on the edge-contact area. Hence, to verify
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54 the viewpoints proposed above, we calculated the binding energy of the edge-contact model. The binding
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56 energy is defined as follows:
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$$E_{\text{bind}}=(E_{\text{PbS/MoS}_2}-E_{\text{PbS}}-E_{\text{MoS}_2})/N \quad (1-1)$$

where $E_{\text{PbS/MoS}_2}$ represents the total energy of the PbS-MoS₂ heterostructures, and E_{PbS} and E_{MoS_2} represent the total energy of the PbS and MoS₂, respectively; moreover, N represents the number of Mo-S bonds that are formed between Mo of MoS₂ and S of PbS (110). To verify the chemical bonding on the edge, we calculated the charge density difference ρ , which is defined as follows:

$$\rho=\rho(\text{PbS/MoS}_2)-\rho(\text{PbS})-\rho(\text{MoS}_2) \quad (1-2)$$

where $\rho(\text{PbS/MoS}_2)$, $\rho(\text{PbS})$, $\rho(\text{MoS}_2)$ represents the charge density of the PbS/MoS₂ edge-contact model, PbS(110) and MoS₂ nanoribbon, respectively.

The schematic diagram of the edge-contact between the MoS₂ and PbS is presented in Figure 4a. The Mo atoms of the zigzag edge of MoS₂ were bonded with the S atom chain that is exposed to PbS (110). The inset is a top view of the schematic diagram. In our calculations, due to the very low lattice mismatch (0.1%) along the zigzag direction of MoS₂, each Mo atom perfectly bonded with two S atoms. Therefore, there are 6 Mo-S bonds in the edge-contact region in a repeat unit. Additionally, every single bond length was marked in Figure 4b. We clearly obtained an average Mo-S bond ca. 2.433 Å in the edge-contact region, which is similar with the value in MoS₂ (2.412 Å). Furthermore, the binding energy of the edge-contact model is -1.73eV per Mo-S bond. Both the bond length and binding energy indicated that the chemical bonds were formed in the edge-contact region. For further confirmation, we calculated the charge density difference (Figure. 4c) and PDOS (Figure. 4d) of the edge-contact region. Here, the red region indicates the charge accumulation region and the green region indicates the charge depletion region in Figure. 4c. We clearly observe that the charge density increases in the bonding region and decreases around the Mo-S bonds; in addition, the charge density increases to a larger extent at the Mo atoms. In other words, it is obvious that the ionic bonds were formed in the edge-contact region. Moreover, the bonding detail was presented by the PDOS of the edge-contact region, as observed in Figure. 4d. The Mo-4d orbital and S-3p orbital clearly undergo orbital hybridization. Overall, the evidence from both the binding energy and electronic structure indicate that an ionic bond is formed by the S-3p and Mo-4d in the edge-contact region.

Periodic arrays have shown superiority in active matrix displays and sensor arrays, which require minimal cross-talk between neighboring devices. In addition, detector technologies should be compatible with

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3 traditional metal-oxide semiconductor (CMOS) electronics for integration into high-sensitivity sensing and
4 imaging systems. Therefore, integrated PbS-MoS₂ heterostructure periodic arrays on Si chips are of great
5 importance in practical applications. Figure 5a shows MoS₂ patterning performed by a photolithography
6 process followed by O₂ plasma etching. The width of the MoS₂ pattern is 5 μm. The PbS nanoplates selectively
7 grew on MoS₂ via a carefully controlled growth condition, as displayed in Figure 5b. This behavior could be
8 explained by classical nucleation kinetics. Compared with the larger surface energy of SiO₂ ($\gamma_{\text{SiO}_2} \approx 300\text{--}400$
9 mJ m⁻²), the surface energy of MoS₂ is 70–75 mJm⁻².^{40, 41} Therefore, epitaxial PbS on SiO₂/Si must overcome
10 a larger free energy barrier. In contrast, the growth of PbS on MoS₂ should be relatively easy. In particular,
11 PbS nanoplates grew on the entire MoS₂ pattern. This phenomenon could be attributed to the presence of
12 photoresist residues as a result of the photolithography process because PbS is easily nucleated at certain
13 defect sites. As is evident from Figure 5c, PbS nanoplates-MoS₂ heterostructures 3×3 arrays were directly
14 fabricated by electron beam lithography for practical high-performance infrared phototransistor applications;
15 this indicates the potential scalability of our heterostructure. Figure 5d shows a schematic illustration of the
16 PbS nanoplates-MoS₂ heterostructure arrays. As observed in Figure S5 c, although the device from the PbS-
17 MoS₂ pattern exhibits rapid photoswitching speeds of $\tau_{\text{rising}} = 51.3$ ms and $\tau_{\text{decay}} = 102.7$ ms, the photoswitching
18 speeds are much slower than those of the previously mentioned edge-contact heterostructure. The photoresist
19 residues can act as trap state centers, which can trap the photo-excited carriers. The performance will be
20 improved and the potential trapping will be overcome as the fabrication technology becomes mature.

21
22 In summary, we have synthesized non-layered PbS and layered MoS₂ heterostructures via van der Waals
23 epitaxy. The device exhibits fast response ($\tau_{\text{rising}} = \tau_{\text{decay}} = 7.8$ ms), high photoresponsivity (4.5×10^4 AW⁻¹) and
24 $I_{\text{light}}/I_{\text{dark}}$ (1.3×10^2), and an ultrahigh detectivity of 3×10^{13} Jones. In addition, the photoresponse wavelengths
25 extend to the infrared region (0.8–1.34 μm). Moreover, in conjunction with density functional theory (DFT)
26 calculations, we elucidate that PbS nanoplates form edge-contacts with MoS₂ via a strong chemical
27 hybridization. Furthermore, we achieved precise control of the PbS nanoplates-MoS₂ heterostructure periodic
28 arrays on the Si/SiO₂ substrate. This study enables the large-scale fabrication of PbS-MoS₂ phototransistors for
29 practical high-performance photodetector applications.

Methods

Device fabrication. Large-area MoS₂ was synthesized by chemical vapor deposition (CVD) from molybdenum trioxide (MoO₃) and pure sulfur powders. Insulating SiO₂/Si layers were used as the growth substrates, and the zone with MoO₃ is heated to 850 °C. The process is performed at atmospheric pressure under the flow of ultrahigh-purity argon. The mechanically exfoliated MoS₂ was prepared by standard mechanical exfoliation with scotch tape. The PbS powder was thermally evaporated into vapor molecules and then self-assembled into single crystalline nanoplates on MoS₂ under a flow of 20 sccm of H₂ gas. The source and substrate temperatures were set to 750 °C and 500 °C, respectively. Back-gated phototransistors were fabricated on n-doped silicon substrates. Electron beam lithography (EBL) was employed to pattern source/drain electrodes on as-synthesized PbS-MoS₂ heterostructures. Subsequently, metal electrodes (Cr/Au, 10 nm/ 60 nm) were deposited by thermal evaporation.

Microstructure characterization. The PbS nanoplates-MoS₂ heterostructure was characterized by optical microscopy (Leica DM4000M microscope), FESEM S4800 (Tokyo, Japan), AFM (Dimension 3100), FEI Tecnai F20 equipped with SAED, and Raman spectroscopy (Renishaw InVia, 532 nm excitation laser).

DFT computational details. The structural properties of MoS₂ and PbS were calculated by density functional theory (DFT), which was performed by the Vienna Ab Initio Simulation Package (VASP) version 5.3.5.^{42, 43} The projector augmented wave potentials and GGA functional of PBE were applied.^{44, 45} A kinetic energy cutoff of 500 eV was set for wave functions in a plane-wave basis. For representing the experiment, we simulated the edge-contact between MoS₂ and PbS by adopting the MoS₂ ribbons model and the PbS (110), whose surface is composed of Pb atomic chains and S atomic chains. Hence, 1 × 9 × 1 Monkhorst–Pack k-point meshes⁴⁶ for the edge-contact model and a vacuum region of ~15 Å along the Z-direction were used to achieve the converged results. In addition, all the MoS₂ and only two layers of the PbS slab were relaxed until the maximum Hellmann–Feynman force acting on each atom was less than 0.03 eV/Å and the convergence for the total energy between two consecutive steps was less than 1 × 10⁻⁴ eV.

Photoresponse measurement. All the measurements were performed by a manual probe station at room temperature. The devices were placed in a sealed chamber at a vacuum of 10⁻⁶ torr. The parameters were analyzed using a Keithley 4200 semiconductor characterization system.

ASSOCIATED CONTENT

Supporting Information

Experimental details, AFM images, SAED, Raman spectra. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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Notes

The authors declare no competing financial interest. †These authors contributed equally to the work.

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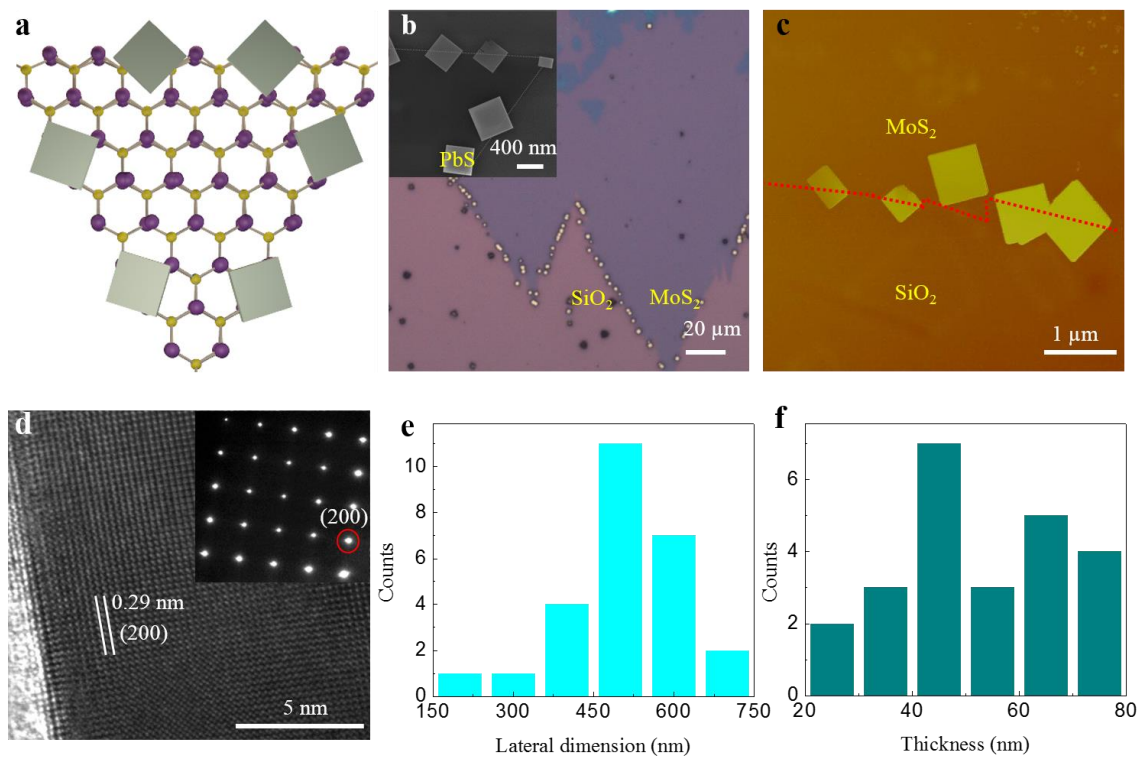


Figure 1. Morphology of non-layered PbS nanoplates-MoS₂ heterostructures. (a) Schematic illustration of PbS nanoplates-MoS₂ heterostructures. (b) OM image with selectively growing PbS nanoplates along the edge of MoS₂ triangle. The inset in (b) is an amplified SEM image of edge contact heterostructures. (c) AFM image of edge of MoS₂ triangle. (d) HRTEM image with crystal fringes of (200) planes. The SAED cubic pattern is shown in the inset of (d). The histogram statistics of PbS nanoplates: the lateral dimension and thickness distribution ranging from 150 to 750 nm and 20 to 80 nm, respectively.

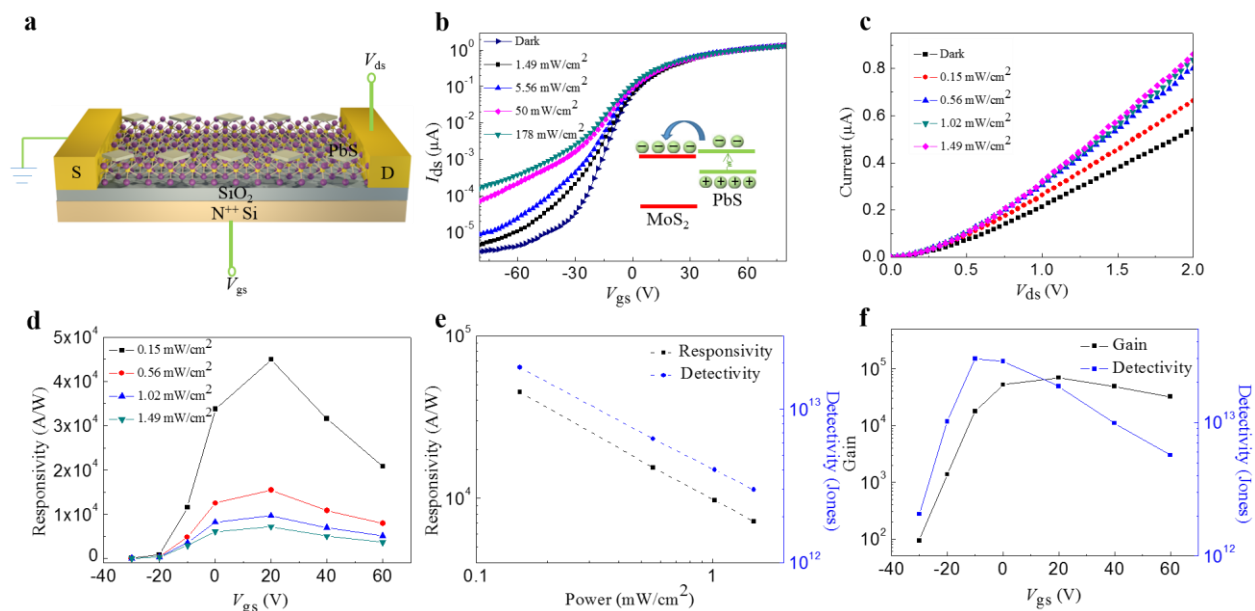


Figure 2. Gate voltage-tunable responsivity and detectivity of epitaxial PbS nanoplates-MoS₂ heterostructures. (a) Schematic of a typical device. (b) Transfer characteristic (I_{ds} - V_{gs}) measurements under laser illumination (wavelength, $\lambda = 800$ nm) with power densities varied from 1.49 to 178 mW/cm² were performed at $V_{ds} = 2$ V and $T = 300$ K. (c) The output characteristic (I_{ds} - V_{ds}) curves with different power densities at $V_{gs} = 20$ V. (d) The responsivity of the heterostructures is plotted as a function of the applied back-gated voltage. (e) The plots of R and D^* versus P for different power densities at $V_{gs} = 20$ V. (f) The gate-tunable behavior for photo-gain (G) and detectivity with $P = 0.15$ mW/cm².

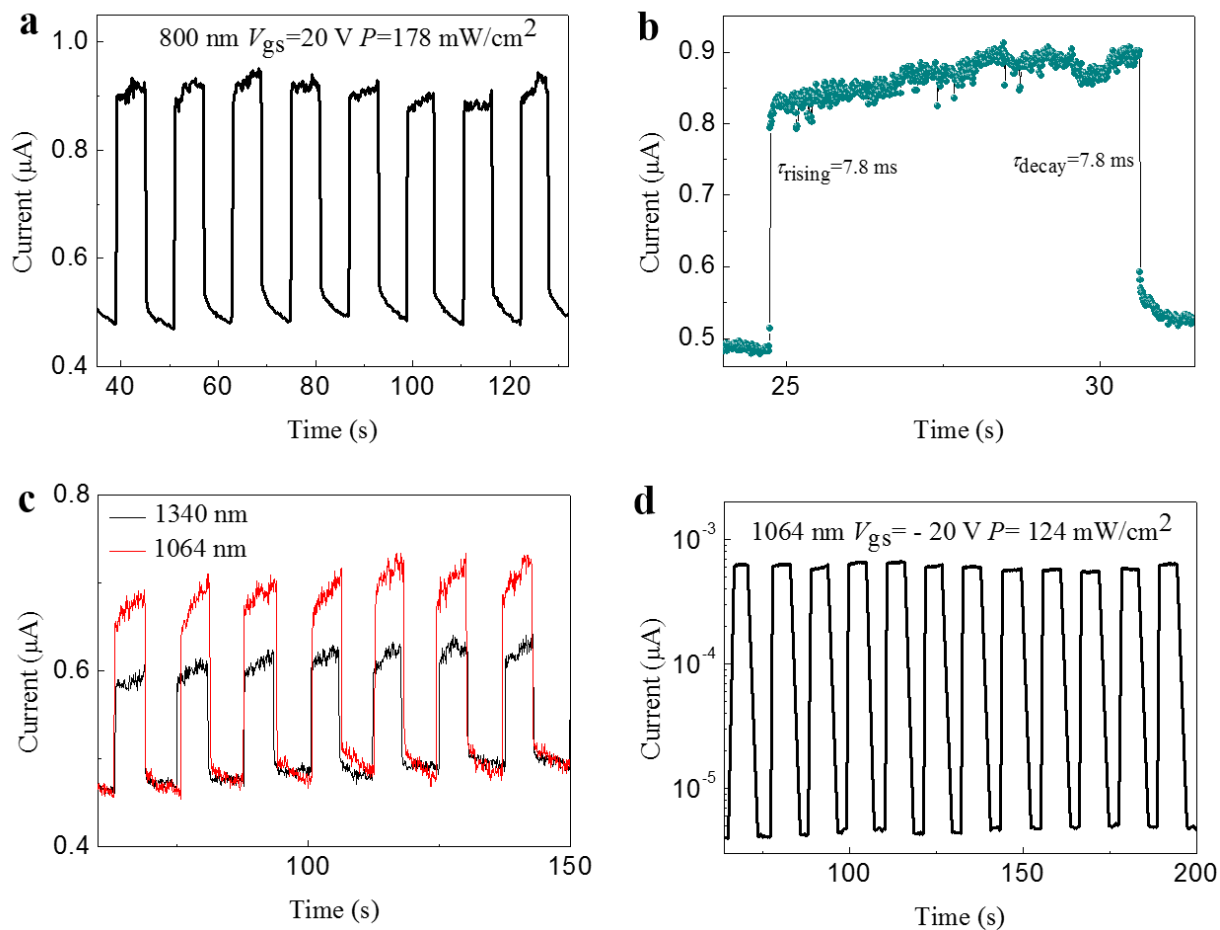


Figure 3. Time-resolved photoresponse of epitaxial PbS nanoplates-MoS₂ heterostructures. (a) Time-resolved photoresponse was measured at 800 nm, which indicates a highly stable and reversible response of the phototransistor. (b) Temporal response exhibits fast photoswitching ($\tau_{\text{rising}} = \tau_{\text{decay}} = 7.8$ ms). (c) Time-resolved photoresponse in the near-infrared region at 1064, 1340 nm with power densities of 124 and 145 mW/cm², respectively. (d) The ratio of photocurrent on-off switching is as high as 10^2 at $V_{gs} = -20$ V and $P = 124$ mW/cm².

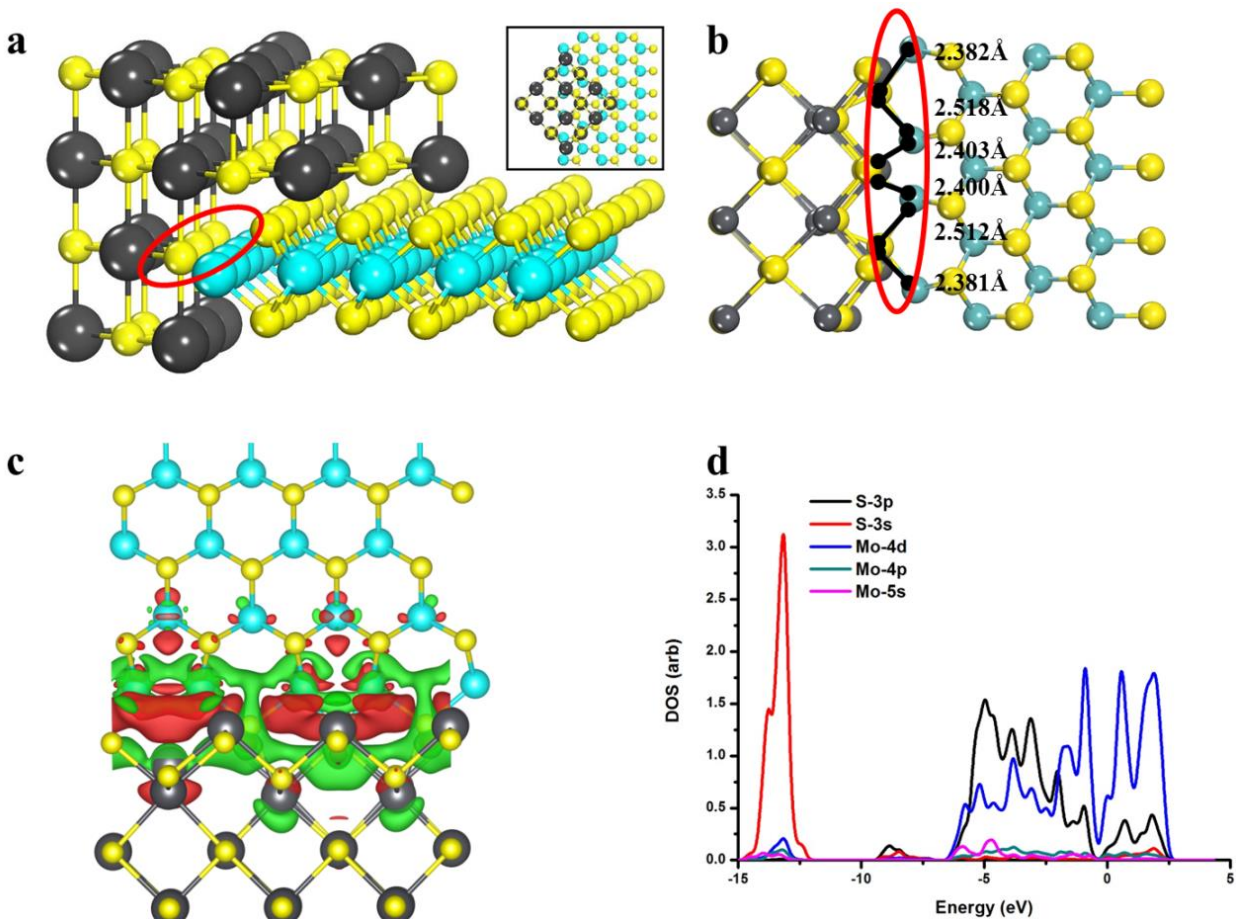


Figure 4. The edge-contacts of PbS nanoplates-MoS₂ heterostructures were calculated by density functional theory (DFT) (a) The schematic diagram of edge-contact between the MoS₂ and PbS, and the inset is a top view of the schematic diagram. (b) The bonding situation in the edge-contact region. (c) The charge density difference of the edge-contact; the red and green regions represent charge accumulation and depletion regions, respectively. (d) PDOS of the edge-contact region, the Fermi energy was set to zero. The black ball is C, the yellow ball is S and the blue-green ball is Mo.

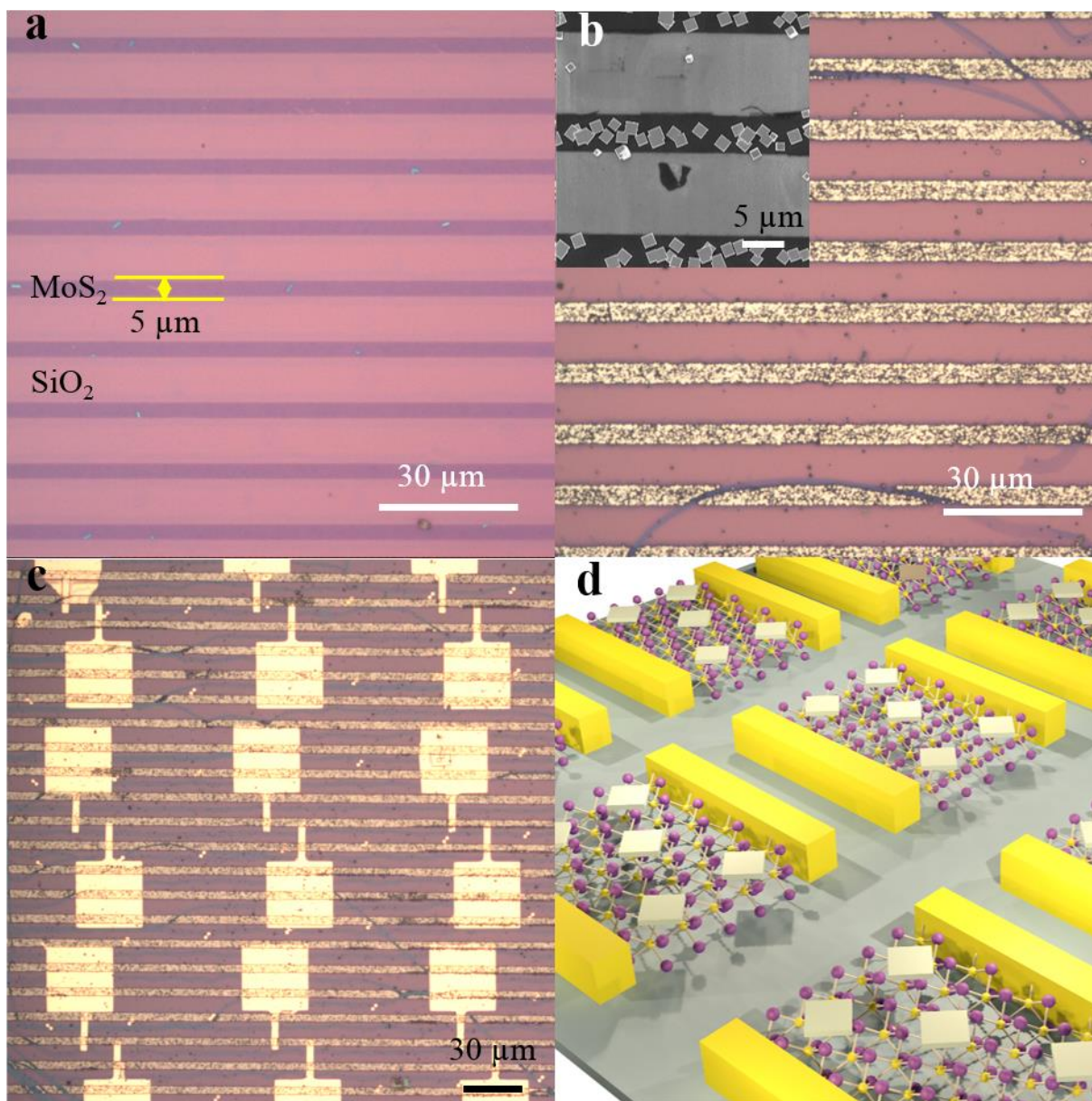


Figure 5. Scalable fabrication of the heterostructure. (a) MoS₂ patterning by a photolithography process followed by O₂ plasma etching. (b) The PbS nanoplates selectively grew on MoS₂ via a carefully controlled growth condition. (c) The PbS nanoplates-MoS₂ heterostructure 3×3 arrays were directly fabricated by an electron beam lithography process. (d) Schematic illustration of PbS nanoplates-MoS₂ heterostructure arrays.

	Wavelength	Response time	I_{ph}/I_{dark}	Detectivity	Responsivity	Ref.
commercial Si	-	-	-	$\sim 10^{12}$	-	47
commercial InGaAs	-	-	-	$\sim 10^{12}$	-	48
GaAs/AlGaAs NW	-	-	10^2	7.2×10^{10}	0.57 AW^{-1}	47
CdS-MoS ₂	610 nm	$\tau_{\text{rising}} = 100 \text{ ms}$	-	-	3.91 AW^{-1}	40
PbS quantum dots-MoS ₂	635 nm	$\tau_{\text{decay}} = 350 \text{ ms}$	-	$\sim 10^{11}$ ($V_g = -60 \text{ V}$)	$6 \times 10^5 \text{ AW}^{-1}$	15
PbS nanoplates-MoS ₂	800 nm	$\tau_{\text{rising}} = \tau_{\text{decay}} = 7.8 \text{ ms}$	10^2	10^{13} ($V_g = 0 \text{ V}$)	$4.5 \times 10^4 \text{ AW}^{-1}$	This work

Table 1. Comparison of critical optoelectronic parameters in various photodetectors.