

ratio of photocurrent to dark current, response speed (millisecond level) and low responsivity (mA/W level) [27]; these aspects hinder its further application in light detection. Based on this situation, many researchers have carried out numerous studies to improve the photoelectric performance by applying a gate voltage or constructing heterostructures. Wright *et al.* [28] fabricated four-layer MoTe₂ field effect transistors (FETs) with a photoresponse time of ~160 μs under 685 nm laser illumination, and the photoresponsivity was only 6 A/W even when a large gate voltage of -40 V was applied. He *et al.* [29] also reported a MoTe₂ phototransistor, which exhibited a responsivity of ~10² A/W at zero gate voltage with an increase reaching up to 2560 A/W at 80 V gate voltage under 473 nm laser illumination. Chu *et al.* [30] developed a few-layer MoTe₂ photodetector, which demonstrated a photoresponsivity of 50 mA/W and a detectivity of 3.1 × 10⁹ cm·Hz^{1/2}·W⁻¹ for 637 nm light at a back gate voltage of 10 V. Then, they designed a self-powered MoTe₂/MoS₂ vertical heterojunction photodetector, which possessed a high on/off ratio (>10⁵) and a fast response time of 60 μs. However, the photoresponsivity remained relatively low at 46 mA/W [31]. Lu *et al.* [32] fabricated a 1D Ga-doped CdS nanowire/2D MoTe₂ flake heterojunction device, achieving a self-driven photodetector with a response time of less than 50 ms and a photoresponsivity of less than 60 mA/W. Chen *et al.* [33] fabricated a MoTe₂/WSe₂ heterojunction photodetector, achieving a high on/off ratio of ~10⁴ and a low response time of 72 μs under illumination with a 633 nm laser; however, the responsivity was only 1.8 mA/W. Jie *et al.* [34] reported a few-layer MoTe₂/Si 2D-3D vertical heterojunction photodetector. Their device possessed an ultrahigh response speed up to 150 ns and a large on/off ratio of 3.2 × 10⁶ under 980 nm light illumination; however, the responsivity and the EQE were only 0.19 A/W and 24%, respectively. Although much work has been carried out in MoTe₂-based photodetectors, some key parameters (such as responsivity) still need significant improvement. Moreover, numerous research has been performed on visible light imaging of MoTe₂-based photodetectors. Therefore, more efforts need to be made to improve the photodetection performance of the MoTe₂-based photodetectors. Constructing heterojunctions is considered an effective method to improve the photoelectric performance of photodetectors via energy band engineering; this engineering can enhance the photocurrent, suppress the dark current and promote the effective separation of the photogenerated carriers. Loutfy *et al.* [35] reported that CdS_{0.5}Se_{0.5} had the highest photoelectrochemical conversion efficiency, achieved by optimizing the composition in CdS_{1-x}Se_x thin films. Additionally, we successfully fabricated high-quality CdS_{0.42}Se_{0.58} flakes, in which the composition was similar to that of CdS_{0.5}Se_{0.5}. To the best of our knowledge, few-layer MoTe₂ and CdS_{0.42}Se_{0.58}

flake heterojunctions have not yet been reported.

In this work, we fabricated a MoTe₂/CdS_{0.42}Se_{0.58} flake heterojunction to achieve a high-performance photodetector, with high responsivity (7221 A/W), large current on/off ratio (1.73 × 10⁴), fast response speed (90/120 μs), large EQE (1.52 × 10⁶ %) and detectivity (1.67 × 10¹⁵ Jones). In view of the high performance, we investigated the visible light imaging of heterojunction devices, which indicated the great potential of MoTe₂/CdS_{0.42}Se_{0.58} photodetectors for room-temperature visible imaging. The microphysical mechanism of the excellent performance of the MoTe₂/CdS_{0.42}Se_{0.58} photodetectors was determined based on DFT calculations. Our study has shown that the MoTe₂/CdS_{0.42}Se_{0.58} heterojunction has potential applications in high-performance photodetectors due to its excellent optical properties and imaging capabilities.

2 Experimental section

2.1 Preparation of CdS_{0.42}Se_{0.58} and MoTe₂ flakes

The CdS_{0.42}Se_{0.58} material was synthesized within a tubular furnace outfitted with a quartz tube measuring 25 mm in diameter. Initially, high-purity CdS and CdSe powders with a purity of 99.99% were combined in a ceramic boat in a 1:1 molar ratio. Subsequently, the mixture was placed in the central evaporation zone within the quartz tube. Next, silicon wafers were cut into slices measuring 1.5 cm × 1.0 cm and immersed in a beaker containing acetone. After undergoing ultrasonic cleaning for 10 minutes in the acetone solution, the silicon substrates were further rinsed for 10 minutes with deionized water. Following the cleaning process, the silicon substrates were coated with a layer of gold particles and positioned at the upstream and downstream ends of the quartz tube. Finally, the quartz tube was introduced into the tubular furnace, and the temperature was gradually increased to 835 °C at a rate of 5.6 °C/min. The system was maintained at this temperature for 150 minutes under a flow of 17 sccm Ar:H₂ gas mixture with a ratio of 95:5. Subsequently, the furnace was allowed to naturally cool to room temperature, and the CdS_{0.42}Se_{0.58} flake was obtained. The few-layer MoTe₂ flake was obtained by a mechanical exfoliation method.

2.2 Preparation of the MoTe₂/CdS_{0.42}Se_{0.58} heterojunction photodetector

First, Cr and Au were sequentially deposited on a clean silicon substrate by using electron beam evaporation equipment to form electrodes with a channel length of 5 μm. Second, the MoTe₂ flake was transferred to the gold electrodes by a 2D material transfer system. Subsequently, the fabricated CdS_{0.42}Se_{0.58} flakes were soaked

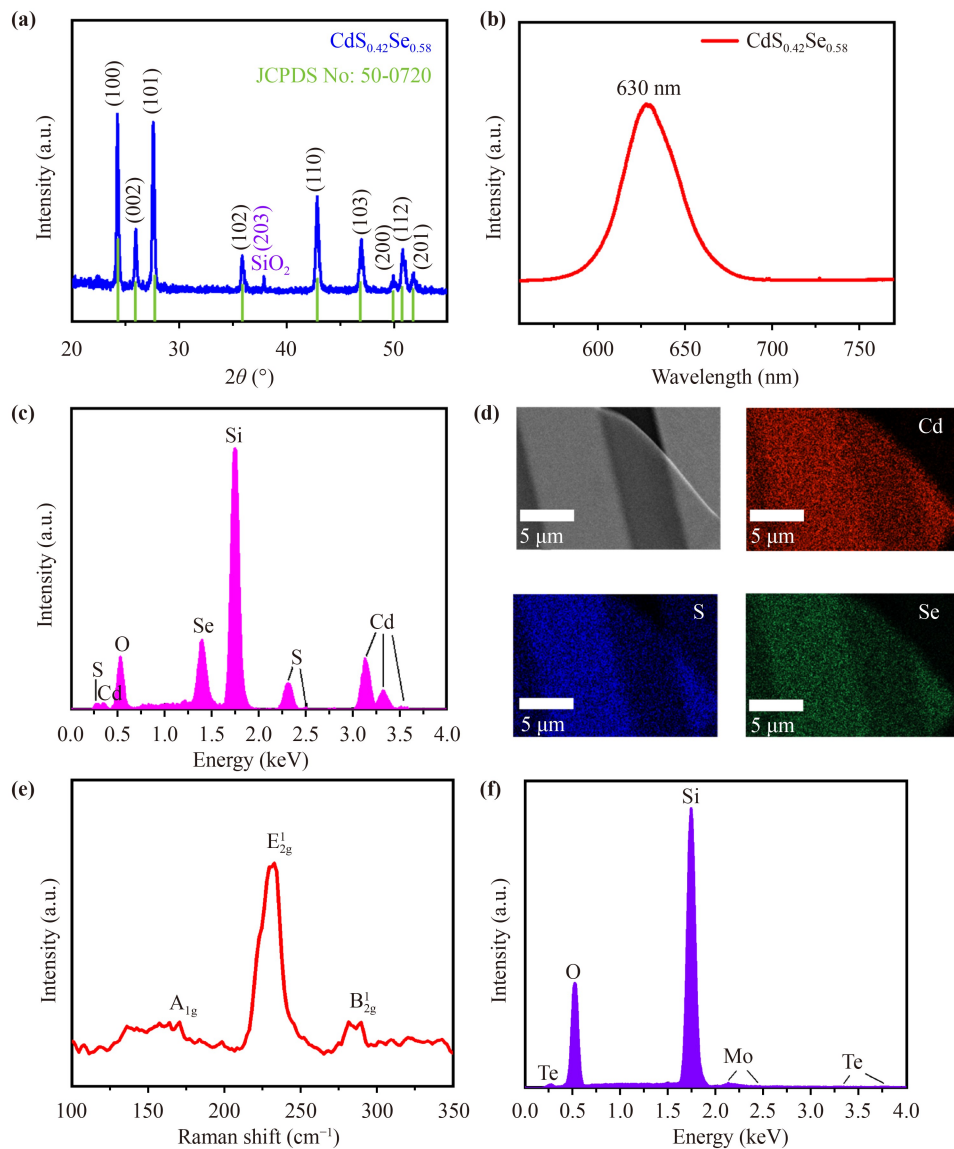


Fig. 1 (a) XRD pattern, (b) PL, (c) EDS and (d) elemental mapping of $\text{CdS}_{0.42}\text{Se}_{0.58}$ flake. (e) Raman spectroscopy and (f) EDS of few-layer MoTe_2 flake.

in an ethanol solution and then added dropwise onto another clean silicon substrate. The $\text{CdS}_{0.42}\text{Se}_{0.58}$ flake remained on the substrate following the evaporation of ethanol. Finally, the $\text{CdS}_{0.42}\text{Se}_{0.58}$ flake was detached from the initial substrate and placed above the MoTe_2 flake using a PDMS film, thus completing the construction of the heterojunction device.

2.3 First-principles calculations

All first-principles calculations were performed within the framework of the DFT-LCAO method as implemented in the QuantumATK T-2022.03-SP1 software package [36]. To simulate the trilayer $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ heterojunction, a slab model consisting of 336 atoms was built. The vacuum layer thickness was ~ 22 Å, and the

atomic ratios of Cd, S and Se agreed with the experimental values of 1:0.42:0.58 in $\text{CdS}_{0.42}\text{Se}_{0.58}$ NB. The Grimme DFT-D2 method [37] was adopted in the structure optimization to consider the van der Waals correction. The total energy and force converged to $\sim 10^{-4}$ eV and 0.05 eV/Å, respectively. The cutoff energy and k -point grid were set to 105 Hartrees and $4 \times 4 \times 1$, respectively. The HSE hybrid functional was used to more accurately calculate the projected density of states of the heterostructure.

3 Results and discussion

Figure 1(a) shows the X-ray diffraction (XRD) pattern of the $\text{CdS}_{0.42}\text{Se}_{0.58}$ flake. Evidently, the diffraction peak

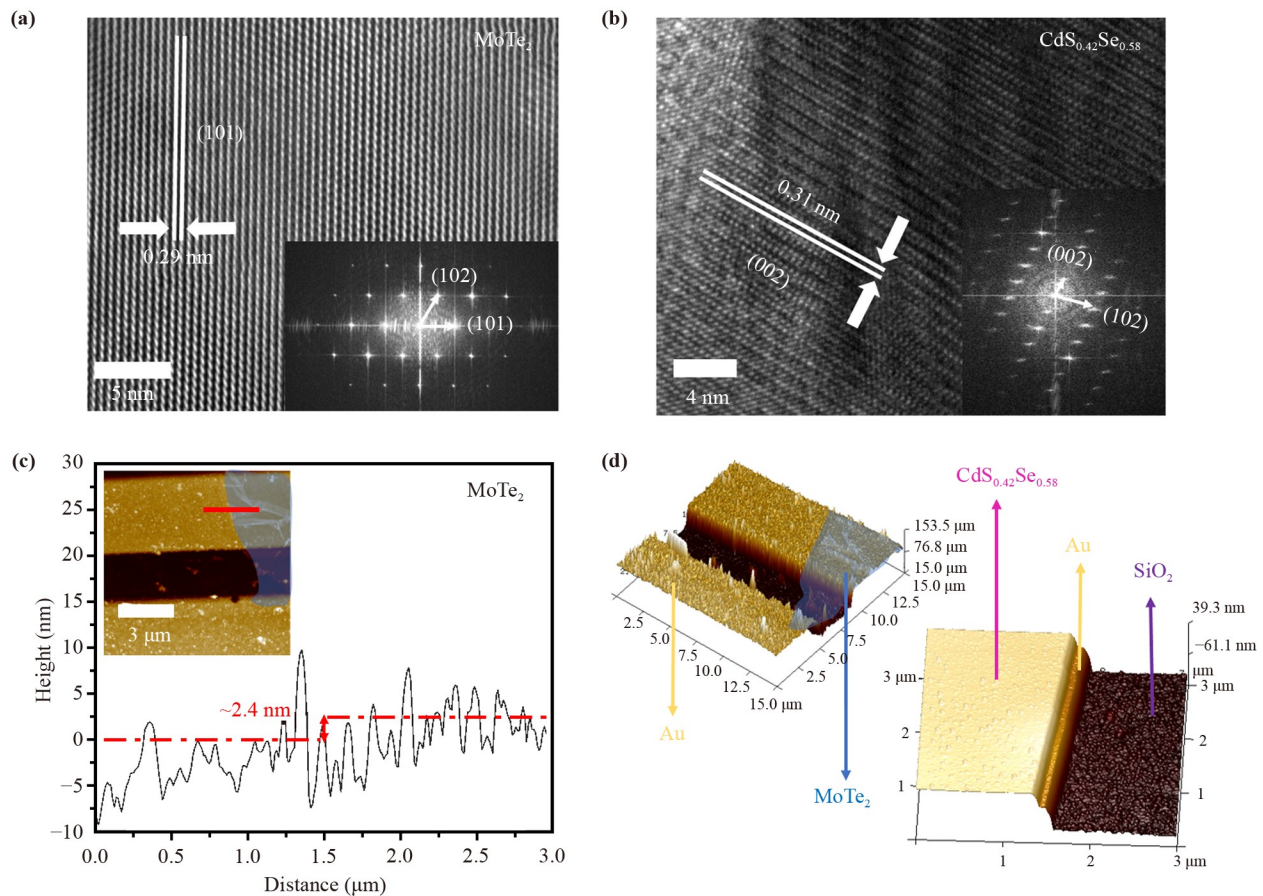


Fig. 2 HRTEM images of (a) MoTe₂ and (b) CdS_{0.42}Se_{0.58}. Insets are the corresponding FFT patterns. (c) AFM profile along the red line in the inset. The inset is the 2D AFM of the isolated MoTe₂ region in the heterojunction device. (d) 3D AFM topographies of isolated MoTe₂ and CdS_{0.42}Se_{0.58} regions in the heterojunction device.

is consistent with the JCPDS No: 50-0720 standard card [38], in which the ratio of S to Se is 0.42:0.58. A small peak emerged at approximately $2\theta = 37.8^\circ$, which correlates to the (203) diffraction peak of the SiO₂ substrate by comparison with the JCPDS No: 47-1300 card. Figure 1(b) shows the photoluminescence (PL) spectra for the CdS_{0.42}Se_{0.58} flake. A peak is observed at a wavelength of 630 nm; from this peak, the band gap of CdS_{0.42}Se_{0.58} is calculated to be approximately 1.9 eV. Figure 1(c) shows the energy-dispersive X-ray spectroscopy (EDS) of the CdS_{0.42}Se_{0.58} flake. Si, O, Cd, Se, and S are present in the CdS_{0.42}Se_{0.58} flake, in which Si and O originate from the SiO₂ substrate. Figure 1(d) shows the elemental mapping image of the CdS_{0.42}Se_{0.58} flake. These elements are evenly distributed in the CdS_{0.42}Se_{0.58} flake. The magnified SEM showed exceptionally smooth surfaces of the CdS_{0.42}Se_{0.58} flake, demonstrating the excellent crystalline quality of the CdS_{0.42}Se_{0.58} flakes. Figure 1(e) shows the Raman spectrum of the few-layer MoTe₂ flake; the layer number of MoTe₂ obtained by mechanical exfoliation is 3 layers according to the positions and intensities of the A_{1g}, E_{2g}¹ and B_{2g}¹ vibration modes in the Raman spectrum [32]. Figure 1(f) shows the EDS

diagram of the few-layer MoTe₂ flake. The EDS diagram clearly shows the presence of Mo, Te, O, and Si, providing conclusive evidence that the few-layer MoTe₂ flake consists of Mo and Te, while elements O and Si can be attributed to the SiO₂ substrate. The relatively low detected values of Te and Mo can be attributed to the thin MoTe₂ layer and the abundant distribution of the SiO₂ substrate.

To further investigate the morphology, crystal structure, and surface roughness of the fabricated MoTe₂ and CdS_{0.42}Se_{0.58} flakes, we carried out high-resolution transmission electron microscopy (HRTEM) and atomic force microscopy (AFM) measurements. Figures 2(a) and (b) show the HRTEM images of the MoTe₂ and CdS_{0.42}Se_{0.58} flakes, respectively; the lattice spacings of 0.29 and 0.31 nm corresponded to the (101) and (002) planes of MoTe₂ and CdS_{0.42}Se_{0.58}, respectively. The fast Fourier transform (FFT) images (insets) of the two materials indicated their high-quality single-crystalline nanostructure. The step height of 2.4 nm at the edge of the flake indicated that the exfoliated MoTe₂ flake was trilayer [see Fig. 2(c)]; this result effectively agreed with the Raman spectral analysis. The 3D AFM topographies [see

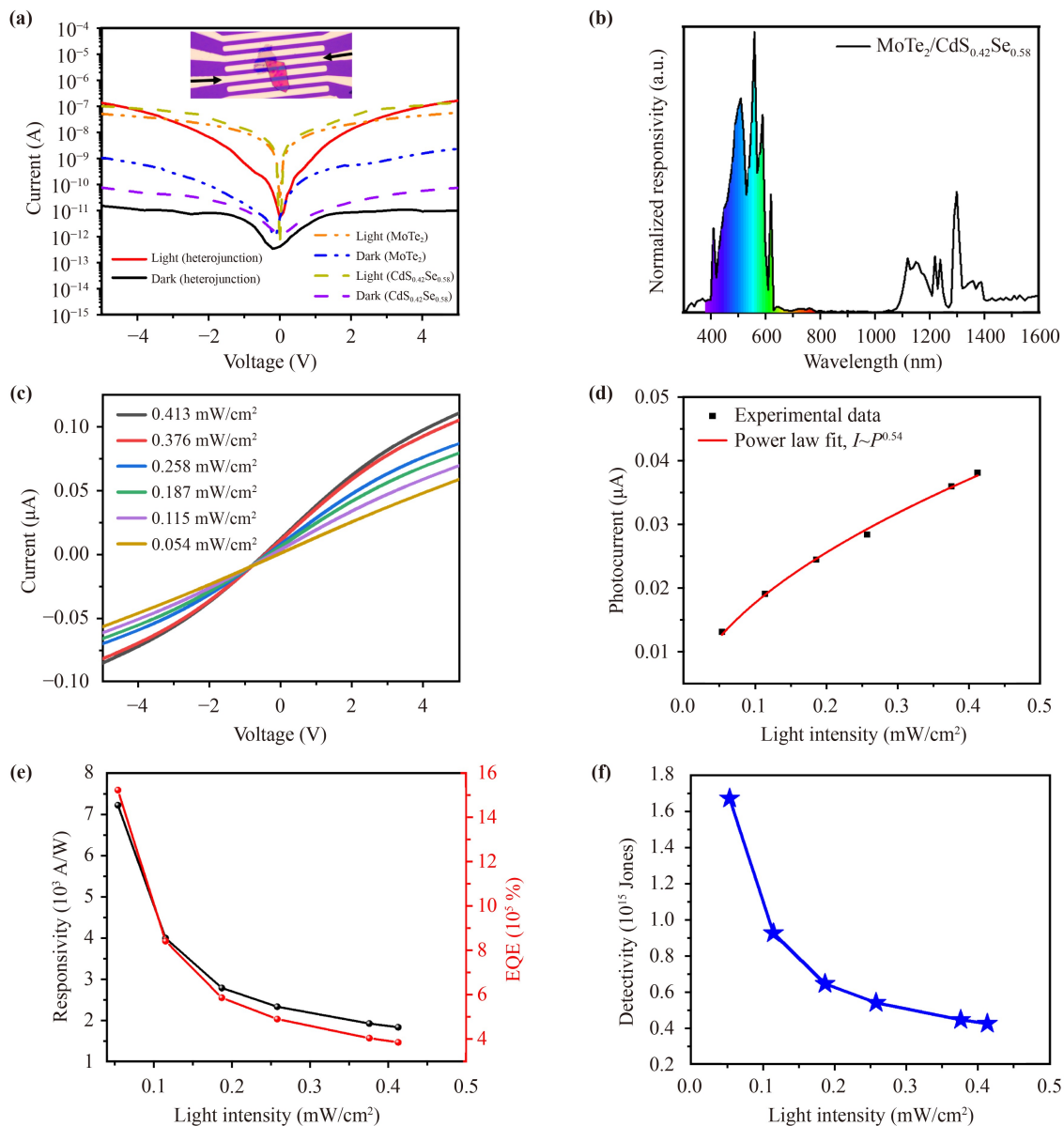


Fig. 3 (a) I - V curves of the single few-layer MoTe₂ flake, single CdS_{0.42}Se_{0.58} flake and MoTe₂/CdS_{0.42}Se_{0.58} flake heterojunction devices under white light of 1.5 mW/cm². The inset is an optical image of the device, in which the black arrows point to the selected test electrodes. (b) Responsivity as a function of the illumination wavelength. (c) I - V curves of the device with different light power densities. (d) Photocurrent as a function of light intensity. (e) Dependence of R , EQE and (f) D^* with respect to power density under 560 nm light irradiation at a 5 V bias.

Fig. 2(d)] indicated a smooth surface of the fabricated MoTe₂ and CdS_{0.42}Se_{0.58} flakes.

Figure 3(a) plots the current-voltage (I - V) curve of the few-layer MoTe₂/CdS_{0.42}Se_{0.58} flake heterojunction photodetector under dark and white-light illumination. For comparison, the I - V characteristics of the single MoTe₂ and CdS_{0.42}Se_{0.58} devices were also plotted under the same conditions. The photocurrent drastically increased with increasing bias voltage. At a bias of 5 V, the MoTe₂ device showed a low on/off current ratio (~ 24) due to its high dark current. For the CdS_{0.42}Se_{0.58} device, the on/off current ratio reached up to 1.87×10^3

because of its high photocurrent and low dark current. When the two materials were coupled to form a heterojunction, the photocurrent slightly increased (1.66×10^{-7} A), while the dark current decreased by almost one order of magnitude (9.62×10^{-12} A) compared to that of the CdS_{0.42}Se_{0.58} device. This significant dark current suppression resulted in the on/off current ratio of the device reaching a high value of 1.73×10^4 . Therefore, the MoTe₂/CdS_{0.42}Se_{0.58} flake heterojunction photodetector exhibited enhanced photoelectric performance. Figure 3(b) shows the spectral response of the heterojunction photodetector ranging from 300 to 1600 nm.

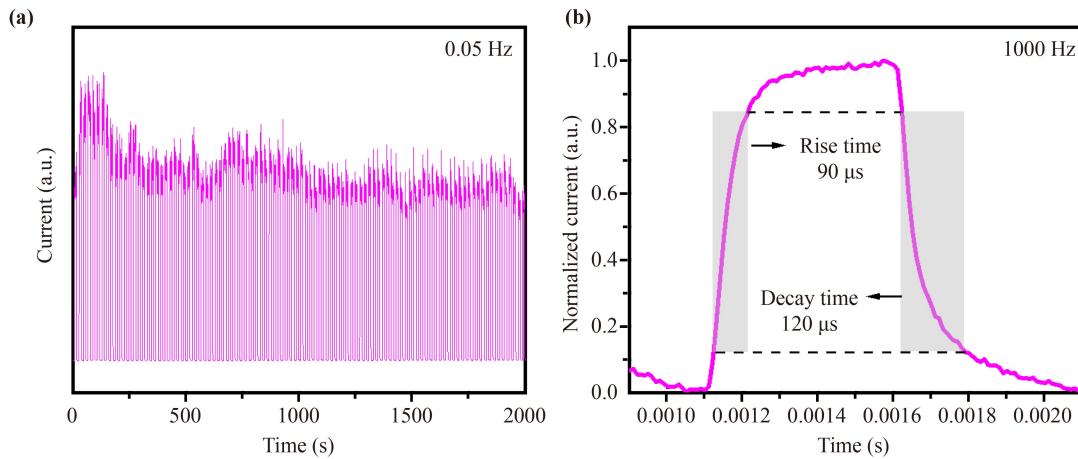


Fig. 4 (a) Long-cycle stability measurement of the few-layer $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ heterojunction photodetector. (b) Response speeds under a switching frequency of 1 kHz.

Evidently, the heterojunction device showed a strong response to visible (400–600 nm) and near-infrared (1100–1400 nm) wavelengths, corresponding to the bandgap of $\text{CdS}_{0.42}\text{Se}_{0.58}$ and MoTe_2 , respectively. These results indicated that excitons in both blocks contributed to the photocurrent. Figure 3(c) shows the I - V curves of the few-layer $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ flake heterojunction photodetector under different light intensities at 560 nm light illumination. Clearly, the photocurrent increased with increasing light power density at the same bias voltage, indicating that the number of photogenerated carriers was proportional to the absorbed photon flux. When the light power density was 0.413 mW/cm^2 , the photocurrent reached $1.11 \times 10^{-7} \text{ A}$ at 5 V bias, and the switching ratio of the device was calculated to be 1.15×10^4 under monochromatic light. Compared with white light illumination, the on/off ratio was reduced but still remained on the same order of magnitude. Figure 3(d) shows the relationship between the photocurrent and light intensity of the few-layer $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ photodetector at a bias of 1 V. The power law formula is as follows: $I_p = AP^\alpha$ [39], where I_p is the photocurrent, A is the proportional coefficient, P is the light power density, and exponent α is the fitting coefficient and reflects the trapping and recombination activity of photocarriers. The α value was determined to be 0.54 using the above fitting formula; this result indicated the existence of trap states in the contact interface, which affected the capture and recombination of photogenerated carriers [40].

To further investigate the performance of the few-layer $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ flake heterojunction photodetector, some key parameters of the photodetector were calculated. As is well known, R [41], EQE [42] and D^* [43] are three important parameters for the study of the photoelectric performance of the device, and these parameters can be calculated using the following formulae:

$$R = \frac{I_p - I_d}{P \cdot A}, \quad (1)$$

$$\text{EQE} = R \left(\frac{hc}{q\lambda} \right), \quad (2)$$

$$D^* = R \sqrt{\frac{A}{2qI_d}}, \quad (3)$$

where I_p represents the photocurrent, I_d represents the dark current, P represents the light power density, A is the device area, h is Planck's constant, c is the speed of light, q is the electron charge, and λ represents the incident wavelength. Figures 3(e) and (f) show the R , EQE and D^* of the few-layer $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ heterojunction photodetector at a 5 V bias. From Figs. 3(e) and (f), R , EQE and D^* decreased with increasing light power density, which was mainly attributed to the trap states being fully occupied by the excited photocarriers under high power density; this resulted in a reduced density of the available states and a decreased gain [44]. This phenomenon could also be observed in other researchers' works [45]. R , EQE and D^* had maximum values of 7221 A/W, $1.52 \times 10^6 \%$ and 1.67×10^{15} Jones when the power density was 0.054 mW/cm^2 at a 5 V bias, respectively. These key parameters are much better than those of most reported MoTe_2 -based photodetectors, indicating that our device possesses excellent photoelectric performance.

Periodic stability and fast response speed are important parameters of photodetectors. Figure 4 shows the I - t test diagram of the few-layer $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ heterojunction photodetector. Figure 4(a) is the I - t test of the $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ device under continuous switching cycles. Evidently, the on and off states show minimal change after 2000 s periodic illumination, indicating excellent stability and repeatability. Figure 4(b) shows the response speed diagram of the $\text{MoTe}_2/$

Table 1 Comparison of the key parameters of the $\text{CdS}_x\text{Se}_{1-x}$ and MoTe_2 -based photodetectors.

Device structure	Bias voltage (V)	R (A/W)	EQE (%)	D^* (Jones)	$I_{\text{light}}/I_{\text{dark}}$	Rise/Decay time (ms)	Ref.
MoTe_2 nanosheet	0	4×10^{-4}	–	1.07×10^8	–	0.043/0.043	[46]
$\text{MoTe}_2/\text{GeSe}$ sheet	–5	28.4	73.5	6.0×10^9	–	0.003/0.012	[47]
$\text{MoTe}_2/\text{Graphene}$	–1	853	1.4×10^4	1.59×10^{10}	–	0.012/0.005	[48]
$\text{CdS}_{0.49}\text{Se}_{0.51}/\text{CdS}$ NW	1	118	3.1×10^4	–	10^5	0.068/0.137	[49]
MoTe_2/Ge flake	–2	12460	–	3.3×10^{12}	–	5/5	[50]
$\text{CdS}_{0.14}\text{Se}_{0.86}$ flakes	5	703	1.94×10^5	3.41×10^{10}	–	39/39	[51]
$\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ flake	5	7221	1.52×10^6	1.67×10^{15}	1.73×10^4	0.09/0.12	This work

$\text{CdS}_{0.42}\text{Se}_{0.58}$ photodetector in single on and off states under a 5 V bias. The rise time is defined as the time needed for the photocurrent to increase from 10% to 90% of the maximum value during the rising process, while the fall time is defined as the time needed for the photocurrent to decrease from 90% to 10% during the falling process in a single switch periodicity. The rise and fall times of the $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ photodetector are as short as 90/120 μs , respectively. The rapid response speed of the $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ photodetector can be attributed to the good crystal quality of the few-layer MoTe_2 and $\text{CdS}_{0.42}\text{Se}_{0.58}$ materials, as well as the effective interface charge separation between them. Many studies have reported millisecond or even longer response times, which limits the practical applications of photodetectors. In our study, the response time of the $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ photodetector was reduced to just 90 μs , which was significantly lower than the reported values in many photodetector fields; thus, our $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ photodetector has great application prospects.

To facilitate comparison with other reported studies, we summarized the key parameters of our $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ devices and the reported MoTe_2 and $\text{CdS}_x\text{Se}_{1-x}$ based photodetectors; these are provided in Table 1. Compared with other MoTe_2 and $\text{CdS}_x\text{Se}_{1-x}$ devices [46–51], the $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ devices have superior optoelectronic performance, such as high R , EQE and D^* , with a faster response speed.

Figure 5(a) shows an optical image of the few-layer $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ heterojunction device, in which the golden yellow part is the interdigital electrode. The width of the device channel is 5 μm , and the device area of the $\text{MoTe}_2/\text{CdS}_{0.48}\text{Se}_{0.58}$ device is $\sim 1.65 \times 10^{-7} \text{ cm}^2$. To determine the generation and transmission mechanism of the photocurrent in the $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ device, we first investigated the photocurrent imaging of the device corresponding to 560 nm laser excitation at a bias of 5 V, as shown in Fig. 5(b), which corresponds to the green area in Fig. 5(a). From the photocurrent imaging results, the photocurrent is mainly distributed in the overlapping region of the MoTe_2 and $\text{CdS}_{0.42}\text{Se}_{0.58}$ flakes, indicating that photogenerated electron-hole pairs are effectively separated at the interface. This result indicates

that the heterojunction photodetector exhibits the same capability to separate photogenerated carriers as other vertical heterojunction devices. Next, we investigated the band alignment at the interface between MoTe_2 and $\text{CdS}_{0.42}\text{Se}_{0.58}$ based on first-principles calculations. Figure 5(c) shows the calculated projected local density of states (PLDOS) of the $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ heterojunction by using the Quantum ATK package. The calculated band gaps of $\text{CdS}_{0.42}\text{Se}_{0.58}$ and MoTe_2 are 2.18 eV and 0.96 eV, respectively; these are in good agreement with the experimental results. Under dark conditions, the electrons in $\text{CdS}_{0.42}\text{Se}_{0.58}$ diffuse to MoTe_2 driven by the band offset, while the holes in MoTe_2 cannot move to $\text{CdS}_{0.42}\text{Se}_{0.58}$ due to the large energy barrier at the valence band and accumulate at the interface. Due to the small band gap of MoTe_2 and the presence of a minority of electrons in MoTe_2 under dark conditions, there is a higher probability of recombination with the accumulated holes, consequently resulting in a lower dark current. Under illumination, $\text{CdS}_{0.42}\text{Se}_{0.58}$ and MoTe_2 absorb photons of different energies, producing a large number of photogenerated carriers. The electrons in $\text{CdS}_{0.42}\text{Se}_{0.58}$ move to MoTe_2 , and the holes in MoTe_2 can also easily transfer to $\text{CdS}_{0.42}\text{Se}_{0.58}$ because the accumulation of a large number of holes in the valence band leads to a decrease in the energy barrier height. Therefore, photogenerated carriers were effectively separated [see Fig. 5(c)]. The co-absorption and efficient separation of photogenerated carriers results in a higher photocurrent and responsivity of the $\text{CdS}_{0.42}\text{Se}_{0.58}/\text{MoTe}_2$ detector. Therefore, the device exhibits a higher $I_{\text{on}}/I_{\text{off}}$ ratio, higher responsivity, faster response speed, higher EQE and detectivity compared with the other reported MoTe_2 -based detectors.

The imaging capability of photodetectors plays a significant role in both civilian and military fields. To explore the actual potential of the few-layer $\text{MoTe}_2/\text{CdS}_{0.42}\text{Se}_{0.58}$ photodetector, the imaging capability of the device was investigated by an imaging system [see Fig. 6(a)]. The laser passed through the U-shaped mask and illuminated the device. After the semiconductor analyzer test and imaging computer control, the U-shaped pattern on the mask plate was finally successfully reproduced, as shown in Fig. 6(b). The uniform

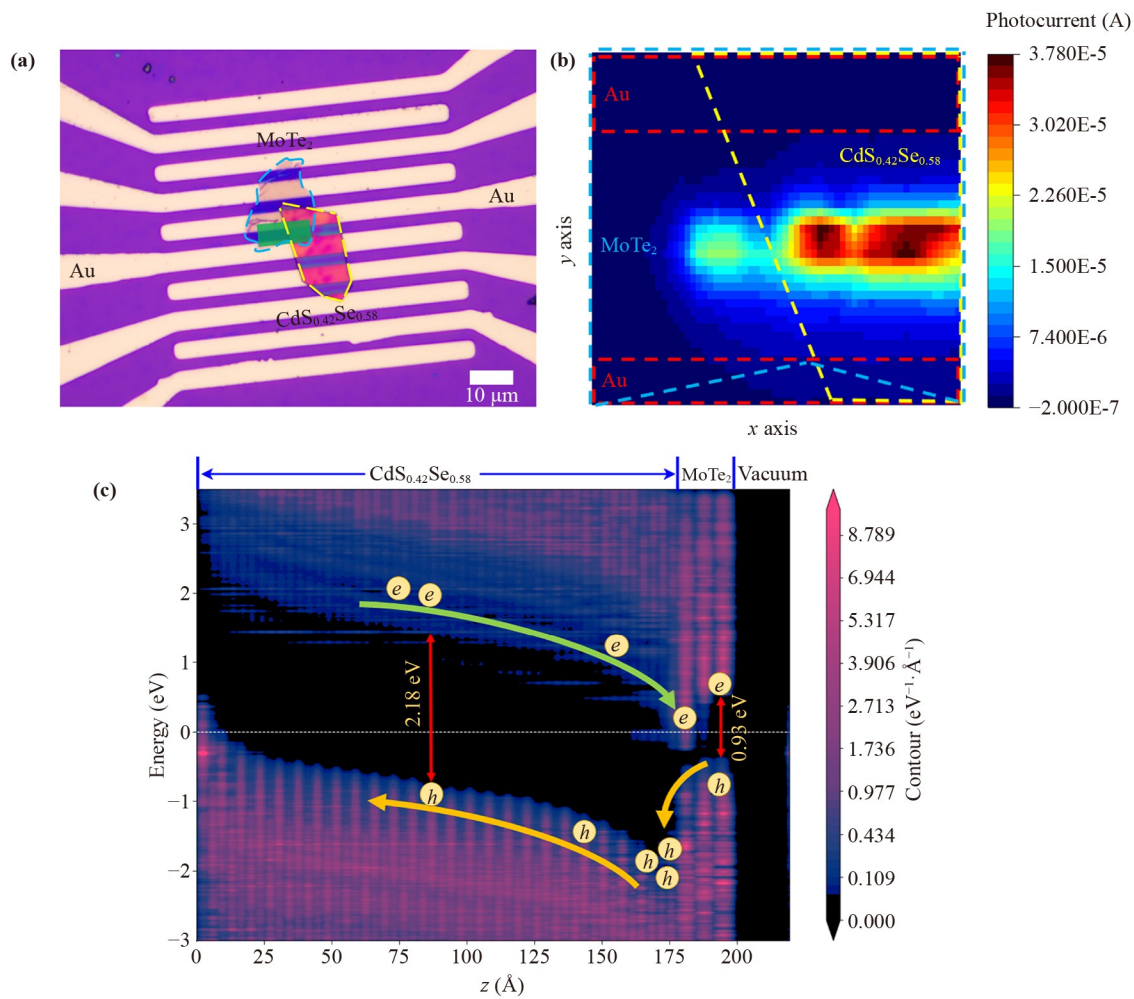


Fig. 5 (a) Optical microscope image and (b) photocurrent mapping image of MoTe₂/CdS_{0.42}Se_{0.58} heterojunction device. (c) Calculated energy band diagram of the MoTe₂/CdS_{0.42}Se_{0.58} heterojunction.

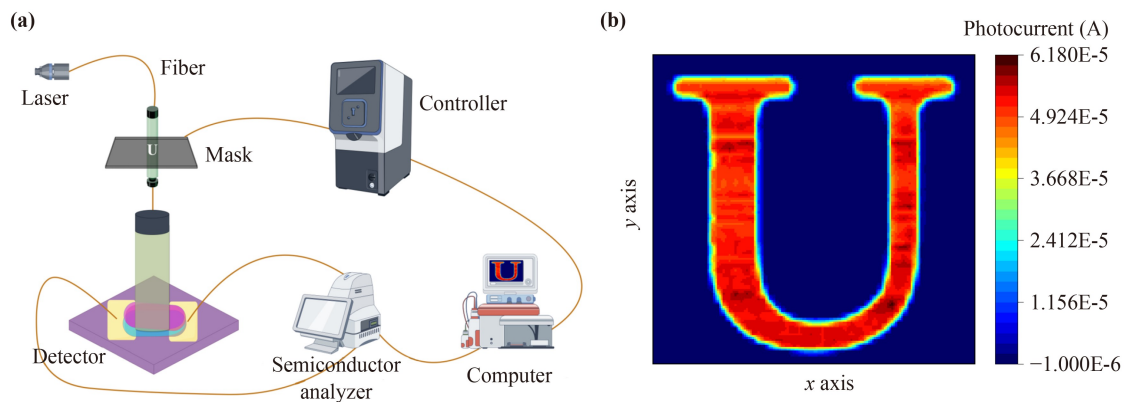


Fig. 6 (a) Schematic diagram of the imaging measurement system. (b) Imaging results of the “U” letter under visible light.

photocurrent was distributed in the U-shaped pattern, indicating that the fabricated photodetector had excellent visible light imaging capability and confirming that the few-layer MoTe₂/CdS_{0.42}Se_{0.58} photodetector had unique advantages in the imaging field.

4 Conclusion

In summary, a few-layer MoTe₂/CdS_{0.42}Se_{0.58} flake heterojunction photodetector was first fabricated by mechanical exfoliated and CVD methods combined with



a typical dry transfer technique. The MoTe₂/CdS_{0.42}Se_{0.58} photodetector exhibited excellent photoelectric performance, with a higher switching ratio of 1.73×10^4 , a higher responsivity of 7221 A/W, a higher EQE of 1.52×10^6 %, a larger detectivity of 1.67×10^{15} Jones and a faster response speed (rise/fall time of 90/120 μs); based on these results, our photodetector was superior to other MoTe₂-based photodetectors. The first-principles calculations indicated that the excellent performance of our heterojunction photodetector was attributed to the unique band arrangement at the interface. Finally, the visible light imaging function was successfully achieved on the MoTe₂/CdS_{0.42}Se_{0.58} photodetector devices. Based on these results, our few-layer MoTe₂/CdS_{0.42}Se_{0.58} flake photodetectors have excellent application prospects in optoelectronic devices.

Declarations The authors declare that they have no competing interests and there are no conflicts.

Acknowledgements This work was supported by the National Natural Science Foundation of China (Nos. 11864046 and 11764046), the Basic Research Program of Yunnan Province (Nos. 202001AT070064 and 202101AT070124), the Spring City Plan (High-level Talent Promotion and Training Project of Kunming) (No. 2022SCP005), and Yunnan Expert Workstation (No. 202205AF150008).

References

1. K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov, Electric field effect in atomically thin carbon films, *Science* 306(5696), 666 (2004)
2. J. Yao, Z. Zheng, and G. Yang, All-layered 2D optoelectronics: A high-performance UV-Vis-NIR broadband SnSe photodetector with Bi₂Te₃ topological insulator electrodes, *Adv. Funct. Mater.* 27(33), 1701823 (2017)
3. A. Castellanos-Gomez, M. Poot, G. A. Steele, H. S. J. van der Zant, N. Agrait, and G. Rubio-Bollinger, Elastic properties of freely suspended MoS₂ nanosheets, *Adv. Mater.* 24(6), 772 (2012)
4. J. Yao and G. Yang, Flexible and high-performance all-2D photodetector for wearable devices, *Small* 14(21), 1704524 (2018)
5. W. Wu, L. Wang, Y. Li, F. Zhang, L. Lin, S. Niu, D. Chenet, X. Zhang, Y. Hao, T. F. Heinz, J. Hone, and Z. L. Wang, Piezoelectricity of single-atomic-layer MoS₂ for energy conversion and piezotronics, *Nature* 514(7523), 470 (2014)
6. S. Mouri, Y. Miyauchi, and K. Matsuda, Tunable photoluminescence of monolayer MoS₂ via chemical doping, *Nano Lett.* 13(12), 5944 (2013)
7. C. J. Heard, J. Čejka, M. Opanasenko, P. Nachtigall, G. Centi, and S. Perathoner, 2D oxide nanomaterials to address the energy transition and catalysis, *Adv. Mater.* 31(3), 1801712 (2019)
8. L. Tang, X. Meng, D. Deng, and X. Bao, Confinement catalysis with 2D materials for energy conversion, *Adv. Mater.* 31(50), 1901996 (2019)
9. F. R. Fan, R. Wang, H. Zhang, and W. Wu, Emerging beyond-graphene elemental 2D materials for energy and catalysis applications, *Chem. Soc. Rev.* 50(19), 10983 (2021)
10. L. X. Chen, Z. W. Chen, M. Jiang, Z. Lu, C. Gao, G. Cai, and C. V. Singh, Insights on the dual role of two-dimensional materials as catalysts and supports for energy and environmental catalysis, *J. Mater. Chem. A* 9(4), 2018 (2021)
11. P. Tao, S. Yao, F. Liu, B. Wang, F. Huang, and M. Wang, Recent advances in exfoliation techniques of layered and non-layered materials for energy conversion and storage, *J. Mater. Chem. A* 7(41), 23512 (2019)
12. Y. Tian, Y. Chen, Y. Liu, H. Li, and Z. Dai, Elemental two-dimensional materials for Li/Na-ion battery anode applications, *Chem. Rec.* 22(10), e202200123 (2022)
13. B. Li, H. Xu, Y. Ma, and S. Yang, Harnessing the unique properties of 2D materials for advanced lithium-sulfur batteries, *Nanoscale Horiz.* 4(1), 77 (2019)
14. N. Deng, Y. Feng, G. Wang, X. Wang, L. Wang, Q. Li, L. Zhang, W. Kang, B. Cheng, and Y. Liu, Rational structure designs of 2D materials and their applications toward advanced lithium-sulfur battery and lithium-selenium battery, *Chem. Eng. J.* 401(20), 125976 (2020)
15. G. Gao, F. Zheng, F. Pan, and L. W. Wang, Theoretical investigation of 2D conductive microporous coordination polymers as Li-S battery cathode with ultrahigh energy density, *Adv. Energy Mater.* 8(25), 1801823 (2018)
16. R. Rojasee and R. Shahbazian-Yassar, Two-dimensional materials to address the lithium battery challenges, *ACS Nano* 14(3), 2628 (2020)
17. A. Dodda, D. Jayachandran, A. Pannone, N. Trainor, S. P. Stepanoff, M. A. Steves, S. S. Radhakrishnan, S. Bachu, C. W. Ordonez, J. R. Shallenberger, J. M. Redwing, K. L. Knappenberger, D. E. Wolfe, and S. Das, Active pixel sensor matrix based on monolayer MoS₂ phototransistor array, *Nat. Mater.* 21(12), 1379 (2022)
18. U. Khan, Y. Luo, L. Tang, C. Teng, J. Liu, B. Liu, and H. M. Cheng, Controlled vapor-solid deposition of millimeter-size single crystal 2D Bi₂O₂Se for high-performance phototransistors, *Adv. Funct. Mater.* 29(14), 1807979 (2019)
19. S. G. Kim, S. H. Kim, J. Park, G. S. Kim, J. H. Park, K. C. Saraswat, J. Kim, and H. Y. Yu, Infrared detectable MoS₂ phototransistor and its application to artificial multilevel optic-neural synapse, *ACS Nano* 13(9), 10294 (2019)
20. S. Namgung, J. Shaver, S. H. Oh, and S. J. Koester, Multimodal photodiode and phototransistor device based on two-dimensional materials, *ACS Nano* 10(11), 10500 (2016)
21. Y. Cai, J. Yang, F. Wang, S. Li, Y. Wang, X. Zhan, F. Wang, R. Cheng, Z. Wang, and J. He, Ultrasensitive solar-blind ultraviolet detection and optoelectronic neuromorphic computing using α-In₂Se₃ phototransistors, *Front. Phys.* 18(3), 33308 (2023)

22. Z. Q. Li, T. T. Yan, and X. S. Fang, Low-dimensional wide-bandgap semiconductors for UV photodetectors, *Nat. Rev. Mater.* 8(9), 587 (2023)
23. H. Zeng, Y. Wen, L. Yin, R. Cheng, H. Wang, C. Liu, and J. He, Recent developments in CVD growth and applications of 2D transition metal dichalcogenides, *Front. Phys.* 18(5), 53603 (2023)
24. G. Zhang, Y. Shu, J. Yao, Q. Shu, H. Deng, G. Jia, and Z. Wang, Characteristics and developments of quantum-dot infrared photodetectors, *Front. Phys. China* 1(3), 334 (2006)
25. K. He, J. Zhu, Z. Li, Z. Chen, H. Zhang, C. Liu, X. Zhang, S. Wang, P. Zhao, Y. Zhou, S. Zhang, Y. Yin, X. Zheng, W. Huang, and L. Wang, High-sensitive two-dimensional PbI₂ photodetector with ultrashort channel, *Front. Phys.* 18(6), 63305 (2023)
26. C. H. An, F. Nie, R. Zhang, X. Ma, D. Wu, Y. Sun, X. Hu, D. Sun, L. Pan, and J. Liu, Two-dimensional material-enhanced flexible and self-healable photodetector for large-area photodetection, *Adv. Funct. Mater.* 31(22), 2100136 (2021)
27. R. Maiti, C. Patil, M. A. S. R. Saadi, T. Xie, J. G. Azadani, B. Uluutku, R. Amin, A. F. Briggs, M. Miscuglio, D. Van Thourhout, S. D. Solares, T. Low, R. Agarwal, S. R. Bank, and V. J. Sorger, Strain-engineered high-responsivity MoTe₂ photodetector for silicon photonic integrated circuits, *Nat. Photonics* 14(9), 578 (2020)
28. T. Octon, V. Nagareddy, S. Russo, M. F. Craciun, and C. D. Wright, Fast high-responsivity few-layer MoTe₂ photodetectors, *Adv. Opt. Mater.* 4(11), 1750 (2016)
29. L. Yin, X. Zhan, K. Xu, F. Wang, Z. Wang, Y. Huang, Q. Wang, C. Jiang, and J. He, Ultrahigh sensitive MoTe₂ phototransistors driven by carrier tunneling, *Appl. Phys. Lett.* 108(4), 043503 (2016)
30. H. Huang, J. Wang, W. Hu, L. Liao, P. Wang, X. Wang, F. Gong, Y. Chen, G. Wu, W. Luo, H. Shen, T. Lin, J. Sun, X. Meng, X. Chen, and J. Chu, Highly sensitive visible to infrared MoTe₂ photodetectors enhanced by the photogating effect, *Nanotechnology* 27(44), 445201 (2016)
31. Y. Chen, X. Wang, G. Wu, Z. Wang, H. Fang, T. Lin, S. Sun, H. Shen, W. Hu, J. Wang, J. Sun, X. Meng, and J. Chu, High-performance photovoltaic detector based on MoTe₂/MoS₂ van der Waals heterostructure, *Small* 14(9), 1703293 (2018)
32. M. Y. Lu, Y. T. Chang, and H. J. Chen, Efficient self-driven photodetectors featuring a mixed-dimensional van der Waals heterojunction formed from a CdS nanowire and a MoTe₂ flake, *Small* 14(40), 1802302 (2018)
33. J. Chen, Y. Shan, Q. Wang, J. Zhu, and R. Liu, P-type laser-doped WSe₂/MoTe₂ van der Waals heterostructure photodetector, *Nanotechnology* 31(29), 295201 (2020)
34. Z. Lu, Y. Xu, Y. Yu, K. Xu, J. Mao, G. Xu, Y. Ma, D. Wu, and J. Jie, Ultrahigh speed and broadband few-layer MoTe₂/Si 2D-3D heterojunction-based photodiodes fabricated by pulsed laser deposition, *Adv. Funct. Mater.* 30(9), 1907951 (2020)
35. R. O. Loutfy and D. S. Ng, Electrodeposited polycrystalline thin films of cadmium chalcogenides for backwall photo-electrochemical cells, *Solar Energy Mater.* 11(4), 319 (1984)
36. S. Smidstrup, D. Stradi, J. Wellendorff, P. A. Khomyakov, U. G. Vej-Hansen, M. E. Lee, T. Ghosh, E. Jónsson, H. Jónsson, and K. Stokbro, First-principles green's-function method for surface calculations: A pseudopotential localized basis set approach, *Phys. Rev. B* 96(19), 195309 (2017)
37. S. Grimme, Semiempirical GGA-type density functional constructed with a long-range dispersion correction, *J. Comput. Chem.* 27(15), 1787 (2006)
38. M. A. Hossain, J. R. Jennings, N. Mathews, and Q. Wang, Band engineered ternary solid solution CdS_xSe_{1-x} sensitized mesoscopic TiO₂ solar cells, *Phys. Chem. Chem. Phys.* 14(19), 7154 (2012)
39. S. Yang, S. Tongay, Y. Li, Q. Yue, J. B. Xia, S. S. Li, J. Li, and S. H. Wei, Layer-dependent electrical and optoelectronic responses of ReSe₂ nanosheet transistors, *Nanoscale* 6(13), 7226 (2014)
40. G. Chen, Z. Liu, B. Liang, G. Yu, Z. Xie, H. Huang, B. Liu, X. Wang, D. Chen, M. Q. Zhu, and G. Shen, Single-crystalline p-type Zn₃As₂ nanowires for field-effect transistors and visible-light photodetectors on rigid and flexible substrates, *Adv. Funct. Mater.* 23(21), 2681 (2013)
41. Y. H. Chen, L. X. Su, M. M. Jiang, and X. S. Fang, Switch type PANI/ZnO core-shell microwire heterojunction for UV photodetection, *J. Mater. Sci. Technol.* 105, 259 (2022)
42. S. Y. Li, Y. Zhang, W. Yang, H. Liu, and X. Fang, 2D Perovskite Sr₂Nb₃O₁₀ for High-Performance UV Photodetectors, *Adv. Mater.* 32(7), 1905443 (2020)
43. Y. Zhang, J. Yao, Z. Zhang, R. Zhang, L. Li, Y. Teng, Z. J. Shen, L. X. Kang, L. M. Wu, and X. S. Fang, Two-dimensional perovskite Pb₂Nb₃O₁₀ photodetectors, *J. Mater. Sci. Technol.* 164, 95 (2023)
44. B. X. Liu, Y. Sun, Y. Wu, K. Liu, H. Ye, F. Li, L. Zhang, Y. Jiang, and R. Wang, Enhanced photoresponse of TiO₂/MoS₂ heterostructure phototransistors by the coupling of interface charge transfer and photogating, *Nano Res.* 14(4), 982 (2021)
45. J. Yan, X. Yang, X. Liu, C. Du, F. Qin, M. Yang, Z. Zheng, and J. Li, Van der Waals heterostructures with built-in Mie resonances for polarization-sensitive photodetection, *Adv. Sci.* 10(9), 2207022 (2023)
46. J. Lai, X. Liu, J. Ma, Q. Wang, K. Zhang, X. Ren, Y. Liu, Q. Gu, X. Zhuo, W. Lu, Y. Wu, Y. Li, J. Feng, S. Zhou, J. H. Chen, and D. Sun, Anisotropic broadband photoresponse of layered type-II Weyl semimetal MoTe₂, *Adv. Mater.* 30(22), 1707152 (2018)
47. P. Chen, L. Pi, Z. Li, H. Wang, X. Xu, D. Li, X. Zhou, and T. Zhai, GeSe/MoTe₂ vdW heterostructure for UV-VIS-NIR photodetector with fast response, *Appl. Phys. Lett.* 121(2), 021103 (2022)
48. J. W. Park, Y. S. Jung, S. H. Park, H. J. Choi, and Y. S. Cho, High-performance photoresponse in ferroelectric d1T-MoTe₂-based vertical photodetectors, *Adv. Opt. Mater.* 10(21), 2200898 (2022)
49. P. Guo, J. Xu, K. Gong, X. Shen, Y. Lu, Y. Qiu, J. Xu, Z. Zou, C. Wang, H. Yan, Y. Luo, A. Pan, H. Zhang, J. C. Ho, and K. M. Yu, On-nanowire axial heterojunction design for high-performance photodetectors, *ACS Nano*



- 10(9), 8474 (2016)
50. W. Chen, R. Liang, S. Zhang, Y. Liu, W. Cheng, C. Sun, and J. Xu, Ultrahigh sensitive near-infrared photodetectors based on MoTe_2 /germanium heterostructure, *Nano Res.* 13(1), 127 (2020)
51. J. Xia, Y. X. Zhao, L. Wang, X. Z. Li, Y. Y. Gu, H. Q. Cheng, and X. M. Meng, Van der Waals epitaxial two-dimensional $\text{CdS}_x\text{Se}_{1-x}$ semiconductor alloys with tunable-composition and application to flexible optoelectronics, *Nanoscale* 9(36), 13786 (2017)