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A review of molybdenum disulfide (MoS₂) based photodetectors: from ultra-broadband, self-powered to flexible devices

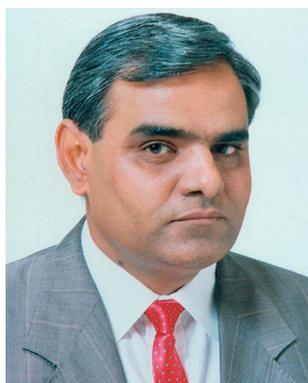
 Hari Singh Nalwa 

Two-dimensional transition metal dichalcogenides (2D TMDs) have attracted much attention in the field of optoelectronics due to their tunable bandgaps, strong interaction with light and tremendous capability for developing diverse van der Waals heterostructures (vdWHs) with other materials. Molybdenum disulfide (MoS₂) atomic layers which exhibit high carrier mobility and optical transparency are very suitable for developing ultra-broadband photodetectors to be used from surveillance and healthcare to optical communication. This review provides a brief introduction to TMD-based photodetectors, exclusively focused on MoS₂-based photodetectors. The current research advances show that the photoresponse of atomic layered MoS₂ can be significantly improved by boosting its charge carrier mobility and incident light absorption *via* forming MoS₂ based plasmonic nanostructures, halide perovskites–MoS₂ heterostructures, 2D–0D MoS₂/quantum dots (QDs) and 2D–2D MoS₂ hybrid vdWHs, chemical doping, and surface functionalization of MoS₂ atomic layers. By utilizing these different integration strategies, MoS₂ hybrid heterostructure-based photodetectors exhibited remarkably high photoresponsivity ranging from mA W⁻¹ up to 10¹⁰ A W⁻¹, detectivity from 10⁷ to 10¹⁵ Jones and a photoresponse time from seconds (s) to nanoseconds (10⁻⁹ s), varying by several orders of magnitude from deep-ultraviolet (DUV) to the long-wavelength infrared (LWIR) region. The flexible photodetectors developed from MoS₂-based hybrid heterostructures with graphene, carbon nanotubes (CNTs), TMDs, and ZnO are also discussed. In addition, strain-induced and self-powered MoS₂ based photodetectors have also been summarized. The factors affecting the figure of merit of a very wide range of MoS₂-based photodetectors have been analyzed in terms of their photoresponsivity, detectivity, response speed, and quantum efficiency along with their measurement wavelengths and incident laser power densities. Conclusions and the future direction are also outlined on the development of MoS₂ and other 2D TMD-based photodetectors.

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Dr Hari Singh Nalwa is a distinguished independent scientist and scholar working in the fields of nanotechnology and materials science. Dr Nalwa has authored more than 170 scientific articles, 26 book chapters, and 125 volumes of scientific books, as well as 18 patents in cross-disciplinary research areas of nanotechnology, materials science and polymer science. Dr Nalwa's research interests include ferroelectric polymers, conducting polymers, organic nonlinear optical materials for integrated optics, low- and high-dielectric constant materials for microelectronics packaging, 3D printing, two-dimensional (2D) nanomaterial-based bulk heterojunction and dye-sensitized solar cells, and multifunctional sensors for wearable technology. He received the "Award of Excellence" from the Association of American Publishers for the "Handbook of Nanostructured Materials and Nanotechnology," a 5-volume set (Academic Press, 2000), and "Best Reference Work Award" from the American Society for Engineering Education for "The Encyclopedia of Nanoscience and Nanotechnology," a 10-volume set (American Scientific Publishers, 2004). He is Founder, President, and Chief Executive Officer (CEO) of American Scientific Publishers (<http://www.aspbs.com>), which he established in 2000.



1. Introduction

Advances in the fields of electronics, optoelectronics and photonics have created a great demand for new functional materials that possess ease of synthesis, processing and fabrication and enable desired tailoring of the physical and chemical properties by chemical functionalization and/or formation of hybrid structures for potential applications in electronic and optoelectronic devices.^{1,2} In this context, earth-abundant two-dimensional (2D) nanomaterials, including graphene^{3–12} and transition metal dichalcogenides (TMDs),^{13–20} have emerged as novel functional materials of choice due to their low-cost production, easy processing and easy deposition on different substrates with precise thickness control of the atomic layers *via* mechanical and chemical exfoliation, chemical vapor deposition (CVD) and atomic layer etching (ALE) methods. Flexible graphene nanosheets have been explored for developing wearable and portable devices, including field-effect transistors (FETs),^{21,22} sensors,^{23–25} supercapacitors,^{26–28} lithium-ion batteries,²⁹ triboelectric nanogenerators,³⁰ solar cells^{31–33} and photodetectors.^{34,35} Following the research progress of graphene, a similar wide range of applications for flexible atomic layered TMDs have been anticipated and are now slowly emerging, from wearable electronics to optoelectronics.^{36–40} Flexible photodetectors are becoming a key component of optoelectronic technology for a wide range of applications in the fields of surveillance, soft robotics, sensors for wearable and portable healthcare and sports, light-emitting diodes (LEDs), high-speed optical communication, and biomedical imaging.^{34,35,41} Flexible photodetectors are gaining much attention for use in wearable optoelectronics, for which many different types of nanostructures, such as atomic layered nanosheets, nanowires (NWs), fibers, quantum dots and 3D networks of inorganic and organic materials and their nano-hybrids, have been studied. These nanostructures have been used as phototransistors, photoconductors and photodiodes for developing photodetectors from a wide variety of nanomaterials, including silicon (Si) and germanium (Ge),^{42,43} selenium (Se),⁴⁴ GaP and InP,^{45,46} CdS,^{47,48} ZnSe,⁴⁹ ZnO and its hybrids with PbS, ZnS, CdO, gold (Au) and polymers,^{50–55} ZnGa₂O₄,⁵⁶ Zn₂GeO₄ and In₂Ge₂O₇,⁵⁷ CuInSe₂,^{58,59} In₂S₃,⁶⁰ In₂Se₃,⁶¹ Sb₂S₃,⁶² Sb₂Se₃,⁶³ Bi₂S₃,⁶⁴ SnS,⁶⁵ SnS₂,⁶⁶ ZrS₃,⁶⁷ Zn₃P₂,⁶⁸ PbI₂,⁶⁹ MoO₃,⁷⁰ GaS,⁷¹ SnO₂,⁷² ZnTe,⁷³ GaTe,⁷⁴ perovskites,^{75,76} their hybrid composites with ZnO, gold, poly(diketopyrrolopyrrole-terthiophene) (PDPP3T) conjugated polymer, and graphene,^{77–80} polythiophene,⁸¹ carbon nanotubes (CNTs),⁸² graphene nanocomposites with CNTs⁸³ and with ZnO quantum dots,⁸⁴ transition-metal trichalcogenides (MX₃, where M represents a transition metal, Ti, Zr, Hf, Nb, or Ta, and X is a chalcogen, S, Se, or Te), *e.g.*, hafnium trisulfide (HfS₃),⁸⁵ HfS₃ and HfSe₃ nanocomposites with graphene,⁸⁶ and graphene-based materials.^{87–89} The important parameters including photoresponsivity, specific detectivity, noise equivalent power (NEP), photogain, external quantum efficiency (EQE), linear dynamic range (LDR), and response speed of photodetectors have already been well defined in the literature;¹ here, these parameters are discussed in reference to the nanomaterial-based photodetectors and especially for the MoS₂ atomic layer-based hybrid photodetectors.

Many research articles have been published on atomically thin layered MoS₂ based photodetectors, however, a comprehensive review summarizing the recent developments in MoS₂ photodetectors is completely lacking in the scientific literature. This review briefly introduces TMDs, including the applications of MoS₂ atomic layers in developing photodetectors. The tuning of optoelectronic properties by boosting the carrier mobility of and incident light absorption by MoS₂ atomic layers through the use of plasmonic and halide perovskite–MoS₂ hybrid heterostructures, 2D–0D and 2D–2D MoS₂ heterostructures, interface coupling effect (ICE), or chemical doping of MoS₂ films is discussed in order to evaluate the performance of MoS₂ photodetectors from the perspective of their based phototransistors, photoconductors and photodiode components. Particular emphasis is placed on atomic layered MoS₂-based ultra-broadband photodetectors, from their fundamental development to self-powered to flexible photodetectors for wearable optoelectronics. The performance of pristine MoS₂ atomic layers and MoS₂ hybrid heterostructures with graphene, CNTs, TMDs, ZnO and surface functionalized MoS₂ atomic layers for developing flexible photodetectors is discussed in terms of their broadband photoresponsivity, detectivity, NEP, photogain, EQE, photoresponse speed, mechanical flexibility and environmental stability. Strain-induced and self-powered MoS₂ based hybrid photodetectors has also been summarized. Finally, the challenges in developing flexible photodetectors from TMDs are analyzed. This review should be a useful source for and inspire a wide range of audience, including researchers working in the fields of optoelectronics, sensors, materials science, nanotechnology, physics, electrical engineering, and communications.

2. Molybdenum disulfide (MoS₂) for photodetectors

Fig. 1 shows the wide range of the electromagnetic spectrum covered by 2D nanomaterials, from the near-infrared (NIR) and mid-IR (MIR) to the far-IR (FIR), the related applications of these nanomaterials in electronics, optoelectronics and photonics, and the atomic and band structures of 2D materials, including hexagonal boron nitride (h-BN), molybdenum disulfide (MoS₂), black phosphorus (BP) and graphene.⁹⁰ h-BN is an insulator with a large bandgap of 6.0 eV,^{91–95} whereas MoS₂^{96–98} and BP^{99–101} are semiconductors with sizeable bandgaps that vary with the number of atomic layers. Graphene is a zero bandgap semimetal^{102,103} and monolayer graphene exhibits 97.7% optical transparency independent of the optical wavelength in the 450–800 nm spectral region; the transparency decreases proportionally with an increasing number of graphene layers.¹⁰⁴ Monolayer graphene absorbs only 2.3% of incident white light; therefore, the photoresponsivity of monolayer (1L) graphene is rather limited due to the low absorbance in the visible region. The insulating layers of an h-BN dielectric have been used along with other 2D materials, such as graphene, BP and TMDs, including MoS₂, MoSe₂, WS₂, WSe₂, MoTe₂, *etc.*, to develop electronic and optoelectronic devices.^{105–109} Though graphene-based



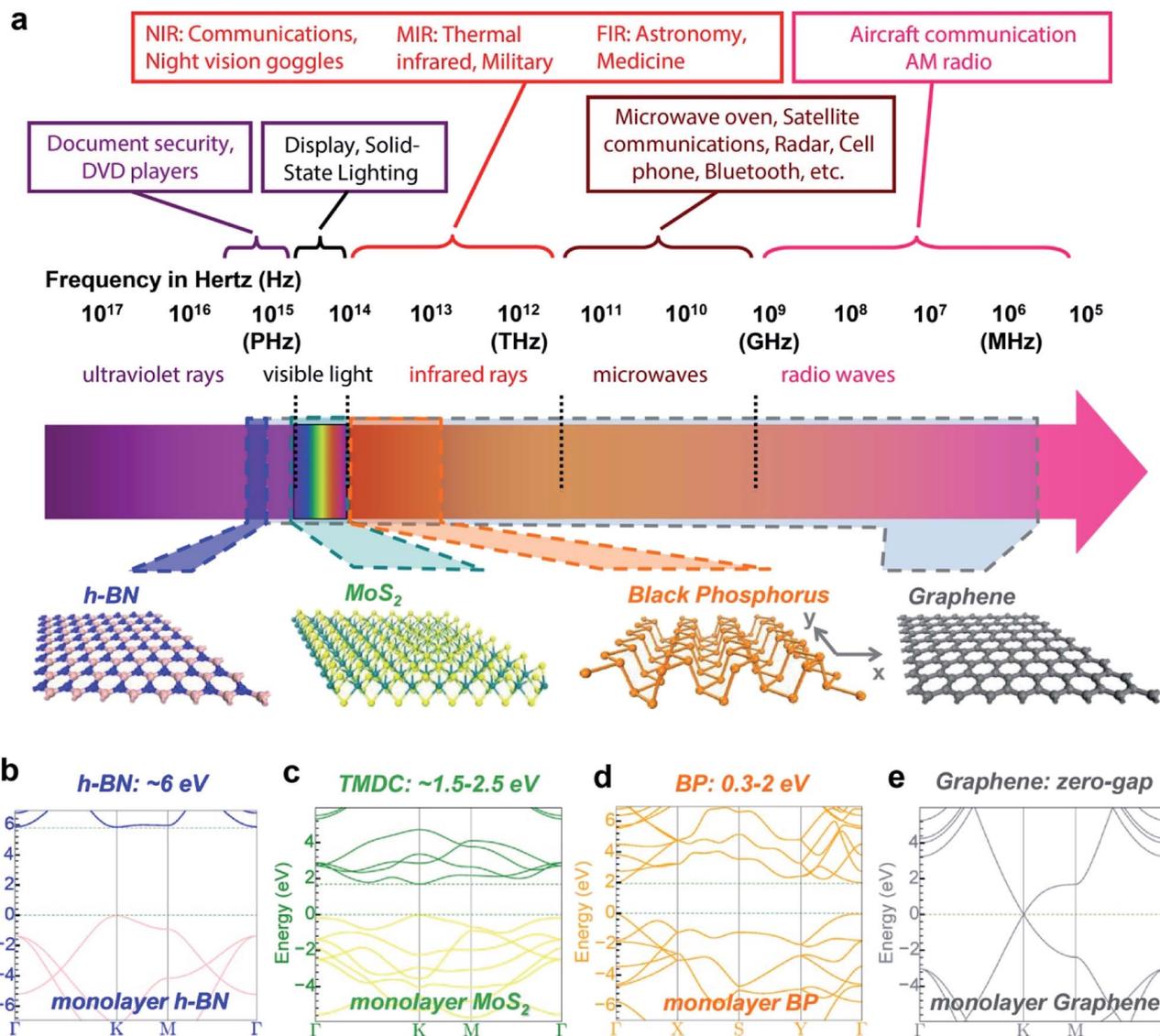


Fig. 1 (a) 2D materials covering a very wide range of the electromagnetic spectrum, from the NIR and MIR to the FIR, and their corresponding applications. The bottom section shows the atomic structures of h-BN, MoS₂, black phosphorus (BP) and graphene from left to right. The electromagnetic spectral ranges covered by different 2D materials are depicted using colored polygons. (b–e) Band structures and bandgaps of monolayer h-BN (b), MoS₂ (c), BP (d) and gapless graphene (e). Reprinted with permission from ref. 90, copyright © 2014 Macmillan Publishers Limited.

materials have been extensively studied for use in photodetectors because of their high carrier mobility and strong interaction with light over a broad spectral range,^{110–113} graphene-based photodetectors suffer from low photoresponsivity due to graphene's weak optical absorption.^{104,114} Broadband photodetectors utilizing multilayer BP operating over the 532 nm to 3.39 μm wavelength range have been fabricated by Guo *et al.*¹¹⁵ Comparatively, TMDs have a large electronic density of states, giving rise to high optical absorption and ultrafast charge transfer, making them more suitable for photodetectors. Bernardi *et al.*^{116a} reported 5–10% incident sunlight absorption by MoS₂, MoSe₂, and WS₂ monolayers for a thickness of >1 nm, which is higher than the value of 2.3% exhibited by monolayer graphene¹⁰⁴ and one

order of magnitude higher than those of thin films of the conventional semiconductors GaAs and Si, generally used for solar energy applications. Fig. 2(a) compares the computed and measured absorbance of monolayer (1L) MoS₂. The computed absorbance of 1L MoS₂ was obtained using density functional theory (DFT) calculations, GW method and the Bethe–Salpeter equation (BSE) while experimental absorbance was reported by Mak *et al.*⁹⁶ The quantitative agreement was observed between the computed and experimentally measured absorbance of 1L MoS₂. Fig. 2(b) compares the optical absorbance of MoS₂, MoSe₂, and WS₂ monolayers with that of graphene, clearly showing that TMD monolayers absorb much more sunlight than graphene. The calculated flux of absorbed photons was 4.6, 3.9, 2.3, and 2.0 mA cm^{-2} for monolayer



MoSe₂, MoS₂, WS₂ and graphene, respectively, compared with 0.3 mA cm⁻² for GaAs and 0.1 mA cm⁻² for Si in the form of 1 nm thin films. A monolayer TMD can absorb as much sunlight as a 50 nm thick Si film. Wang *et al.*^{116b} summarized the band-gaps of different atomic layered 2D nanomaterials including MoS₂, MoSe₂, MoTe₂, WS₂, WSe₂, ReS₂, ReSe₂, SnS₂, SnSe₂, HfS₂, HfSe₂, ZrS₂, ZrSe₂, In₂Se₃, black AsP, black phosphorus (BP), and h-BN with their corresponding photo-detection range, varying from near ultraviolet (NUV) to long infrared (LIR) as shown in Fig. 2(c). MoS₂, WSe₂, SnS₂, and black phosphorus show a broadband spectral range from NUV

wavelength to mid-infrared (MIR) wavelength. The optical absorbance of MoS₂ thin films can be further extended by developing diverse hybrid heterostructures.

The figure-of-merit for a photodetector is generally evaluated in terms of their photoresponsivity (*R*), specific detectivity (*D**), noise equivalent power (NEP), and the photoresponse time. The photoresponsivity is an electrical response to an incident light illumination and detectivity is associated with the lowest required optical power intensity for a photodetector to differentiate signal from the actual noise. The intensities of dark and photo currents generated as a function of applied bias voltage

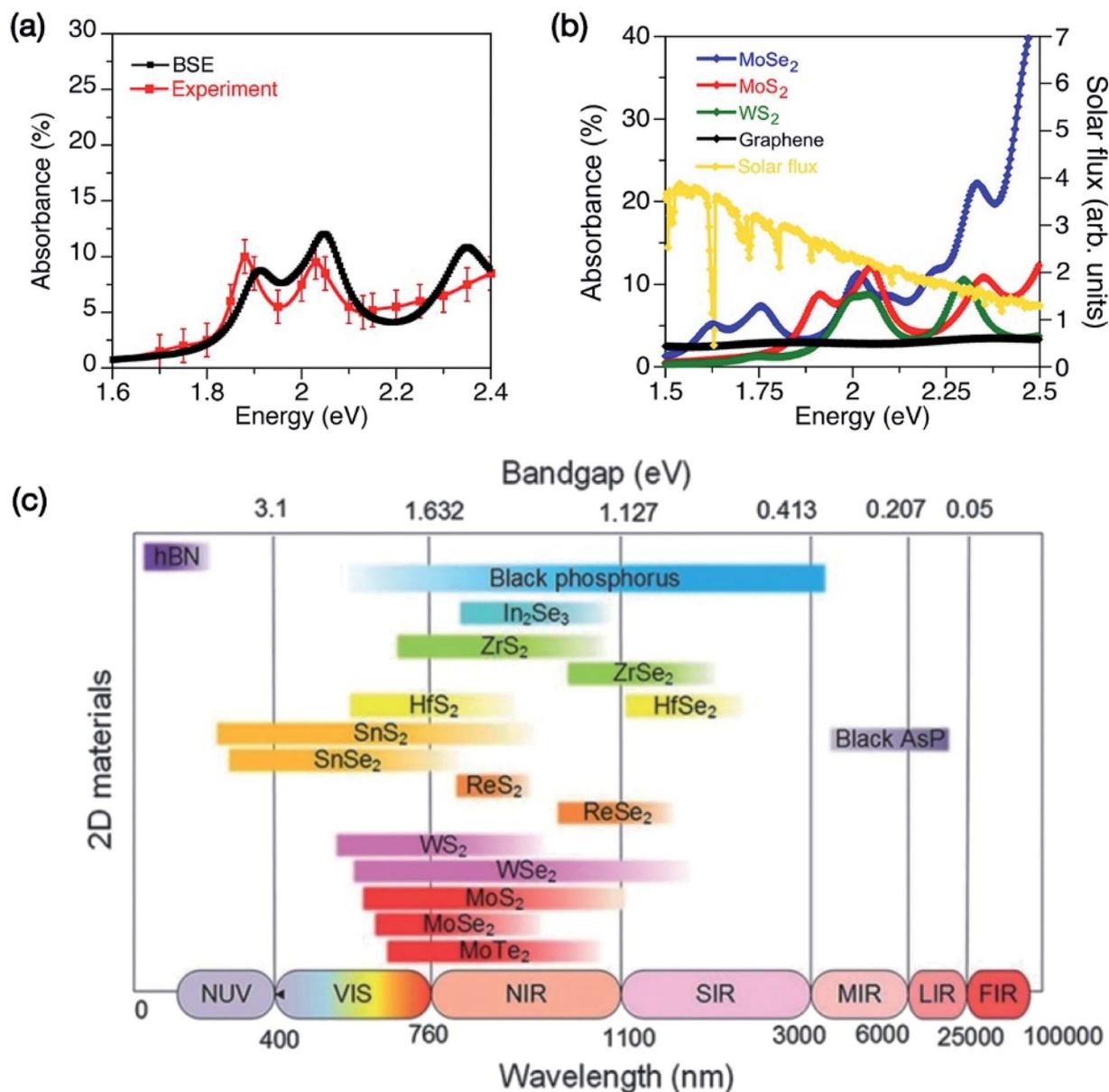


Fig. 2 (a) A comparison of the computed absorbance obtained by Bethe–Salpeter equation (BSE) with experimentally measured absorbance of a MoS₂ monolayer. (b) A comparison of the optical absorbance of MoS₂, MoSe₂, and WS₂ monolayers with that of graphene along with the incident AM1.5G solar flux. Reprinted with permission from ref. 116a, copyright © American Chemical Society. (c) Band-gaps of different atomic layered 2D nanomaterials (MoS₂, MoSe₂, MoTe₂, WS₂, WSe₂, ReS₂, ReSe₂, SnS₂, SnSe₂, HfS₂, HfSe₂, ZrS₂, ZrSe₂, In₂Se₃, black AsP, black phosphorus (BP) and h-BN) with their corresponding photodetection range varying from near ultraviolet (NUV) to long infrared (LIR). Reprinted with permission from ref. 116b, copyright © Wiley.



and the values of photoresponsivity and detectivity at different operation wavelengths and optical power intensity are compared. The photoresponse rise and decay times are also measured to find out the speed of a photodetector. TMDs, such as MoS₂, WS₂, MoSe₂, and WSe₂, show a tunable bandgap that change from a direct bandgap in monolayers to an indirect bandgap in multilayers. Therefore, the optoelectronic

properties of atomically thin layered TMDs can be tailored by utilizing their variable bandgaps and by improving the optical absorption by forming hybrid heterostructures. The large family of atomic layered TMDs is very attractive for their application in broadband photodetectors. For example, the internal quantum efficiency (IQE) of 85% for MoS₂,¹¹⁷ IQE of 70% with an ultrafast photoresponse time of 5.5 ps for WSe₂,¹¹⁸ IQE of 91% for WSe₂/

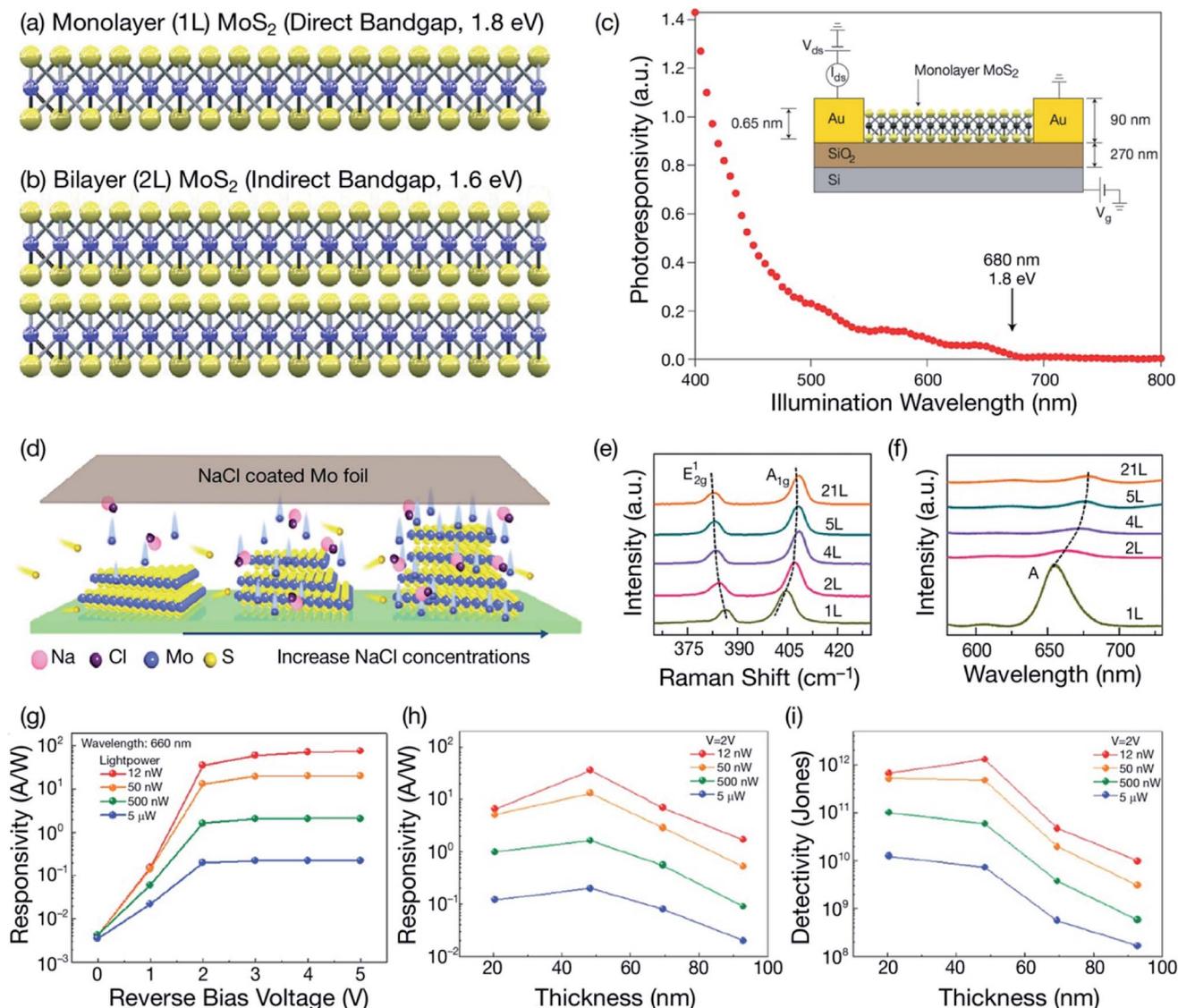


Fig. 3 (a and b) Schematic illustration of monolayer and bilayer MoS₂ on an atomic scale. The blue balls represent Mo atoms, while the yellow balls represent S atoms in MoS₂. The direct bandgap of 1.8 eV observed in monolayer (1L) MoS₂ transits to the indirect bandgap of 1.6 eV for bilayer (2L) MoS₂ and to 1.2 eV for multilayer (ML) MoS₂.^{96–98} In a bilayer (2L), MoS₂ single layers are bound by van der Waals forces having a nanoscale distance between the adjacent layers. (c) Photoresponsivity of a monolayer MoS₂ photodetector measured as a function of illumination wavelength in the 400 nm to 800 nm range. The photodetector shows an increasing photoresponsivity as the illumination wavelength is decreased from 680 nm to 400 nm. The monolayer MoS₂-based photodetectors can be used over a broad spectral range. The inset shows the structural view of a monolayer (6.5 Å thick) MoS₂ photodetector deposited on a back gate substrate with Au electrodes. Reprinted with permission from ref. 171, copyright © 2013 Macmillan Publishers Limited. (d) Schematic of NaCl-assisted layer-controlled low pressure CVD growth of MoS₂ flakes. (e) Layer-dependent Raman spectra of MoS₂ flakes showing the variation in the modes with increasing layer thickness. (f) Layer-dependent PL spectra of MoS₂ flakes. Reprinted with permission from ref. 174, copyright © American Chemical Society. (g) Photoresponsivity of the MoS₂/Si heterojunction photodetector as a function of reverse bias voltage at a 660 nm illumination wavelength under different incident laser powers. (h) Photoresponsivity of the MoS₂/Si photodetector as a function of the thickness of MoS₂ flakes at a bias voltage of 2.0 V under different incident powers. (i) Detectivity of the MoS₂/Si photodetector as a function of the thickness of MoS₂ flakes. Reprinted with permission from ref. 175, copyright © American Chemical Society.



MoSe₂ heterostructures,¹¹⁹ photoresponsivity of 2578 A W⁻¹ for monolayer WSe₂/nitrogen-doped graphene quantum dots (N-GQDs),¹²⁰ ultrafast charge transfer of 50 fs for MoS₂/WS₂ heterostructures after photoexcitation,¹²¹ and, interestingly, photoresponsivity as high as 1×10^{10} A W⁻¹ at 130 K and 5×10^8 A W⁻¹ at room temperature for a monolayer graphene/multilayer MoS₂ hybrid structure illuminated at 632 nm with a power of 376 fW μm⁻² with a response time of 1×10^3 s,¹²² demonstrate the suitability of atomic layered TMDs for developing optoelectronic devices, including photodetectors,^{123,124} LEDs,^{125,126} and solar cells.^{127–132}

The bandgap in TMDs can be adjusted by changing the number of layers, which allows tuning of the optical response over a broad range of wavelengths, from the ultraviolet (UV)-visible to NIR.^{133–135} Furthermore, the high carrier mobility and strong interaction of TMDs with light make these 2D materials interesting for optoelectronic applications. Atomically thin layered TMDs, including MoS₂,^{136–140} MoSe₂,^{141–144} MoTe₂,^{145,146} WS₂,^{147,148} WSe₂/WS₂,¹⁴⁹ WSe₂,^{120,150} WSe₂/h-BN,¹⁵¹ HfS₂,^{152,153} ReS₂,^{154,155} ReSe₂,^{156,157} SnS₂,^{158,159} and WSe₂/SnSe₂,¹⁶⁰ and the doped MoS₂ heterostructures¹⁶¹ have been studied for use in broadband photodetectors. Among 2D TMDs, MoS₂ atomic layers have also been extensively investigated for developing MoS₂ hybrid heterostructure-based photodetectors in combination with other materials, including MoS₂/Si,¹⁶² AuNPs/MoS₂,¹⁶³ MoS₂/WS₂,¹⁶⁴ MoS₂/WSe₂,¹⁶⁵ graphene/MoS₂/WSe₂/graphene,¹⁶⁵ MoTe₂/MoS₂,¹⁶⁶ GaTe/MoS₂,¹⁶⁷ PdSe₂/MoS₂,¹⁶⁸ MoS₂/graphene,¹⁶⁹ and MoS₂/BP.¹⁷⁰ The formation of hybrid heterostructures with other materials facilitates the modification of electronic and optoelectronic properties in order to improve the photoresponse of MoS₂-based photodetectors.

Several studies have demonstrated that the bandgap in MoS₂ can be tuned by changing the number of layers (thickness), from 1.8 eV for monolayer MoS₂ to 1.2 eV for multilayer MoS₂.^{96–98} This strategy could be used to adjust the optical response of MoS₂ over a broad spectral range. Mak *et al.*⁹⁶ reported the strongest direct bandgap photoluminescence (PL) in monolayer (1L) MoS₂, with 1000-fold enhancement of the PL intensity compared with bilayer (2L) MoS₂ as well as strong emergence of photoconductivity near the direct bandgap of 1.8 eV in monolayer MoS₂ and approximately 1.6 eV in bilayer MoS₂. These results confirm the occurrence of an indirect to direct bandgap transition using photoconductivity spectroscopy. Fig. 3(a and b) shows a schematic illustration of monolayer and bilayer MoS₂ on an atomic scale. In a bilayer (2L), MoS₂ single layers are bound by van der Waals (vdW) forces having a nanoscale distance between the adjacent layers. Atomic layer MoS₂ consisting of S–Mo–S atomic structures bonded through vdW forces show strong photodetection over a broad optical spectral range from the UV to IR. Lopez-Sanchez *et al.*¹⁷¹ reported a photoresponsivity of 880 A W⁻¹ and a detectivity of 2.5×10^{10} Jones (Jones = cm Hz^{1/2} W⁻¹) for monolayer MoS₂ at a bias gate voltage (V_{BG}) of -70 V and a V_{ds} of 8 V for a 561 nm wavelength under a 150 pW incident power (2.4×10^{-1} mW cm⁻²), along with a photoresponse in the 400–680 nm wavelength range, as shown in Fig. 3(c). CVD-grown monolayer MoS₂ phototransistors exhibit a photoresponsivity as high as

2200 A W⁻¹, a photogain of 5000 and a response time of 500 s at room temperature.¹⁷² Pang *et al.*¹⁷³ developed a MoS₂-based tribotronic phototransistor by combining a few-layer MoS₂ phototransistor with a sliding mode triboelectric nanogenerator (TENG). The photoresponsivity of the MoS₂ tribotronic phototransistor increased from 221.03 A W⁻¹ to 727.87 A W⁻¹ with increasing sliding distance from 0 mm to 8 mm under 10 mW cm⁻² laser power intensity and a 1.0 V drain voltage. This result indicates that the photoresponsivity of the MoS₂ phototransistor can be tuned by controlling the sliding distance, enabling self-powered photodetection with a TENG.

The number of MoS₂ atomic layers also significantly affects the electronic and optoelectronic properties. Yang *et al.*¹⁷⁴ reported the thickness-controlled growth of MoS₂ using the NaCl-assisted low pressure CVD method, where the number of layers of MoS₂ flakes was precisely controlled by simply increasing the concentration of the NaCl promoter. The Raman and PL spectra of 1L, 2L, 4L, 5L, and 21L MoS₂ flakes showed an increase in the frequency difference (Δ) between the two characteristic Raman peaks (E_{2g}^1 and A_{1g} vibration modes) from 18.3 to 25.7 cm⁻¹ and a redshift in the PL peak from 655 nm to 678 nm as the MoS₂ layer thicknesses increased from 1L to 21L (Fig. 3(d–f)). The intensity of the PL emission from MoS₂ flakes decreased dramatically with increasing number of layers because of the direct to indirect bandgap transition, and similarly, the bandgap of MoS₂ layers decreased with increasing layer thickness, from 1.8 eV for 1L to 1.5 eV for 2L and 1.2 eV for 25L. The optical transmittance measured at 550 nm gradually decreased with an increasing number of MoS₂ layers, ranging from 85.0% to 57.7%, 50.1% and 24.8% for 1L, 2L, 5L and 21L MoS₂. The UV-B absorptions of 1L, 2L, 5L and 21L MoS₂ flakes were over 95%. The thickness-dependent optoelectronic properties of multilayer MoS₂ have been studied. The highest values of photoresponsivity and detectivity reached 1×10^4 A W⁻¹ and 8×10^{12} Jones for monolayer-bilayer (1L–2L) heterojunctions and 4×10^3 A W⁻¹ and 6×10^{12} Jones for monolayer-8-layer (1L–8L) MoS₂ heterojunctions at 660 nm.

In another study, Shin *et al.*¹⁷⁵ deposited mechanically exfoliated multilayer MoS₂ flakes onto a Si layer to develop MoS₂/Si p–n heterojunction photodiodes; the optoelectronic properties were improved and optimized by controlling the number of layers of MoS₂ flakes. The photoresponse of the MoS₂/Si photodetector was measured at 405, 520, and 660 nm wavelengths under different incident power intensities and for various thicknesses of multilayer MoS₂ flakes. The 2 nm thick MoS₂ flake-based photodiodes showed very poor performance with low photoresponsivities of 3.36×10^{-3} and 6.64×10^{-5} A W⁻¹ and detectivities of 2.08×10^6 and 4.11×10^5 Jones at a 660 nm wavelength under incident optical powers of 50 nW and 50 μW at 2 V, respectively. The thickness-dependent maximum values of the photoresponsivity and detectivity were obtained for 48 nm MoS₂ flakes at 12 nW and 2 V. Photoresponsivities of 6.54, 35.7, 6.94, and 1.70 A W⁻¹ and detectivities of 2.33×10^9 , 1.52×10^{11} , 8.31×10^9 , and 8.64×10^7 Jones were measured for the 20, 48, 69, and 92 nm thick multilayer MoS₂ flakes at 2 V, respectively. The thickness-dependent optoelectronic properties for atomic layered MoS₂ have been



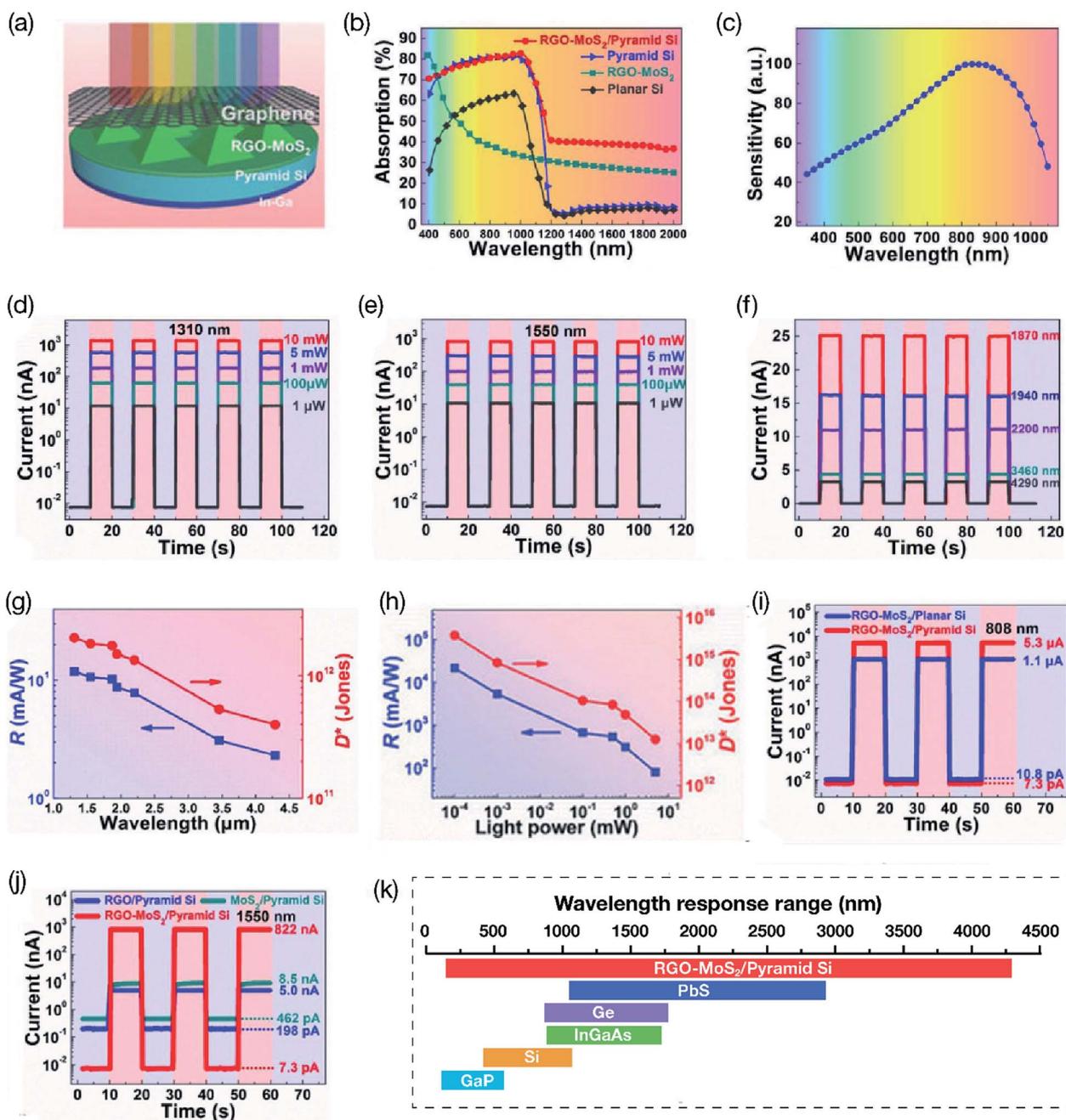


Fig. 4 (a) Schematic illustration of the 3D RGO–MoS₂/pyramid Si heterojunction-based photodetector. (b) A comparison of the absorption spectra of planar and pyramid Si, RGO (also referred as rGO), and RGO–MoS₂/Si heterojunction devices. (c) Photosensitivity of the RGO–MoS₂/pyramid Si heterojunction-based device between 350 to 1100 nm wavelength region. (d and e) Photocurrent switching behavior of heterojunction device measured under light illuminations at 1310 and 1550 nm at zero bias voltage ($V = 0$), self-powered devices. (f) Photocurrent switching behavior of heterojunction devices measured under 1870, 1940, 2200, 3460 and 4290 nm (NIR–MIR) light illuminations. (g) Photoresponsivity (R) and specific detectivity (D^*) curves of the heterojunction device measured as a function of wavelength from 1310 nm to 4290 nm at a fixed laser power intensity of 50 mW under zero bias voltage. (h) R and D^* curves of the heterojunction photodetector measured as a function of laser power intensity at 808 nm at the 100 nW laser power under zero bias voltage. (i) A comparison of the photocurrent switching behavior of RGO–MoS₂/pyramid Si and RGO–MoS₂/planar Si heterojunction devices measured at 808 nm wavelength under 1 μ W laser power intensity. (j) A comparison of the photocurrent switching behavior of RGO–MoS₂/pyramid Si heterojunction device with RGO/pyramid Si and MoS₂/pyramid Si devices measured at 1550 nm wavelength under 10 mW laser power intensity. (k) A comparison of the wavelength coverage by the RGO–MoS₂/pyramid Si heterojunction-based photodetector along with other traditional semiconductor-based photodetectors. The RGO–MoS₂/pyramid Si hybrid heterojunction-based photodetectors operated from 350 nm to 4.3 μ m (UV to MIR) ultra-broadband spectral range due to the bandgap narrowing caused by the S vacancy defects in MoS₂ crystals. Reprinted with permission from ref. 178, copyright © Wiley.



studied. Yang *et al.*¹⁷⁶ fabricated photodetectors using MoS₂ films with thicknesses greater than 6 nm, which showed a fast photoresponse time of <1 ms and a current $I_{\text{on}}/I_{\text{off}}$ ratio of $\sim 10^4$. The MoS₂ photodetector developed from a 9 nm thick MoS₂ film showed an 8 nA current at a 3 V bias voltage in the dark. The current increased to 1.47 and 13.5 μA at 30 and 140 μW , respectively. The current $I_{\text{on}}/I_{\text{off}}$ ratio of the MoS₂ photodetector increased from $\sim 10^1$ to $\sim 10^4$ for the 2 nm to 9 nm thick MoS₂ film but decreased by two orders of magnitude to 10^2 for the 32 nm thick film. Therefore, a MoS₂ film with a 9 nm thickness was used as the active layer for fabricating a MoS₂ photodetector. The current of the MoS₂ photodetector measured by irradiating it with 532 nm laser light at a 3 V bias showed a rapid increase to 25 μA and then decreased to 8 nA after turning off the laser light. Fig. 3(g–i) shows the photoresponsivity of the MoS₂/Si heterojunction photodetector as a function of reverse bias voltage and thickness of MoS₂ flakes under different incident powers. The photodetector based on 48 nm thick MoS₂ flakes showed a low noise equivalent power (NEP) value of $7.82 \times 10^{-15} \text{ W Hz}^{-1/2}$ at a 10 Hz frequency and a reverse bias voltage of 4.0 V, lower than the NEP of $3 \times 10^{-14} \text{ W Hz}^{-1/2}$ for the Si avalanche photodiode.¹⁷⁷ Ling *et al.*¹³⁹ also studied thickness-dependent photoresponse of MoS₂ photodetectors. The photoresponsivity increased from 0.4 A W^{-1} for bilayer MoS₂ photodetector to 1.8 A W^{-1} for 5L MoS₂ at 850 nm wavelength under a bias voltage of 5 V because of the enhanced photoabsorption. The EQE value of a 5L MoS₂ photodetector was found to increase from 30% at 1 V to 263% at 5 V. The 2L MoS₂ photodetector showed detectivity over 10^9 Jones, much higher compared with 3L and 5L MoS₂ photodetectors due to the low dark current. As seen from the above studies, multilayer MoS₂ is more appealing for developing broadband photodetectors due to its smaller indirect bandgap and extended optical spectral range compared with monolayer MoS₂, although further improvement of the photoresponsivity and detectivity is still needed.

3. Strategies for boosting the performance of MoS₂ photodetectors using hybrid heterostructures

The outstanding features of atomic layered MoS₂, such as the high optical transparency and carrier mobility, ultrafast photoresponse, and photodetection from the UV to IR, make MoS₂ highly desirable for developing broadband photodetectors. However, the low optical absorption by MoS₂ atomic layers hinders in achieving the high performance of photodetectors; therefore, different strategies have been applied to improve the photoresponse by generating abundant photo-excited carriers. Though the absorption of light by the MoS₂ atomic layers is an intrinsic property, the light absorption can be further enhanced in MoS₂ hybrid heterostructures by utilizing the supplementary light absorption of secondary integrated components in the hybrid structures. In addition to the incident light absorption, the carrier mobility of MoS₂ can be dramatically increased by forming hybrid heterostructures and nanocomposites with

a diverse range of inorganic, organic and polymeric materials to develop MoS₂ hybrid heterostructure-based high-performance ultrabroadband photodetectors. The large bandgap and limited absorption of visible light ($\sim 10\%$) displayed by monolayer MoS₂ hinder the attainment of a high power conversion efficiency, which eventually restricts the application of MoS₂ atomic layers in broadband photodetectors. The intrinsic photoresponse of atomic layered MoS₂ is rather constrained; therefore, different strategies have been explored to boost the carrier mobility and optical absorption of MoS₂ layers to improve the overall optoelectronic properties by maneuvering the electronic band structure. Different strategies such as control of atomic-layer thickness, hybrid heterostructures formation, chemical doping, surface functionalization, strain and defect engineering have been used for boosting the performance of MoS₂ photodetectors which are discussed throughout this article.

Diverse MoS₂ hybrid heterostructures with other inorganic, organic and 2D nanomaterials have been developed for extending the light absorption wavelengths and improving the charge transfer process. For example, Xiao *et al.*¹⁷⁸ reported reduced graphene oxide (RGO)–MoS₂/pyramid Si heterostructure-based photodetectors where 3L graphene and indium–gallium (In–Ga) alloy were used as top and bottom electrodes, respectively. Fig. 4 shows a schematic illustration of the 3D RGO–MoS₂/pyramid Si heterojunction-based photodetector, a comparison of the absorption spectra of planar and pyramid Si, RGO (also referred to as rGO), and RGO–MoS₂/Si heterojunction devices, photosensitivity between 350 to 1100 nm wavelength region, photocurrent switching behavior under light illuminations, wavelength and laser-power dependent photoresponsivity (R) and specific detectivity (D^*) of RGO–MoS₂/Si heterojunction-based photodetector under zero bias voltage and wavelength range covered by the RGO–MoS₂/pyramid Si heterojunction-based photodetector. The photo-voltage of RGO–MoS₂/Si heterojunction photodetector increased from 180 to 276 mV as the light power intensity was increased from 100 nW to 1 mW, which evidenced the self-powered operation of this photodetector at zero bias voltage ($V = 0$). Furthermore, hetero-junction photodetector showed the photoresponsivity of 21.8 A W^{-1} and detectivity of 3.8×10^{15} Jones at an 880 nm wavelength and a very broad optical spectrum range from the UV (350 nm) to mid-IR (4.3 μm). The self-driven heterojunction photodetectors exhibited photoresponsivity values of 2 to 11 mA W^{-1} and detectivity of 0.4 to 2×10^{12} Jones in the NIR–MIR (1870–4290 nm) range. The origin of high photoresponse over such an ultra-broadband range lies on several factors. In this RGO–MoS₂/pyramid Si heterostructure, the light absorption was increased by the pyramid Si structure while highly conductive RGO assisted in enhancing the charge separation and transfer process. The RGO–MoS₂/Si heterojunction shows maximum photoresponse at 800–900 nm. The light-harvesting by nanostructured pyramid Si surface was found to be 20% higher compared to planar Si in the 400 to 1000 nm spectral range. The photocurrent of RGO–MoS₂/pyramid Si device (5.3 μA) was found to be five times higher than that of the RGO–MoS₂/planar Si device (1.1 μA) at 808 nm.



Both pyramid and planar Si substrates barely absorb light over 1100 nm due to the bandgap of 1.1 eV. The integration of pyramid Si nanostructured surface assisted in extending the light absorption of the heterojunction in the NIR range. On the other hand, the RGO–MoS₂ composite exhibits strong light absorption up to 2 μm and dominant photovoltaic property at 1310 and 1550 nm where the corresponding current $I_{\text{on}}/I_{\text{off}}$ ratios of 1.9×10^6 and 1.2×10^6 and detectivity values of 2.04×10^{12} and 1.8×10^{12} Jones were measured, respectively. Electron–hole pairs are generated in the RGO–MoS₂/Si heterojunction under light illumination and then get separated at the heterojunction interface due to the built-in-electric field. The RGO–MoS₂/pyramid Si heterojunction device also displayed faster rise/decay times of 2.8 μs/46.6 μs compared with 32.6 μs/87.8 μs for the MoS₂/pyramid Si device due to the increased conductivity and internal electric field. The defects in MoS₂ assist in absorbing the light from NIR to MIR wavelength range, as a result photoexcited carriers from MoS₂ are transferred to the RGO layer, giving rise to the photocurrent in the NIR–MIR region. Therefore, the RGO–MoS₂/pyramid Si heterojunction-based photodetector was able to operate from 350 nm to 4.3 μm (UV to MIR) ultrabroad spectral range due to the bandgap narrowing caused by the S vacancy defects in MoS₂ crystals. XPS measurements and theoretical calculation also confirmed the existence of S vacancies where Mo/S atomic ratios of 1 : 2, 1 : 1.87 and 1 : 163 yielded bandgaps of 1.18, 0.30 and 0.28 eV, respectively, indicating a dramatic reduction in the bandgap of MoS₂. These results demonstrated that the formation of a hybrid heterostructure and occurring S vacancy defects in MoS₂ crystals contributed to the high performance of the RGO–MoS₂/pyramid Si heterojunction-based ultra-broadband photodetectors. In another study, Peng *et al.*¹⁷⁹ used an rGO layer as the conducting channel and a perovskite/MoS₂ bulk heterojunction (BHJ) as a photosensitizer for developing a hybrid photodetector that showed a photoresponsivity of 1.08×10^4 A W⁻¹, a detectivity of 4.28×10^{13} Jones, an EQE value of $2.0 \times 10^6\%$, and a >45 ms photoresponse time. The high photoresponse in the hybrid heterostructured system originated from the hole transfer from the perovskite to the rGO layer, facilitated by the suppression of the recombination of photocarriers from the perovskite/MoS₂ BHJ along with electron trapping in the MoS₂ layers.

Another common technique of improving the performance of MoS₂ photodetectors is *via* chemical doping. For example, Kang *et al.*¹⁸⁰ demonstrated that the field-effect mobility increased from $28.75 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ to $142.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and the photoresponsivity increased from 219 A W^{-1} to $5.75 \times 10^3 \text{ A W}^{-1}$ for APTES/MoS₂ hybrid phototransistors after aminopropyltriethoxysilane (APTES) doping of monolayer MoS₂. The 24.5-fold increase in the photoresponsivity of the APTES/MoS₂ photodetector resulted from the enhanced photocurrent after APTES doping. The photoresponsivity of APTES/MoS₂ devices reached 1212.8 A W^{-1} at a gate bias voltage of $V_g = 0$ and a V_{ds} of 5 V for a 520 nm wavelength under an incident power of 5.8 mW cm^{-2} . Yu *et al.*¹⁸¹ used mechanically exfoliated monolayer MoS₂ nanosheets sensitized with rhodamine 6G (R6G) organic dye to develop a photodetector with an enhanced photoresponse. The

dye-sensitized MoS₂/R6G-based photodetector showed a photoresponsivity of 1.17 A W^{-1} , a detectivity of 1.5×10^7 Jones, an EQE of 280% at 520 nm under an incident power of 1 μW and a photoresponse between wavelengths of 405 and 980 nm arising from charge transfer from the rhodamine 6G dye to monolayer MoS₂. In addition to photosensitive dyes, the high-κ Al₂O₃ dielectric has been used for enhancing the photoresponse of MoS₂ photodetectors. Huang *et al.*¹⁸² used zinc phthalocyanine (ZnPc) organic dye on the surface of monolayer MoS₂ to create a charge transfer interface. Dark and photocurrent of the dye-sensitized ZnPc-treated MoS₂ photodetectors increased by 10^3 to 10^5 times after a 30 nm thick top Al₂O₃ passivation layer was used. The photoresponsivity of 281 and 1.74 A W^{-1} were recorded for the bare MoS₂ and ZnPc-treated MoS₂ devices at 532 nm under light intensity of 0.07 mW cm^{-2} , respectively. The photoresponse of the Al₂O₃-passivated MoS₂/ZnPc hybrid photodetector was significantly improved: the photoresponsivity increased from 430 A W^{-1} to $1.4 \times 10^4 \text{ A W}^{-1}$ as the light intensity changed from 3.64 mW cm^{-2} to 0.07 mW cm^{-2} under a gate bias of 40 V, respectively. The photoresponse of the MoS₂/ZnPc hybrid device was 100 times faster than that of a pristine MoS₂ device. Wu *et al.*¹⁸³ demonstrated a photoresponsivity of $2.7 \times 10^4 \text{ A W}^{-1}$ for a MoS₂ photodetector using an Al₂O₃/ITO/SiO₂ substrate, which increased the light absorption of MoS₂ thin films. The high-κ Al₂O₃ dielectric also yielded a current $I_{\text{on}}/I_{\text{off}}$ ratio of 10^9 under a 2 V gate bias voltage, a mobility of $84 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and a subthreshold swing of 104 mV dec^{-1} .

Several different strategies including MoS₂ plasmonic heterostructures, chemical doping of MoS₂ layers, halide perovskites, 2D–0D MoS₂/QDs heterostructures, and 2D–2D MoS₂/vdWHs hybrid heterostructures have been employed for boosting the light absorption efficiency of MoS₂ thin films, hence improving the performance of MoS₂ photodetectors. The strategies assist in terms of increasing the carrier mobility and the absorption of incident sunlight by MoS₂ hybrid heterostructures.

3.1 MoS₂ plasmonic heterostructures

The plasmonic nanostructures have been used for enhancing the optical absorption of semiconductor-based photodetectors. The photoresponse of atomic layered MoS₂ photodetectors has been optimized by forming hybrid plasmonic heterostructures for light-harvesting. The gold nanoparticles (AuNPs)@MoS₂ heterostructure-based plasmonic photodetectors having AuNPs core and ML MoS₂ shell showed 10-fold increase in photoresponsivity (0.5 A W^{-1}) compared with pristine MoS₂ phototransistors (0.057 A W^{-1}).¹⁶³ The maximum photoresponsivity of 30 A W^{-1} was achieved for the Si-supported AuNPs@MoS₂ heterojunction-based gateless photodetector greater than that of Si/MoS₂ heterojunction (1.1 A W^{-1}) phototransistors. Bang *et al.*¹⁸⁴ developed a monolayer (1L) MoS₂ photodetector exploiting the surface plasmon of a one-dimensional (1D) silver nanowire (AgNW) network. Fig. 5(a–d) shows the schematic diagram of the 1L-MoS₂/AgNW-based photodetector, dark field image of a hybrid photodetector, and photocurrent–voltage



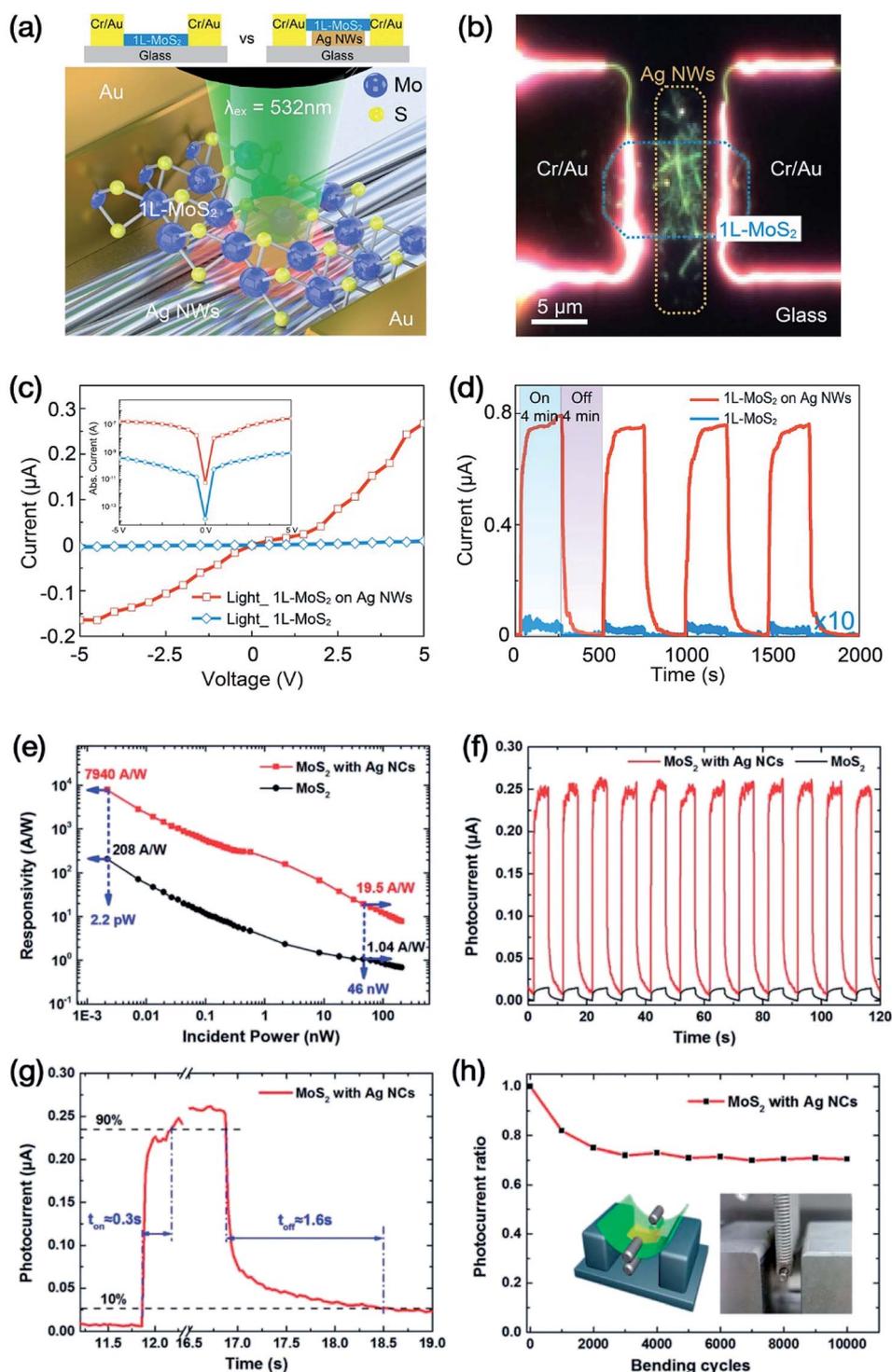


Fig. 5 (a) Schematic diagram of the 1L-MoS₂ and Ag nanowires (NWs) based photodetector. Upper area shows the schematic diagrams of the pristine 1L-MoS₂ and the 1L-MoS₂/AgNWs hybrid photodetector on a glass substrate along with Cr/Au electrodes. (b) Dark field image of the 1L-MoS₂/AgNWs hybrid photodetector (60% density of AgNWs). (c) A comparison of the photocurrent–voltage curves between pristine 1L-MoS₂ (blue line) and the 1L-MoS₂/AgNWs hybrid photodetector (red line) measured at 532 nm. The inset represents a logarithmic scale of the photocurrent–voltage characteristic. (d) Time dependent photocurrent of the pristine 1L-MoS₂ (blue) and 1L-MoS₂/AgNWs hybrid photodetectors (red) recorded with an $I_{\text{on}}/I_{\text{off}}$ time of 4 min. Reprinted with permission from ref. 184, copyright © American Chemical Society. (e) Photoresponsivity of MoS₂ and MoS₂/Ag nanocubes (NCs) hybrid-based photodetectors as a function of incident laser power. (f) A comparison of the transient photocurrent of MoS₂ and MoS₂/Ag nanocubes (NCs) photodetectors under periodical $I_{\text{on}}/I_{\text{off}}$ illumination at 1 V. (g) Photocurrent rise (t_{on}) and decay (t_{off}) times of MoS₂/Ag NCs hybrid-based photodetector. (h) Photocurrent of MoS₂/Ag NCs hybrid-based photodetector as a function of number of bending cycles, where the insets show schematic and photograph of a bending setup. Reprinted with permission from ref. 186, copyright © Wiley.



curves of pristine 1L-MoS₂ and the 1L-MoS₂/AgNWs hybrid photodetectors as a function of applied voltage (from -5 V to +5 V) and the time-dependent photocurrents. The current $I_{\text{on}}/I_{\text{off}}$ ratios of 1.39×10^4 and 62.97 were measured for the 1L MoS₂/AgNW hybrid and pristine 1L-MoS₂ photodetectors, respectively. The 1L MoS₂/AgNW hybrid heterostructure-based device showed 560- and 250-fold increases in the PL and photocurrent, respectively, compared with the pristine 1L MoS₂ device. Similarly, the photoresponsivity of 59.60 A W⁻¹ and detectivity of 4.51×10^{10} Jones for the 1L MoS₂/AgNW hybrid photodetector were 1000 times enhanced compared to those of the pristine 1L-MoS₂ photodetector (0.05 A W⁻¹ and 4.11×10^7 Jones). The significant improvement in the optoelectronic properties of the 1L MoS₂/AgNW hybrid photodetector occurred due to the surface plasmon coupling of the AgNW network. In another study, Jing *et al.*¹⁸⁵ enhanced the localized surface plasmon resonance (LSPR) by depositing Ag nanoparticles onto monolayer MoS₂ for developing phototransistors. AgNPs/MoS₂ hybrid-based phototransistors showed a 470% increase in photoresponsivity, changing from 5.35×10^3 A W⁻¹ to 2.97×10^4 A W⁻¹ at 610 nm for 25 nm AgNPs. The interactions between MoS₂ layers and light are enhanced by the integrated Au or Ag nanostructures due to the LSPR, which significantly improves the photoresponse of plasmon heterostructure-based hybrid MoS₂ photodetectors. Sun *et al.*¹⁸⁶ used silica layer-coated silver nanocubes (AgNCs) with optimized LSP in the gap mode for developing a flexible MoS₂ photodetector. The PL spectrum of MoS₂/Ag nanocubes hybrid was found to be significantly enhanced compared with MoS₂ film after modifying of Ag NCs with underneath Ag thin film due to interactions between the MoS₂ excitons and LSP in the gap mode. Fig. 5(e-g) compare the photoresponsivity and photocurrent of MoS₂ and flexible MoS₂/Ag nanocubes (NCs) hybrid-based photodetectors as a function of incident laser power and time, respectively. Photocurrent rise (τ_{rise}) and decay (τ_{decay}) times and mechanical stability of flexible MoS₂/AgNCs hybrid-based photodetector are also depicted. The photocurrents of the photodetectors fabricated using pristine MoS₂ film and MoS₂ film with underneath Ag film were almost the same (48–50 nA) at 3 V under 520 nm laser illumination with 46 nW incident power whereas the photocurrent of flexible photodetector having MoS₂ film in the gap of Ag layer and AgNCs was considerably increased to 0.91 μ A under similar experimental conditions due to the Ag nanocubes. The AgNCs decorated MoS₂ film shows 19-fold increment in photocurrent compared with pristine MoS₂ film. The photoresponsivity of MoS₂ with AgNCs based flexible photodetector increased 38 times to 7940 A W⁻¹ at 3 V under 2.2 pW incident power compared with pristine MoS₂ film with decreasing incident power intensity due to the reduced scattering and recombination of the photogenerated charge carriers. Photocurrent rise time (τ_{rise}) of 0.3 s and decay time (τ_{decay}) of 1.6 s were estimated for the flexible MoS₂/AgNCs hybrid-based photodetector. The flexible photodetector showed good mechanical stability after bending at a curvature of 8 mm for 10 000 times because 71% photocurrent ratio of devices was retained. The photocurrent initially decreased up to 3000 bending cycles, but no noticeable decrease in photocurrent ratio was observed thereafter. These

studies show that the integration of plasmonic nanostructures with MoS₂ atomic layers can significantly increase the photoresponsivity of MoS₂ based photodetectors.

3.2 Chemical doping of MoS₂ layers

As discussed above, performance of MoS₂ photodetectors can be significantly increased by doping MoS₂ layers with APTES,¹⁸⁰ rhodamine 6G¹⁸¹ and ZnPc¹⁸² organic dyes, which efficiently generate charge transfer interfaces between MoS₂ and chemical dopants. A few examples of boosting the performance of MoS₂ photodetectors *via* chemical doping are presented here. Heo *et al.*¹⁸⁷ used n-type doping of MoS₂ flakes with triphenylphosphine (PPh₃) to improve the electronic and optoelectronic properties of multilayer MoS₂ by utilizing charge transfer from PPh₃ to MoS₂, in which the PPh₃ doping concentration ranged from 1.56×10^{11} to 9.75×10^{12} cm⁻². The PPh₃ doping considerably increased the mobility from 12.1 to 241 cm² V⁻¹ s⁻¹ and the $I_{\text{on}}/I_{\text{off}}$ current ratio from 8.72×10^4 to 8.70×10^5 for the MoS₂ transistor. The photoresponsivity of the MoS₂ photodetector similarly increased from 2.77×10^3 to a maximum value of 3.92×10^5 A W⁻¹ under an applied laser power of 5 pW. The detectivity decreased by three orders of magnitude, from 6.82×10^{13} Jones to 2.36×10^{10} Jones, as the device doping temperature was increased from 150 °C to 350 °C. The MoS₂ photodetector also showed long-term stability, where the photoresponsivity decreased by 1.58% after 14 days of exposure to air; additionally, the PPh₃ doping was reversible, allowing repetitive use of the photodetector device. For example, a MoS₂/Si heterojunction-based photodetector showed no degradation in the photovoltage after storing in an air atmosphere over a period of a month.¹⁶²

The chemical doping of monolayer MoS₂ film over a 13 cm² area for developing photodetector devices was reported by Kim *et al.*¹⁶¹ The n-type MoS₂ semiconductor transitions into the p-type semiconductor after doping with Nb. Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), and photoluminescence (PL) spectroscopy were used to characterize the doped MoS₂. The Nb-doped MoS₂ showed binding energy peaks at lower values compared with MoS₂. The Nb- and Mn-doped monolayer MoS₂ FETs showed p-type and n-type doped MoS₂ characteristics, respectively. Fig. 6 shows doped MoS₂ photodetectors, transfer curves (drain current *versus* gate voltage) and photoresponsivity of Nb-doped MoS₂ photodetectors as a function of optical power and compares the photoresponsivity, EQE, and detectivity of Nb-doped MoS₂ photodetectors with up-doped MoS₂ photodetectors at different laser wavelengths. A sublinear relationship ($R \propto P^\alpha$) was observed for Nb-doped MoS₂ photodetector between photoresponsivity (R) and optical power (P), which yielded an α value of -0.9 using the power function fitting method. The Nb-doped MoS₂ photodetector showed photoresponsivity of 2×10^5 A W⁻¹, which is 10^6 times higher compared with the photoresponsivity of 0.52 A W⁻¹ for the MoS₂ film measured under similar conditions at a 550 nm laser wavelength. The highest photoresponsivity of 4.83×10^5 A W⁻¹ was recorded at 750 nm wavelength for the Nb-doped MoS₂ photodetector. Likely, the EQE for Nb-doped MoS₂



photodetector was found to be $9.31 \times 10^7\%$, which was 10^6 times higher compared with EQE of 107% for the MoS₂ device at 450 nm wavelength. The Nb-doped MoS₂ photodetector showed detectivity of 5.0×10^{12} Jones, compared with the detectivity of 3.93×10^8 Jones for the updoped MoS₂ photodetector at 750 nm wavelength, which resulted in 10^5 times higher detectivity after Nb-doping. The photocurrent values of Nb-doped MoS₂ devices were also increased by 10^5 folds due to the charge-transfer process. The Nb-doped MoS₂ devices also exhibited faster rise/decay times of 4.9 ms and 5.7 ms compared with 0.63 s and 1.63 s for the updoped MoS₂ devices at 550 nm laser wavelength, respectively. The Nb-doped MoS₂ photodetector devices showed stability for 30 days under ambient conditions.

The phototransistors utilizing hybrid MoS₂/PbS QDs, were fabricated by Pak *et al.*¹⁸⁸ in which the PbS QDs were surface modified with n-type tetrabutylammonium iodide (TBAI) and p-type 1,2-ethanedithiol (EDT) ligands to develop p-n junctions. The MoS₂/PbS-TBAI/PbS-EDT hybrid device showed a photoresponsivity of $5700 \pm 71.2 \text{ A W}^{-1}$ for a single-junction device and $6120 \pm 96.9 \text{ A W}^{-1}$ for a double-junction device at an incident laser power of 20 pW. The photocurrent rise time of 40 ms for the single-junction device was reduced to 950 μs for the double-junction device. The efficient charge transfer occurring in the MoS₂/PbS QD hybrid photodetector yielded a maximum

photoresponsivity of $5.4 \times 10^4 \text{ A W}^{-1}$ and a detectivity of 1×10^{11} Jones. High photoresponsivity of 2570 A W^{-1} and detectivity of 2.2×10^{12} Jones at 635 nm in a few-layer MoS₂ photodetector have been demonstrated under zero gate voltage with the use of a poly(vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE)] ferroelectric polymer gate.¹³⁷ This MoS₂/P(VDF-TrFE) hybrid photodetector also showed photodetection over the 0.85 μm to 1.55 μm spectral region and excellent stability over 90 000 repeated cycles of operation.

Organic molecules and polymers have been hybridized with MoS₂ to develop photodetectors. Sun *et al.*¹⁸⁹ used narrow bandgap organic semiconducting polymer, poly(diketopyrrolopyrrole-terthiophene) (PDPP3T), into large bandgap monolayer MoS₂ to develop UV-NIR photodetectors. The photoresponsivities of MoS₂/PDPP3T based phototransistors were enhanced by one to two orders of magnitude compared with MoS₂. Flexible MoS₂/PDPP3T photodetectors that were fabricated on a PET substrate. The MoS₂/PDPP3T hybrid showed photoresponsivities of 276, 445, and 269 mA W⁻¹ and detectivity values of 2.59×10^8 , 3.14×10^8 , and 2.53×10^8 Jones at 365, 660, and 850 nm, respectively. The flexible MoS₂/PDPP3T photodetectors were developed on a PET substrate which exhibited photoresponse from 365 nm to 940 nm. The bending testing of flexible MoS₂/PDPP3T

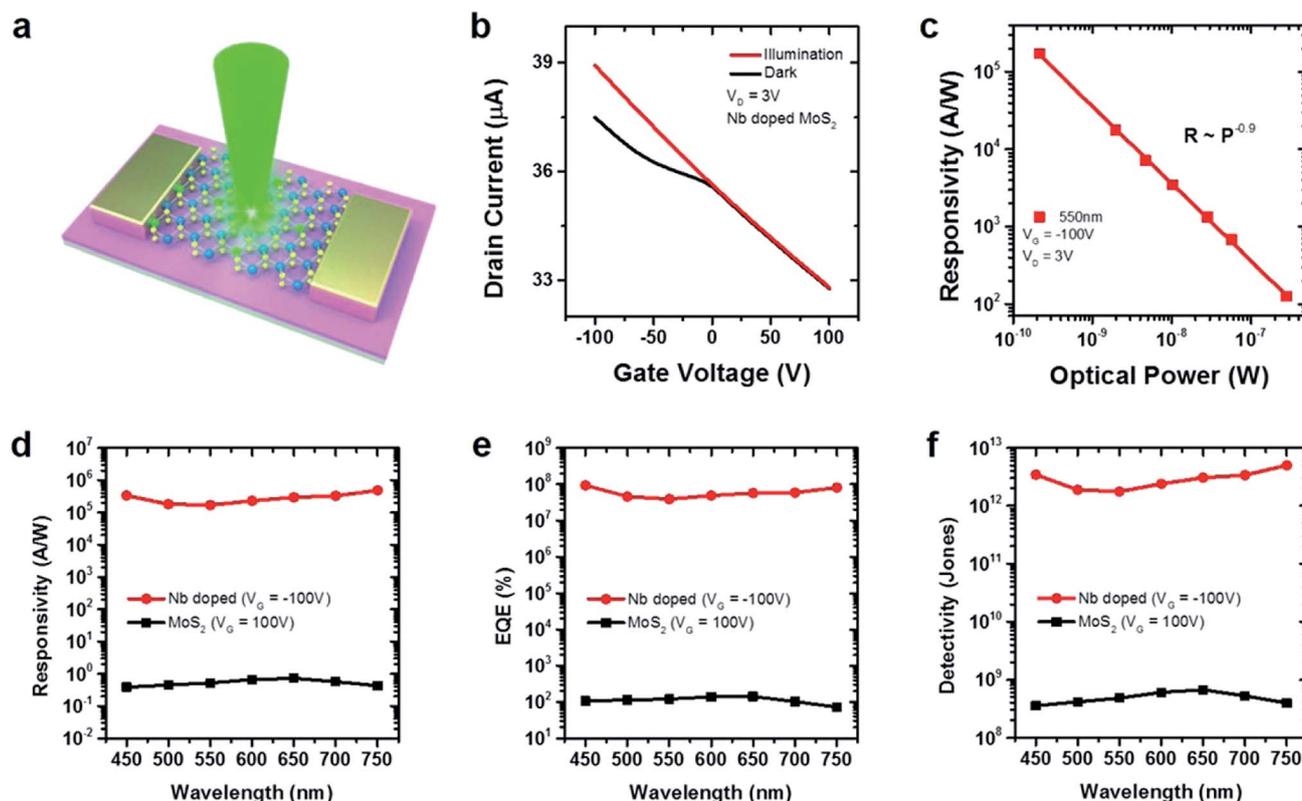


Fig. 6 (a) Schematic illustration of doped MoS₂ photodetectors. (b) Transfer curves of Nb-doped MoS₂ photodetectors at 550 nm laser wavelength under 282 nW optical power. (c) Photoresponsivity (R) of Nb-doped MoS₂ photodetectors as a function of optical power (P) at 550 nm laser wavelength and at $V_d = 3 \text{ V}$ and $V_g = -100 \text{ V}$. A comparison of photoresponse characteristics of Nb-doped MoS₂ photodetectors with up-doped MoS₂ photodetectors. (d) Photoresponsivity, (e) EQE, and (f) detectivity of Nb-doped MoS₂ photodetectors and up-doped MoS₂ photodetectors at $V_g = -100 \text{ V}$ as a function of laser wavelength. Reprinted with permission from ref. 161, copyright © American Chemical Society.



photodetector was conducted under 660 nm light illumination a 9 mm bending radius where some decrease in the photocurrent was noticed after 10 000 bending cycles. Likely, the environmental stability of the flexible MoS₂/PDPP3T photodetector was also evaluated without any encapsulation in an air atmosphere for 35 days where slight changes were observed both in the dark and photo currents. MoS₂/PDPP3T hybrid photodetector devices demonstrated both mechanical stability and durability in the air.

3.3 Halide perovskite MoS₂ heterostructures

Organometallic halide perovskites have emerged as an important class of light harvesting materials for developing solar cells^{8,9,130,131} because of their tunable bandgap, extended light absorption over a broad spectral range, high photoconversion efficiencies, charge-carrier mobilities, and ease of solution processing into ultrathin layers. These unique optoelectronic properties of perovskites have been utilized in improving the performance of MoS₂ photodetectors. Wu *et al.*¹⁹⁰ developed hybrid phototransistors using 0D cesium lead halides perovskite (CsPbI_{3-x}Br_x) QDs and 2D monolayer MoS₂. The CsPbI_{3-x}Br_x QD/1L MoS₂ 0D–2D mixed-dimensional vdW heterostructure-based photodetectors showed a high photoresponsivity of 7.7×10^4 A W⁻¹, a detectivity of 5.6×10^{11} Jones, and an EQE value greater than 10⁷% at 532 nm light illumination under optical power of 1.5 μW due to the photogating effect. Also under similar illumination conditions, the photocurrent of CsPbI_{3-x}Br_x QDs/1L MoS₂ hybrid phototransistor was found to be increased by 15.3 times that of a pristine MoS₂ phototransistor, indicating the efficient separation of photo-generated carriers and the strong absorption of 0D CsPbI_{3-x}Br_x QDs.

MoS₂ layers combined with perovskite CsPbBr₃ nanosheets have been investigated for fabricating photodetectors, as reported by Song *et al.*¹⁹¹ The MoS₂/CsPbBr₃ hybrid-based photodetector showed a photoresponsivity of 4.4 A W⁻¹, a detectivity of 2.5×10^{10} Jones, and a 302% EQE arising from photoexcited carrier separation at the interface of the perovskite CsPbBr₃ nanosheet and MoS₂ layer. The photoresponsivity of the MoS₂/CsPbBr₃ hybrid-based photodetector increased by three orders of magnitude compared to the MoS₂ photodetector without the perovskite CsPbBr₃. The response time of the hybrid photodetector decreased from 65.2 to 0.72 ms after adding MoS₂ layers. The high photon absorption by the CsPbBr₃ perovskite layer and the carrier transport by the MoS₂ layer contributed to the high performance of the photodetector devices. The 3 MoS₂/CsPbBr₃ hybrid, CsPbBr₃ and MoS₂ photodetector devices showed current in the 0.1–1 nA range. Photocurrents of 5030, 1230, and 54 nA were measured for the MoS₂/CsPbBr₃ hybrid, CsPbBr₃, and MoS₂, respectively. Light on/off ratios of 16 700, 14 300, and 150 were calculated for the MoS₂/CsPbBr₃ hybrid, CsPbBr₃, and MoS₂ photodetector devices, respectively. PL quenching, as well as a fast decay time, was observed for the MoS₂/CsPbBr₃ hybrid due to the charge transfer from the perovskite CsPbBr₃ nanosheet to the MoS₂ atomic layers. Kang *et al.*¹⁹² reported perovskite/MoS₂/APTES

hybrid heterostructure-based photodetector devices. The photoresponsivity of a MoS₂ photodetector was enhanced from 636 to 4.9×10^3 A W⁻¹ and detectivity from 1.53×10^9 to 8.76×10^{10} Jones for a perovskite/MoS₂ hybrid device after applying the perovskite layer at 520 nm. The similar increment was also observed for 655 and 785 nm laser wavelengths, but for 850 nm. The photocurrent of the perovskite/MoS₂/APTES hybrid photodetector was increased by 25.9 times that of the MoS₂ photodetector at 520 nm wavelength. The photoresponsivity and detectivity of the perovskite/MoS₂/APTES hybrid photodetector device were enhanced from 816.6 to 2.11×10^4 A W⁻¹ and from 3.93×10^9 to 1.38×10^{10} Jones after APTES n-doping, respectively. The photoresponsivity of the perovskite/MoS₂/APTES hybrid photodetector was increased by 94.2 times, from 2.11×10^4 to 1.94×10^6 A W⁻¹ at 520 nm as a function of incident laser power (1.57 nW to 4.63 pW), while the highest detectivity value was 1.29×10^{12} Jones due to the decreased scattering between photogenerated electrons. The perovskite/MoS₂/APTES hybrid photodetectors showed significantly higher photoresponsivity values compared with the MoS₂/APTES photodetector.¹⁸⁰ The performance of photodetector devices followed the sequence as: perovskite/MoS₂/APTES > perovskite/MoS₂ > MoS₂, as clearly evident from the above-mentioned results. Since perovskites are susceptible to environmental degradation,^{8,130} therefore, the stability of perovskite/MoS₂/APTES photodetectors was improved by applying encapsulating layers of octadecyltrichlorosilane (OTS) and PMMA. The stability of non-encapsulated, PMMA and OTS/PMMA encapsulated perovskite/MoS₂/APTES photodetector devices in air was measured at different wavelengths: 520, 655, 785 and 850 nm. Photoresponsivity and detectivity deteriorated by 50% and 70% after 68 h for PMMA and 20.4% and 28.7% after 200 h for OTS/PMMA encapsulated devices, and 20.4–24.8% and 28.7–34.2% for perovskite/MoS₂/APTES photodetectors at different wavelengths, respectively. These data showed that performance of MoS₂ photodetectors can be improved by applying a perovskite absorption layer and furthermore *via* a chemical doping process.

The flexible perovskite/MoS₂ hybrid photodetectors were developed using a polyethylene terephthalate (PET) substrate. Sun *et al.*⁷⁶ used CVD grown atomic layered MoS₂ film, deposited a 20 nm Ti/50 nm Au thick electrode and then spin-coated a triple cations [Cs_{0.05}(MA_{0.17}FA_{0.83})_{0.95}Pb(I_{0.83}Br_{0.17})]₃ lead mixed-halide perovskite layer to fabricate a hybrid photodetector. Fig. 7 shows the schematics and energy-band diagram of the perovskite/MoS₂ hybrid photodetector under illumination, photoresponse of the pristine MoS₂ and perovskite/MoS₂ hybrid photodetector devices and mechanical stability test of the flexible hybrid photodetector device up to 20 000 bending cycles. The flexible perovskite/MoS₂ hybrid photodetectors showed photoresponsivity of 342 A W⁻¹ at 2 V bias potential without any gate voltage at 520 nm under 2.2 pW incident power. The hybrid photodetector devices showed high stability without any encapsulation, photoresponse and decay times of 27 ms and 21 ms, respectively. The hybrid devices were fabricated on rigid SiO₂/Si, glass and flexible PET substrates. The pristine MoS₂ photodetector device showed the photocurrent of 32 nA at 2 V



bias potential with 20 nW incident power, which increased to 84 nA after spin-coating the perovskite layer. The perovskite/MoS₂ hybrid device showed photocurrent of 2 pA at 1 V and 3.8 pA at 2 V bias potential. At 1 V bias potential, the photocurrent of the hybrid device increased from 2 pA in the dark to 40 nA under illumination. The hybrid photodetector device having 10 μm channel exhibited the highest photoresponsivity value of 1.7 A W⁻¹ under 50 nW incident power. The estimated detectivity was 1.14 × 10¹² Jones. The photocurrent of the hybrid devices increased as a function of increased incident power. The photoresponsivity of the hybrid photodetector device was dramatically increased to 342 A W⁻¹ as the incident power decreased to 2.2 pW because of the suppressed scattering between the photogenerated charge carriers, and mechanical stability test on flexible perovskite/MoS₂ hybrid photodetectors was performed with 5 mm curvature up to 20 000 bending cycles. The device retained 91% photocurrent value under bending compared with the corresponding *I*-*V* curves of a device without bending. There was no noticeable decline in photocurrent ratio from 2000 to 20 000 bending cycles, indicating the strong mechanical stability of the flexible perovskite/MoS₂ hybrid devices.

3.4 2D-0D MoS₂-quantum dots (QDs) heterostructures

As discussed above, photodetectors fabricated with hybrid MoS₂/PbS QDs yield significantly high photoresponsivity.¹⁸⁸ 0D QDs have been used for improving the performance of MoS₂

photodetectors. Huo *et al.*¹⁹³ developed photodetectors by incorporating mercury telluride (HgTe) colloidal QD film into a TiO₂-encapsulated few-layer MoS₂ channel. In the MoS₂/TiO₂/HgTe hybrid heterostructure, TiO₂ is a buffer layer between the MoS₂ channel and the HgTe QD layer that acts as an electron accepting medium to form a p-n junction with the HgTe QDs to facilitate the transfer of photogenerated charges from the HgTe QDs to the MoS₂ channel. MoS₂/TiO₂/HgTe hybrid photodetectors showed a high photoresponsivity of 10⁶ A W⁻¹ under an incident power of 70 fW (0.35 μW cm⁻²) at a gate voltage (*V*_g) of -15 V and a *V*_{ds} of 1 V, which is seven orders of magnitude higher than that of the HgTe QD photodetector due to the low carrier mobility (2 × 10⁻³ cm² V⁻¹ s⁻¹) of the HgTe QDs. The detectivity of MoS₂/TiO₂/HgTe hybrid photodetectors was 1 × 10¹² Jones at a 2.0 μm wavelength, which is four orders of magnitude higher than that of pure HgTe-based photodetectors (10⁷ to 10⁸ Jones). This study is an excellent example of extending the spectral wavelengths of a MoS₂-based photodetector over 2.0 μm. In another study, Zhang *et al.*¹⁹⁴ reported 2D MoS₂-0D zinc cadmium selenide (ZnCdSe)/zinc sulfide (ZnS) colloidal QD-based photodetectors, where monolayer (1L) to multilayer (ML) MoS₂ thin film acts as a carrier transport channel and a core/shell ZnCdSe/ZnS QDs sensitizing thin layer functions as a light harvester. Fig. 8 compared the photoresponsivity and detectivity as a function of laser incident power for the pristine MoS₂ based photodetectors with hybrid MoS₂-ZnCdSe/ZnS QD-based photodetectors having monolayer MoS₂ (1L), bilayer MoS₂ (2L), trilayer MoS₂ (3L), and multilayer MoS₂

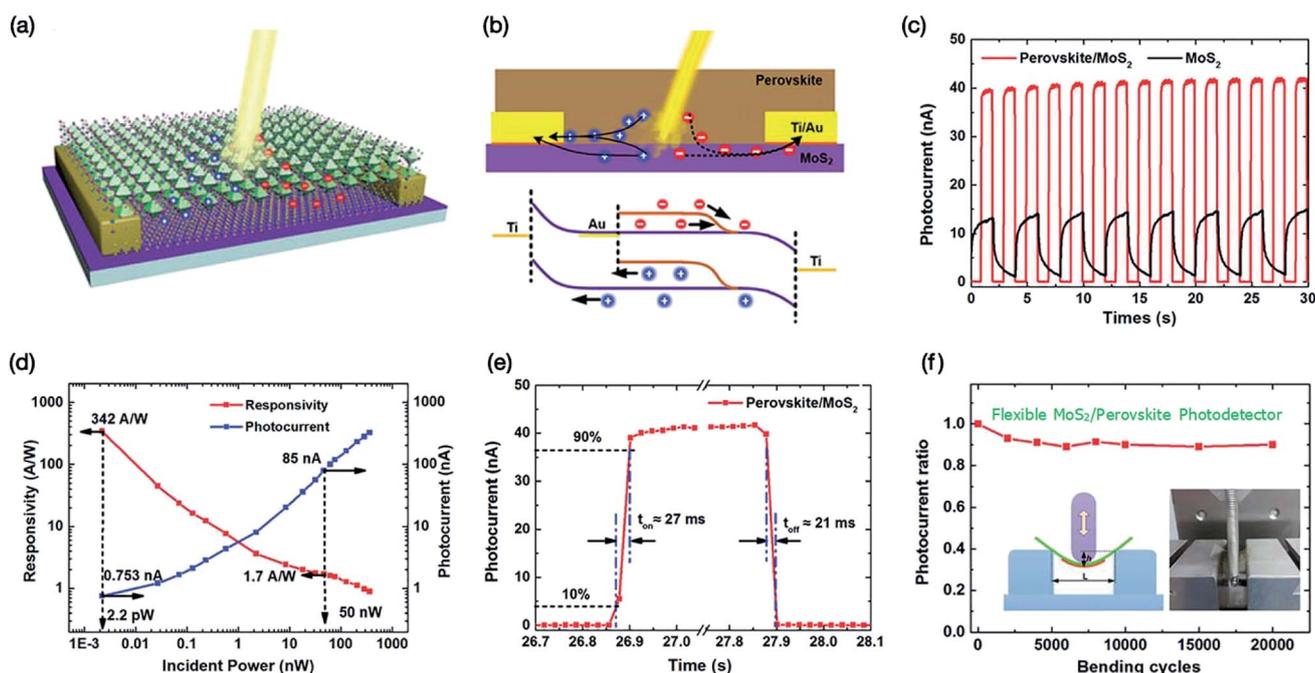


Fig. 7 (a) Schematic illustration of perovskite/MoS₂ hybrid based photodetector. (b) Working mechanism and energy-band diagram of flexible perovskite/MoS₂ hybrid photodetector under illumination. (c) A comparison of the transient photoresponse of the pristine MoS₂ and flexible perovskite/MoS₂ hybrid based photodetectors under on/off illumination. (d) Photoresponsivity and photocurrent of the flexible perovskite/MoS₂ hybrid photodetector devices as a function of laser incident power. (e) Photoresponse rise and decay time of the flexible perovskite/MoS₂ hybrid based photodetector. (f) Mechanical stability test of the flexible perovskite/MoS₂ hybrid photodetector device up to 20 000 bending cycles. Reprinted with permission from ref. 76, copyright @ Elsevier.



(ML). There is a very significant increase in photocurrent, photoresponsivity, and detectivity after adding ZnCdSe/ZnS QD sensitizing layer. The photocurrent of hybrid MoS₂-ZnCdSe/ZnS QD-based photodetectors increased by 1000 folds compared with pristine MoS₂ photodetectors. The response time of the MoS₂-ZnCdSe/ZnS QD-based photodetector was reduced to 0.3 s from 15 s for MoS₂ layers, making the hybrid device 50 times faster. The schematic of the MoS₂-ZnCdSe/ZnS QD interface and their energy diagram depict the transfer of electrons from ZnCdSe/ZnS QDs to MoS₂ layers *via* a tunneling process and the transfer of excitons from ZnCdSe/ZnS QDs to the MoS₂ layer *via* nonradiative energy transfer (NRET) processes after a heterojunction formation. In the NERT process, the MoS₂ layer acts as an acceptor, whereas the ZnCdSe/ZnS QDs thin layer acts as a donor. The photoresponsivity of the hybrid MoS₂-ZnCdSe/ZnS QD-based photodetectors increased by three orders of magnitude to 3.7×10^4 A W⁻¹ compared to pristine MoS₂ layers of 10 A W⁻¹. Likely, the detectivity of the MoS₂-ZnCdSe/ZnS QD-based photodetector increased to 1.0×10^{12} Jones, and the gain increased by five orders of magnitude to 1.08×10^5 after adding a layer of QDs due to the increased absorption and efficient transfer of energy from the photoexcited ZnCdSe/ZnS QDs layer to MoS₂ layers. The better performance of the 2D-0D hybrid photodetector is associated with high carrier mobility in the MoS₂ layer and the creation of effective photon absorption/exciton in the ZnCdSe/ZnS QDs layer. The MoS₂ layers efficiently quench the

fluorescence of the ZnCdSe/ZnS QDs in the 2D-0D hybrid nanostructures exhibiting ultrasensitivity and high gain.

3.5 2D-2D MoS₂-2D van der Waals heterostructures (vdWHs)

MoS₂-based photodetectors have been designed and developed using different electrode materials and van der Waals heterostructures (vdWHs) to improve their optoelectronic properties. The performance of MoS₂-based photodetectors can be increased using rGO layers.^{178,179} Fig. 9(a-c) shows a schematic of the multilayer MoS₂ photodetector developed with Mo bottom contacts (100 nm) on thermally oxidized SiO₂/Si substrates (270 nm) and the photoresponsivity and EQE of the ML MoS₂ photodetector measured over the 400-1100 nm spectral region, as reported by Saenz *et al.*¹⁹⁵ The photodetector exhibited ultrahigh photoresponsivity of 1.4×10^4 A W⁻¹ and detectivity of 2.3×10^{11} Jones at a 700 nm wavelength with a 14.5 pW laser power for the multilayer MoS₂ device fabricated using 100 nm thick Mo bottom electrodes, with a broadband photoresponse from the UV to IR regime. The photoresponsivity increased from 8×10^3 A W⁻¹ at 400 nm to 1.4×10^4 A W⁻¹ at 1100 nm at a bias voltage of 20 V, and the EQE varied from 3.6×10^4 to 1.4×10^4 within the 400 nm to 1100 nm wavelength range. Photoresponsivities of 1×10^3 A W⁻¹ and 42 A W⁻¹ were recorded at incident powers of 70 pW and 15.85 nW, respectively, at a bias voltage of 5 V and 300 K. The decrease in the photoresponsivity resulted from the loss of photocarriers due to

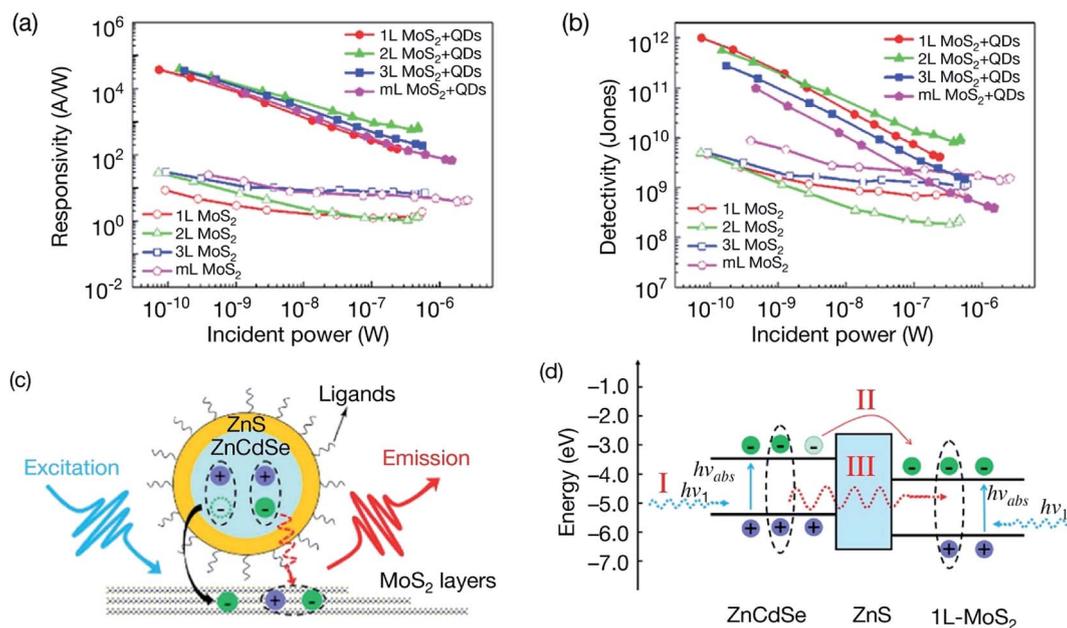


Fig. 8 (a and b) A comparison of photoresponsivity and detectivity of pristine MoS₂ based photodetectors with hybrid MoS₂-ZnCdSe/ZnS QD-based photodetectors having different MoS₂ layers; monolayer MoS₂ (1L), bilayer MoS₂ (2L), trilayer MoS₂ (3L), and multilayer MoS₂ (ML) as a function of laser incident power. Both responsivity and detectivity increased significantly after adding QD sensitizing layer. (c) Schematic of the MoS₂-ZnCdSe/ZnS QD interface showing the transfer of electrons from ZnCdSe/ZnS QDs to MoS₂ layers *via* a tunneling process (represented by black arrow) and the transfer of excitons from ZnCdSe/ZnS QDs to MoS₂ layer *via* nonradiative energy transfer (NRET) processes (represented by red dashed arrow) under an illumination. (d) Energy diagram of the 1L MoS₂-ZnCdSe/ZnS QD interface after a heterojunction formation where three photoelectrical processes are involved; (I) photon excitation in 1L-MoS₂ and ZnCdSe/ZnS QDs; (II) transfer of electrons from the ZnCdSe/ZnS QDs to 1L-MoS₂ *via* a tunneling process and (III) exciton transfer from the ZnCdSe/ZnS QDs to 1L-MoS₂ *via* NRET processes. Reprinted with permission from ref. 194, copyright © American Chemical Society.



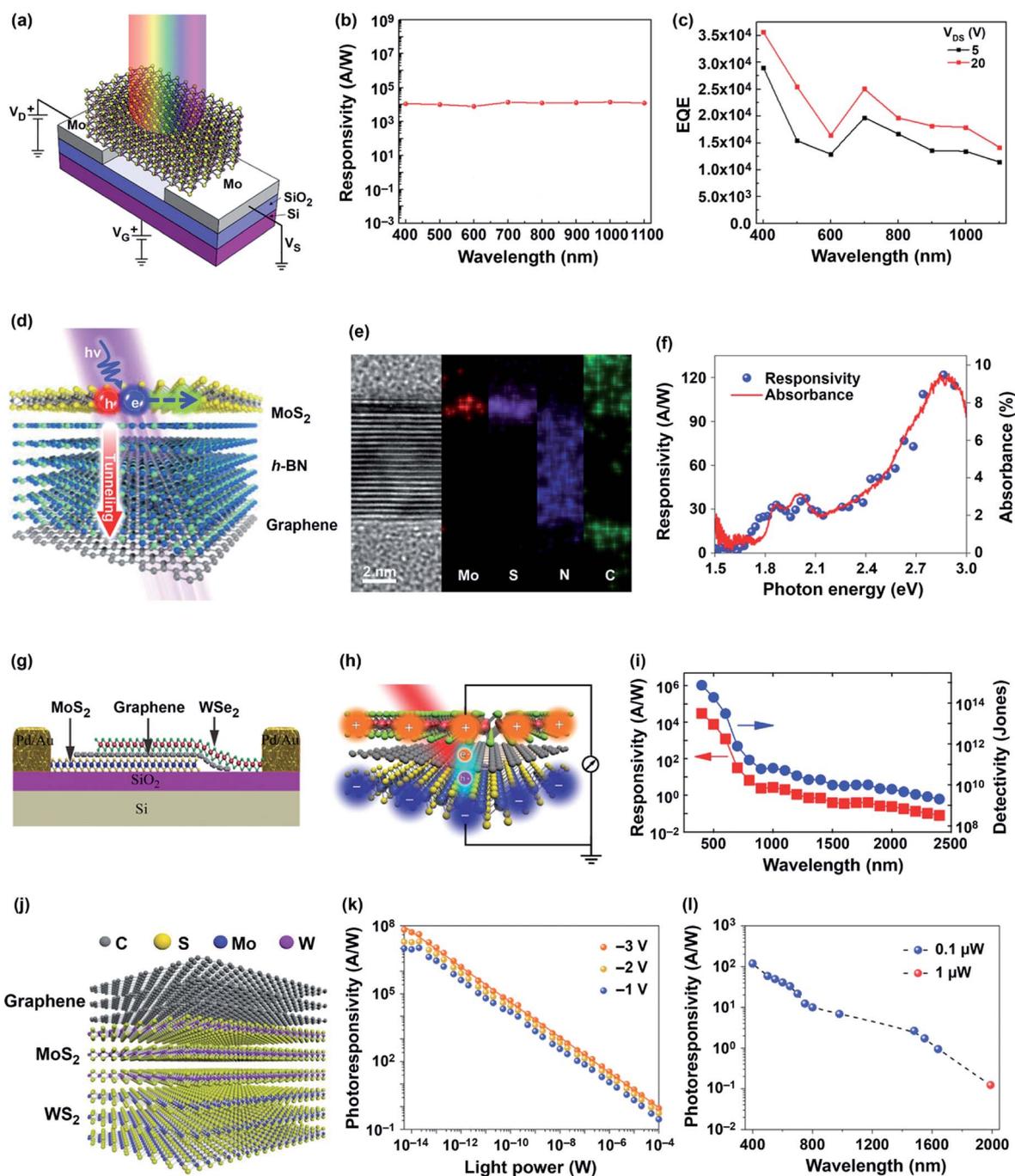


Fig. 9 (a) Schematic of the multilayer MoS₂ photodetector fabricated using 100 nm thick Mo bottom contacts on 270 nm thick thermally oxidized SiO₂/Si substrates. (b) Photoresponsivity and (c) external quantum efficiency (EQE) of the ML MoS₂ photodetector as a function of wavelength over the 400–1100 nm spectral region. Reprinted with permission from ref. 195, copyright © 2018 Springer Nature Limited. (d) Schematic of the MoS₂/h-BN/graphene vdWH photodetector using the MoS₂ layer as a photon absorber, the h-BN layer as a selective hole tunneling layer and the graphene layer as a bottom electrode. The generation of an electron–hole pair upon light illumination and occurrence of selective hole carrier tunneling through the h-BN layer. (e) Cross-sectional STEM image and energy dispersive X-ray spectroscopy (EDS) elemental mapping of the MoS₂/h-BN/graphene vdWHs. (f) Photoresponsivity and absorbance of the MoS₂/h-BN/graphene photodetector as a function of photon energy. Reprinted with permission from ref. 196, copyright © American Chemical Society. (g) Side view of MoS₂/graphene/WSe₂ vdWHs. (h) Schematic drawing of atomically layered MoS₂/graphene/WSe₂ vdWHs for broadband photodetection. (i) Photoresponsivity and detectivity (D^*) of the MoS₂/graphene/WSe₂-based broadband photodetector in the 400 nm to 2400 nm wavelength range. Reprinted with permission from ref. 198, copyright © American Chemical Society. (j) Schematic of the graphene/MoS₂/WS₂ vdWH-based optical-fiber compatible photodetector. (k) Photoresponsivity of the graphene/MoS₂/WS₂-based broadband optical-fiber compatible photodetector as a function of laser illumination power at different bias voltages. (l) Photoresponsivity of the graphene/MoS₂/WS₂-based broadband optical-fiber compatible photodetector as a function of wavelength in the 400 nm to 2000 nm spectral range. Reprinted with permission from ref. 199, copyright © Wiley.



recombination effects. Vu *et al.*¹⁹⁶ used MoS₂/h-BN/graphene vdWHs to develop a photodetector. Fig. 9(d–f) depicts a schematic of the MoS₂/h-BN/graphene photodetector, its cross-sectional STEM image, its energy dispersive X-ray spectroscopy (EDS) elemental mapping and its photoresponsivity and absorbance as a function of photon energy. An h-BN insulating layer was inserted between the MoS₂ photoabsorber and the graphene electrode. In these vdWHs, the dark carriers are suppressed at the graphene/h-BN barrier (2.7 eV), whereas the tunneling of photocarriers is facilitated at the MoS₂/h-BN junction (1.2 eV). Varying the thickness of the h-BN insulating layer from 0–25 nm played an important role in the tunneling of the dark carriers and photocarriers; a 7 nm thick h-BN barrier layer was most effective in blocking the conduction of dark carriers and facilitating the tunneling of photocarriers through the low-hole MoS₂/h-BN junction. The photodetector fabricated from MoS₂/h-BN/graphene vdWHs with a 7 nm thick h-BN layer exhibited a photoresponsivity of 180 A W⁻¹ with a maximum photocurrent/dark current ($I_{\text{photo}}/I_{\text{dark}}$) ratio of 10⁵ and a detectivity of 2.6×10^{13} Jones. The detectivity of the vdWH-based photodetector was 1000 higher compared with the values of 2.5×10^{10} Jones for the lateral MoS₂ p–n junction¹⁷⁰ and 5×10^{10} Jones for monolayer MoS₂-based photodetectors.¹⁹⁷ Furthermore, the rise/fall times of 0.23 s/0.25 s for the vdWH-based photodetector were 1000 times faster than those of the monolayer MoS₂-based photodetector.¹⁷¹

Similarly, atomically thin layered MoS₂/graphene/WSe₂ (p–g–n) vdWHs were developed by Long *et al.*¹⁹⁸ In this vdWH, graphene was sandwiched within the p–n junction to broaden the absorption spectral range and photodetection sensitivity. Fig. 9(g–i) shows a side view of MoS₂/graphene/WSe₂ vdWHs, a schematic for the broadband photodetection and the wavelength-dependent photoresponsivity and detectivity (D) of the MoS₂/graphene/WSe₂-based broadband photodetector over the 400 nm to 2400 nm wavelength range. Both the photoresponsivity and detectivity decreased with increasing excitation laser wavelength. The maximum photoresponsivity and detectivity values of the p–g–n heterostructure-based photodetector were on the order of 10⁴ A W⁻¹ and 10¹⁵ Jones in the visible range and decreased to a few mA W⁻¹ and 10⁹ Jones at the 2400 nm wavelength, respectively. A photoresponsivity of 4250 A W⁻¹, a detectivity of 2.2×10^{12} Jones and an EQE value of $1.0 \times 10^6\%$ were measured at 532 nm with a 0.2 nW laser power intensity for the p–g–n heterostructure-based photodetector, while the photoresponsivity dropped to 306 mA W⁻¹ in the near-IR region, 940 nm with a 17 nW laser power. The photocurrent mapping conducted at 830 nm with a 20.5 μW laser power indicated that the overlapping regions of the MoS₂, graphene and WSe₂ atomic layers are responsible for the strong photoresponse instead of the electrode regions. The MoS₂/graphene/WSe₂ heterostructure-based broadband photodetector also showed a fast photoresponse with a 53.6 μs rise time and a 30.3 μs fall time, and can be used from the visible to near-IR spectral range at room temperature.

Fig. 9(j–l) illustrates a schematic of the graphene/MoS₂/WS₂ vdWH-based optical-fiber compatible photodetector fabricated by Xiong *et al.*¹⁹⁹ The photoresponsivity of this optical-fiber

tuned photodetector was recorded as a function of applied laser power at different bias voltages and in the 400 nm to 2000 nm spectral range. The photoresponsivity increased with increasing bias voltage at a fixed incident light power of 1 pW. The fiber-based photodetector was impacted by the trap state of MoS₂ and WS₂. The optical fiber-based photodetector exhibited a photoresponsivity of 17.1 A W⁻¹ at 1550 nm, and an optimum photoresponsivity of 6.6×10^7 A W⁻¹ was measured at 400 nm under an incident light power of 6.35 nW cm⁻² (5 fW) at a –3 V bias voltage. The EQE value of graphene/MoS₂/WS₂ vdWH reached 2.06×10^8 at a 5 fW incident power input (400 nm) and 13.71 at 1550 nm, orders of magnitude higher compared with multilayer graphene and MoS₂/WS₂ heterostructure. The photoconductive photobolometric mechanism involved in graphene/MoS₂/WS₂ vdWH yielded the photoresponse times of 7 ms and 160 ms at the illumination power of 200 μW and 5 mW, respectively, quite different than those of graphene (photobolometric) and MoS₂/WS₂ heterostructure (photoconductive) resulting from different photogeneration mechanisms. The photoresponsivity was measured as a function of the wavelength by maintaining the incident light power at 100 nW for the 400 nm to 1640 nm range and 1.0 μW for the 2.0 μm wavelength. The photodetection limit of graphene/MoS₂/WS₂ vdWH was between 400 nm (5 fW) and 1550 nm (20 nW), compared with 0.1 mW at 1550 nm for graphene and 0.2 nW at 400 nm for MoS₂/WS₂ heterostructure. The photodetector exhibited a decrease in photoresponsivity as the wavelength was increased from 400 nm to 2.0 μm but retained a high photoresponsivity. This result demonstrates the broadband photodetection of the graphene/MoS₂/WS₂ vdWH-based photodetector from the visible to IR ranges.

The MoS₂-based vdWHs are also interesting for developing highly sensitive photodetectors. Liu *et al.*²⁰⁰ developed photodetectors using vertically stacked p-Sb₂Te₃/n-MoS₂ vdWHs, which showed a photoresponsivity of 330 A W⁻¹, a fast response time of <500 μs and a power conversion efficiency of 4.5%. Yang *et al.*²⁰¹ fabricated few-layer SnSe/MoS₂ vdWHs with a current $I_{\text{on}}/I_{\text{off}}$ ratio of 1×10^5 . The type-II SnSe/MoS₂ heterostructure FET showed a fast response time of <10 ms, a photoresponsivity (R_{λ}) of 100 A W⁻¹, and an EQE of $23.3 \times 10^3\%$ under 532 nm light illumination. The gallium telluride (GaTe)–MoS₂ p–n vdW heterojunction FET showed a fast response time of <10 ms, a photoresponsivity of 1.365 A W⁻¹, and an EQE of 266% under 633 nm light illumination,¹⁶⁷ much higher compared with the photoresponsivity of 0.03 A W⁻¹ for a flexible GaTe photodetector measured at a wavelength of 473 nm under an incident laser power of 80.5 mW cm⁻².²⁰² The electronic and optoelectronic properties of MoS₂ have been improved by fabricating vdWH-based FETs with other 2D materials.^{36,203} The vertically stacked vdWHs of SnS₂/MoS₂ monolayers showed improved mobility of 27.6 cm² V⁻¹ s⁻¹, $I_{\text{on}}/I_{\text{off}}$ ratio of 10⁶, and photoresponsivity of 1.36 A W⁻¹.²⁰⁴ 2D layered vdW semiconductor MoTe₂ has been used with MoS₂ to develop vdW heterojunction p–n diodes for extending the range of photodetection in the NIR region. Ding *et al.*¹⁶⁶ fabricated vertically stacked 2H-MoTe₂/MoS₂ bilayer heterostructures using CVD technique for broadband photodetectors ranging from 200 nm (UV) to 1100 nm



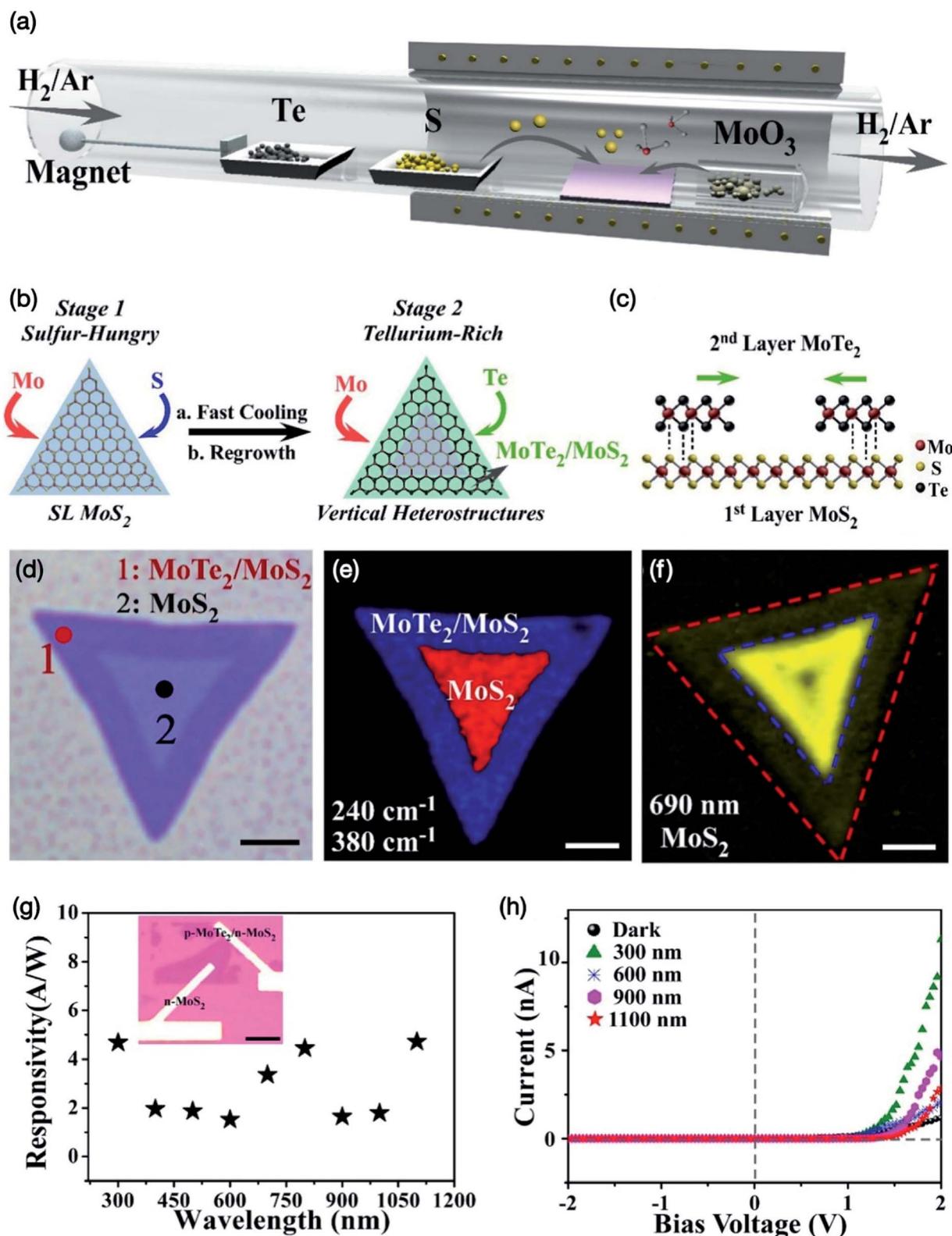


Fig. 10 (a) Schematic diagram of experimental set-up showing magnet-assisted CVD growth method for preparing MoTe₂/MoS₂ heterostructure. (b) Vertically stacking approach of MoTe₂/MoS₂ bilayer heterostructures. (c) Schematic side view of the MoTe₂/MoS₂ bilayer heterostructure where Mo atoms are represented in red, S atoms in yellow, Te atoms in black. (d) Optical image of the MoTe₂/MoS₂ flake. (e) Raman intensity mapping at 240 cm⁻¹ shown in blue and 380 cm⁻¹ in red. (f) PL intensity mapping at 690 nm wavelength where the red dash lines defect the MoTe₂/MoS₂ heterojunction and the blue dash lines indicates the interface of regions with the inner core and outside ring. (g) Photoresponsivity of MoTe₂/MoS₂ photodetectors as a function of wavelength between 300 nm to 1100 nm region. (h) I-V current characteristics of the MoTe₂/MoS₂ photodetectors at different applied bias voltage. Reprinted with permission from ref. 166, copyright © Elsevier.



(NIR) regions. Fig. 10 shows the magnet-assisted CVD growth method, stacking approach of MoTe₂/MoS₂ bilayer heterostructures, optical image of the MoTe₂/MoS₂ flake, Raman intensity and PL mapping, photoresponsivity and I - V current of the MoTe₂/MoS₂ heterostructure-based photodetectors. MoTe₂ was observed to grow along the edges of MoS₂ seeds. During the first stage, the sulfur-hungry environment assisted in growing the initial MoS₂ seeds. In the second stage, MoTe₂ was seen to grow over the first MoS₂ monolayer from the edge nucleation and thereafter epitaxial growth of MoTe₂ occurred from the center area on the MoS₂ surface to create a vertically stacked MoTe₂/MoS₂ bilayer heterostructure. Atomic force microscopy (AFM) evidenced from the height image of a triangular MoTe₂/MoS₂ bilayer heterostructure that center and outside regions have a thickness of 0.8 nm and 1.6 nm, respectively. The center area is the MoS₂ monolayer whereas the outside area is a vertically stacked MoTe₂/MoS₂ bilayer heterostructure. The core-ring structure of vertically stacked MoTe₂/MoS₂ bilayer heterostructures also was confirmed by the Raman mapping of MoTe₂ at 240 cm⁻¹ and MoS₂ mapping at 380 cm⁻¹ and 375 cm⁻¹. PL spectra showed emission peaks at 640 nm and 690 nm for the MoS₂ flake where the peak at 690 nm indicates a significant quenching for the MoS₂. A band alignment occurred when the MoS₂ monolayer was vertically stacked with the MoTe₂ monolayer due to the interlayer coupling interactions. The vertically stacked MoTe₂/MoS₂ bilayer heterostructures also showed higher image contrast than that of the MoS₂ monolayer with high-angle annular dark field (HAADF) STEM. The photoresponsivity and external quantum efficiency (EQE) of 4.71 A W⁻¹ and 532% at 1100 nm and 4.67 A W⁻¹ and 1935% at 300 nm under the light power density of 4.209 mW cm⁻² were measured for the MoTe₂/MoS₂ bilayer heterostructure-based photodetectors, respectively, which was up to two orders of magnitude higher compared with an exfoliated MoTe₂ heterostructure-based photodetector. Epitaxial growth of MoTe₂ monolayer on the top and along the edges of MoS₂ monolayer having a 2H-stacking mode dramatically enhanced the contact and interfacial interactions of the MoTe₂/MoS₂ bilayer heterostructures, which yielded a better performance of photodetectors from UV to infrared regions. Ahn *et al.*^{205a} fabricated self-powered visible-invisible multiband photodetectors using MoTe₂/MoS₂ multilayer-based semivertical heterojunction p-n diodes, which showed an ideality factor of less than 1.5 and current $I_{\text{on}}/I_{\text{off}}$ ratio of more than 10⁴. The MoTe₂/MoS₂ photodiodes showed photodetection from 405 nm (violet) to 1310 nm (NIR) wavelength region and a linear dynamic range (LDR) of 130 dB between 10⁻⁵ to 1 W cm⁻² applied laser power intensity in the photovoltaic mode. The MoTe₂/MoS₂ photodetectors showed photoresponsivity of 0.62 A W⁻¹ and 0.86 A W⁻¹ at 532 nm laser illumination, and LDR of 132 dB and 84 dB in the photoconduction mode ($V = -2$ V), respectively. The values of detectivity ranged from 10⁸ to 10¹⁰ Jones within 405–1310 nm wavelength range. A prototype self-powered visible-invisible multiband image sensor was also constructed with MoTe₂/MoS₂ photodiodes in the photovoltaic mode operation. Wang *et al.*^{205b} developed p-MoTe₂/n-MoS₂ based van der Waals heterojunctions for photodetectors. The multilayer MoTe₂/MoS₂

vdWH-based FETs showed a photocurrent that increased by several orders of magnitude with increasing laser power density, a photoresponsivity of 0.15 A W⁻¹ and an EQE value of 39.4% at a 473 nm laser wavelength, a photocurrent $I_{\text{on}}/I_{\text{off}}$ ratio of 780 and a field-effect mobility of 1.9 cm² V⁻¹ s⁻¹, much larger compared with pure MoTe₂-based FETs. The photoelectric response of vertically stacked MoTe₂/MoS₂ bilayer heterostructure-based photodetectors¹⁶⁶ was much higher compared with these MoTe₂/MoS₂ FET-based photodetectors. The PdSe₂/MoS₂¹⁶⁸ vdWH-based photodetectors exhibiting a very broad spectral range from 450 nm (visible) to 10.6 μm (LWIR) have been reported. The photoresponsivity of PdSe₂/MoS₂ vdWH-based devices reached 22.86 A W⁻¹ at 450 nm, 11.15 A W⁻¹ at 637 nm, 4.24 A W⁻¹ at 940 nm and 28.83 A W⁻¹ at 4.012 μm at the V_{ds} of 1 V. The photoresponsivity of heterostructure photodetectors at 4.012 μm was higher than those of 2.7 μm, 3.1 μm, and 10.6 μm wavelengths. The detectivity was more than 6.88 × 10⁹ Jones over the full spectral range and reached a maximum value of 6.09 × 10¹⁰ Jones at 4.012 μm. The low NEP value of 0.13 pW Hz^{-1/2} was measured for the heterostructure device in the full spectral region. The photoresponse for heterostructure devices was much faster having rise/fall times of 65.3 μs/62.4 μs at 637 nm illumination.

A significant improvement in mobility was also observed for NbS₂/MoS₂ vdWH-based Schottky-effect FETs, which yielded a mobility of 800 cm² V⁻¹ s⁻¹ at room temperature with graphene source/drain (S/D) contacts, compared with the low mobility values of 15–170 cm² V⁻¹ s⁻¹ for pure MoS₂ FETs with Au contacts, depending on the annealing conditions.²⁰⁶ In contrast, lower photoresponsivities were recorded for GaSe/MoS₂ vertical heterostructure-based FETs at wavelengths from 300 nm to 670 nm under 6.412 mW cm⁻² to 2.169 mW cm⁻² optical power, as were a low mobility of 0.026 cm² V⁻¹ s⁻¹ and a low $I_{\text{on}}/I_{\text{off}}$ current ratio of 10³, compared with pure MoS₂ FETs due to the increased scattering of charge transport carriers at the interface of the vdW heterojunctions.^{207a} Atomic layer vdW heterostructure-based photodiodes were fabricated using n-type MoS₂ on top of p-type GaSe crystals, which showed photoresponsivity of ~3 A W⁻¹ at 532 nm when symmetric FL graphene contacts were used.^{207b} However, the asymmetric GaSe/FL graphene and MoS₂/Au contacts based vdW photodiodes resulted in a low NEP value of 10⁻¹⁴ W Hz^{-1/2} because of the reduced dark current and a large LDR of ≈ 70 dB.

3.6 Interface coupling effect in dual-gated MoS₂ phototransistors

Different strategies have been used to improve photoresponse of photodetectors. Deng *et al.*²⁰⁸ demonstrated that the interface coupling effect (ICE) enhanced photoresponse in fully-depleted silicon-on-insulator (FD-SOI) metal-oxide-semiconductor field-effect transistors (MOSFETs). The thinner Si film-based photodetectors exhibited higher photoresponsivity due to a stronger ICE. CMOS-compatible SOI photodetectors achieved a high photoresponsivity of 3.3 × 10⁴ A W⁻¹. This ICE strategy was also employed for improving the performance of MoS₂-based phototransistors. Liao *et al.*²⁰⁹ first reported interface coupling effect



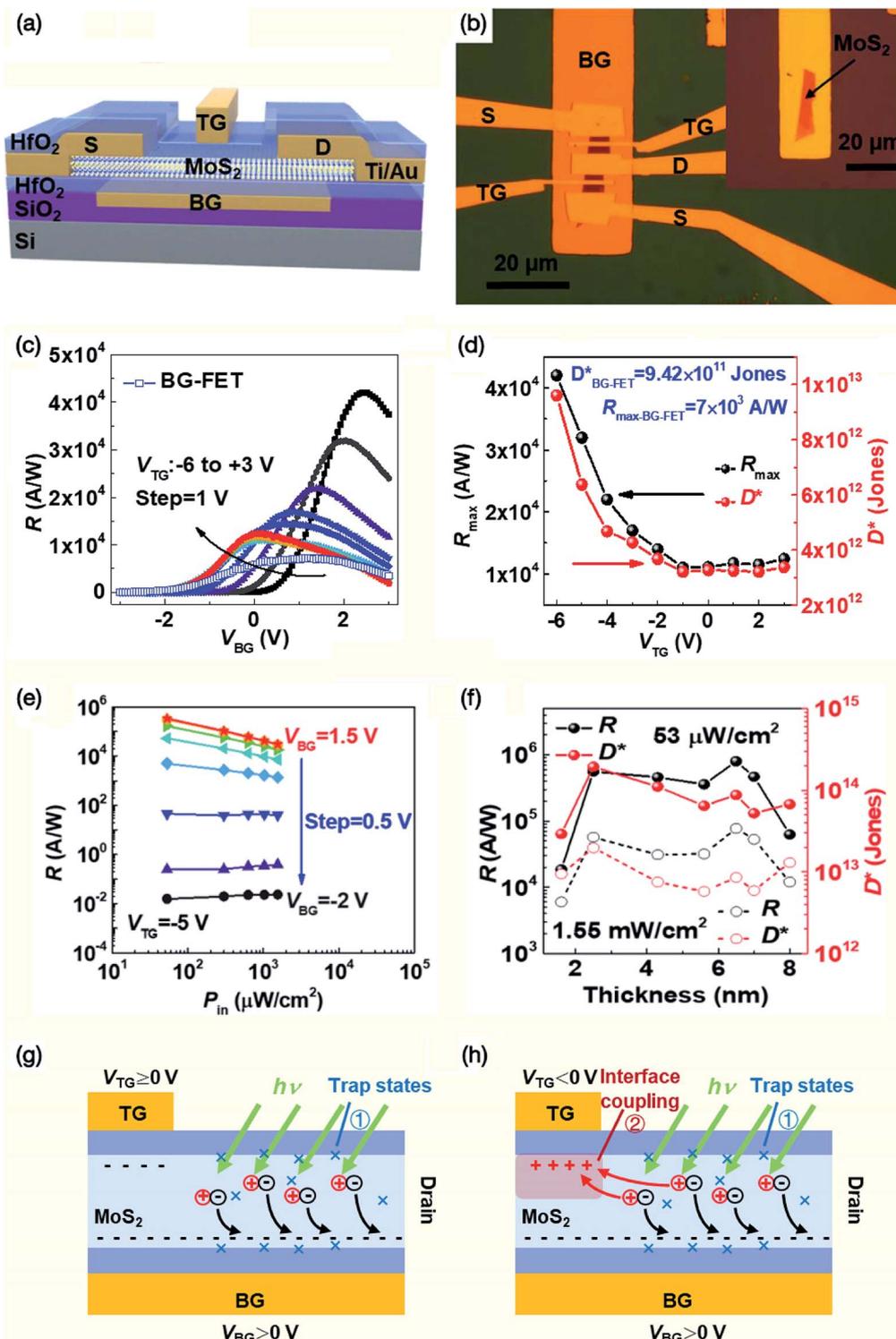


Fig. 11 (a) 3D schematic of a dual-gated (DG) MoS₂ phototransistor on a Si/SiO₂ substrate showing location of top-gate (TG), bottom-gate (BG), source (S), drain (D) and HfO₂ layer as a BG dielectric. (b) Optical microscopic image of a DG MoS₂ phototransistor. Inset is an optical microscopic image of the ML MoS₂ flake. (c) Photoresponsivity (R) of DG MoS₂ phototransistor as a function of back-gate voltage (V_{BG}) under different top-gate voltage (V_{TG}) with illumination power density (P_{in}) of 1.55 mW cm⁻² at 550 nm wavelength. Blue line + square symbol represent photoresponsivity of MoS₂ BG-FET. (d) The maximum photoresponsivity (R_{max}) and detectivity (D^*) as a function of applied V_{TG} . (e) P_{in} dependence of photoresponsivity the DG MoS₂ phototransistor with varying V_{BG} at $V_{TG} = -5$ V. (f) The MoS₂ thickness (1.6 to 8 nm) dependence of R and D^* under the illumination power density of 53 μW cm⁻² (solid sphere) and 1.55 mW cm⁻² (open circle). Schematic illustrating generated electron-hole pairs in the DG MoS₂ phototransistor with a positive V_{TG} bias (g) and a negative (h) V_{TG} bias in connection with interface coupling effect (ICE). Reprinted with permission from ref. 209, copyright © Wiley.



(ICE) phenomenon in dual-gated (DG) MoS₂ phototransistors. Both negative top-gate (TG) voltage (V_{TG}) as well as the positive back-gate (BG) voltage (V_{BG}) is simultaneously applied to the MoS₂ channel in order to facilitate the trapping of photo-generated holes in the depleted region beneath top-gate. In fabricating DG MoS₂ phototransistors, two metal gates were placed on the top and bottom of a MoS₂ layer. Fig. 11 shows a 3D schematic and optical microscopic image of a DG MoS₂ phototransistor, photoresponsivity (R) of DG MoS₂ phototransistor as a function of V_{BG} under different V_{TG} with illumination power density (P_{in}) of 1.55 mW cm⁻² at 550 nm wavelength, R_{max} and detectivity (D^*) as a function of applied V_{TG} , P_{in} dependent R with different V_{BG} , MoS₂ thickness dependent R and D^* values and schematic illustration of ICE in the DG MoS₂ phototransistor with positive and negative V_{TG} biases. The DG MoS₂ photodetector, having varied thickness between 2.5 nm to 6.5 nm, showed the highest photoresponsivity of 7.7×10^5 A W⁻¹ and detectivity of 1.9×10^{14} Jones under illumination power density (P_{in}) of 53 μ W cm⁻² at 550 nm under $V_{TG} = 5.0$ V and $V_{BG} = 2.0$ V. The response and recovery time of the DG MoS₂ photodetector decreased from 11.9 s to 8.3 s and 76.2 s to 46.4 s as the V_{TG} decreased from -4 V to -6 V, respectively. The maximum photoresponsivity and detectivity of BG-MoS₂ FETs were found to be 7×10^3 A W⁻¹ and 9.6×10^{12} Jones, respectively. DG MoS₂ photodetectors showed photoresponsivities in 10^4 to 10^5 A W⁻¹ range independent of the MoS₂ thickness; however, photoresponsivity decreased as the thickness of the MoS₂ channel was less than 2.5 nm or more than 6.5 nm. DG MoS₂ photodetectors having 2.5 and 6.5 nm thickness exhibit a strong ICE under applied negative V_{TG} and positive V_{BG} . The photoresponse speed of the DG MoS₂ phototransistors can be adjusted with ICE. This study showed that the interface coupling effect in the DG MoS₂ phototransistors improves the photoelectric performance of photodetectors.

Flexible photodetectors for wearable applications have been developed from a wide variety of nanomaterials, and their performance is controlled by many conditions, including the material synthesis, atomic layer processing, hybrid heterostructures, device fabrication, measurement wavelengths and incident laser power.^{64,87,208–214} The performance of MoS₂-based photodetectors, such as the detectivity, photoconductive gain, response time and broadband responsivity depends on several factors, including the number of active layers, processing, nature of doping materials, vdW interactions, barrier and junction heterostructures, transport, photocarrier generation and spectral range.²¹⁵ Additionally, ferroelectric polymers, such as poly(vinylidene fluoride) (PVDF), poly(vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE)] copolymer and poly(vinylidene fluoride-trifluoroethylene-chlorofluoroethylene) [P(VDF-TrFE-CFE)] terpolymer are unique organic polymers that have been used in flexible electronics because they offer high piezoelectricity, pyroelectricity and ferroelectricity, tremendous mechanical strength and flexibility to integrate into devices such as transducers, sensors, actuators, nonvolatile ferroelectric random-access memory (NV-FeRAM) and ferroelectric FETs.²¹⁶ Additionally, in the case of MoS₂ photodetectors, these ferroelectric polymers help suppress the dark

current, which ultimately improves the photoresponse of photodetectors.¹³⁷ MoS₂-based flexible photodetectors are discussed in the next section.

4. MoS₂ based flexible photodetectors for wearable technology

Similar to graphene, applications of flexible atomic layered TMDs have been explored for wearable technologies, from electronics to optoelectronics.³⁶ Flexible photodetectors are expected to be important elements of wearable optoelectronic technology in fields from biomedical imaging to surveillance and high-speed broadband optical communication. Flexible photodetectors have been developed using a variety of nanomaterials, from traditional semiconductors, such as Si, Ge, Se, GaP, InP, CdS and ZnSe,^{42–49} to newly emerging perovskites,^{75,76} as a very wide variety of hybrid heterostructures involving polymers, metal oxides, CNTs, graphene, transition metal trichalcogenides, *etc.*^{77–89} Similarly, flexible MoS₂ atomic layers and MoS₂-based hybrid heterostructures have been explored for fabricating photodetectors for the next generation of wearable technologies. The progress made in developing flexible MoS₂ photodetectors is summarized in this section.

Flexible polymeric substrates based on poly(ethylene terephthalate) (PET), Kapton/polyimide (PI), poly(methylmethacrylate) (PMMA), poly(ethylene naphthalate) (PEN), poly(dimethylsiloxane) (PDMS) and poly(4-vinylphenol) (PVP) have been generally used for developing wearable electronic and optoelectronic devices due to their inherent mechanical strength and flexibility.^{23,25,36} In addition, textile fibers and papers have also been considered for developing flexible devices. Zhang *et al.*²¹⁷ reported substrate-dependent performance of MoS₂ photodetectors owing a dual-photogating effect where rigid silicon carbon (SiC) and flexible Kapton (polyimide) films were used as substrates for developing MoS₂ photodetectors. Both SiC and Kapton have strong light absorption, therefore, the dual-photogating effect induced at the interface of the MoS₂/SiC or MoS₂/Kapton assists in enhancing the photoresponse of MoS₂-based photodetectors. Fig. 12 shows the schematic illustrations of MoS₂ photodetectors fabricated on updoed SiC and Kapton substrates, their energy level diagrams and the variation of photoresponsivity (R) and on/off ratio (I_{photo}/I_{dark}) of rigid MoS₂/SiC and MoS₂/SiO₂/Si photodetectors as well as flexible MoS₂/Kapton and MoS₂/PET photodetectors as a function of optical power intensity in the UV and visible wavelengths. The bandgap of 2.7 and 3.2 eV and the electron affinity of 4.2 and 1.8 eV for MoS₂ and SiC are depicted, respectively. The energy level mismatch at the MoS₂/SiC interface yields to a strong built-in electric field (E_{in}) which assists in separating the photoexcited carriers in MoS₂. The enhanced photoresponse was noticed for the MoS₂/SiC photodetectors more than those of MoS₂/SiO₂/Si photodetectors. The energy band diagram in a flexible MoS₂/Kapton photodetector shows the valence band of MoS₂ as -6.0 eV and conduction band as -4.2 eV and the HOMO level at -5.0 eV and LUMO level at -2.1 eV, *versus* vacuum. The highest photoresponsivity of $\sim 10^4$ A W⁻¹ for MoS₂/SiC photodetector and the



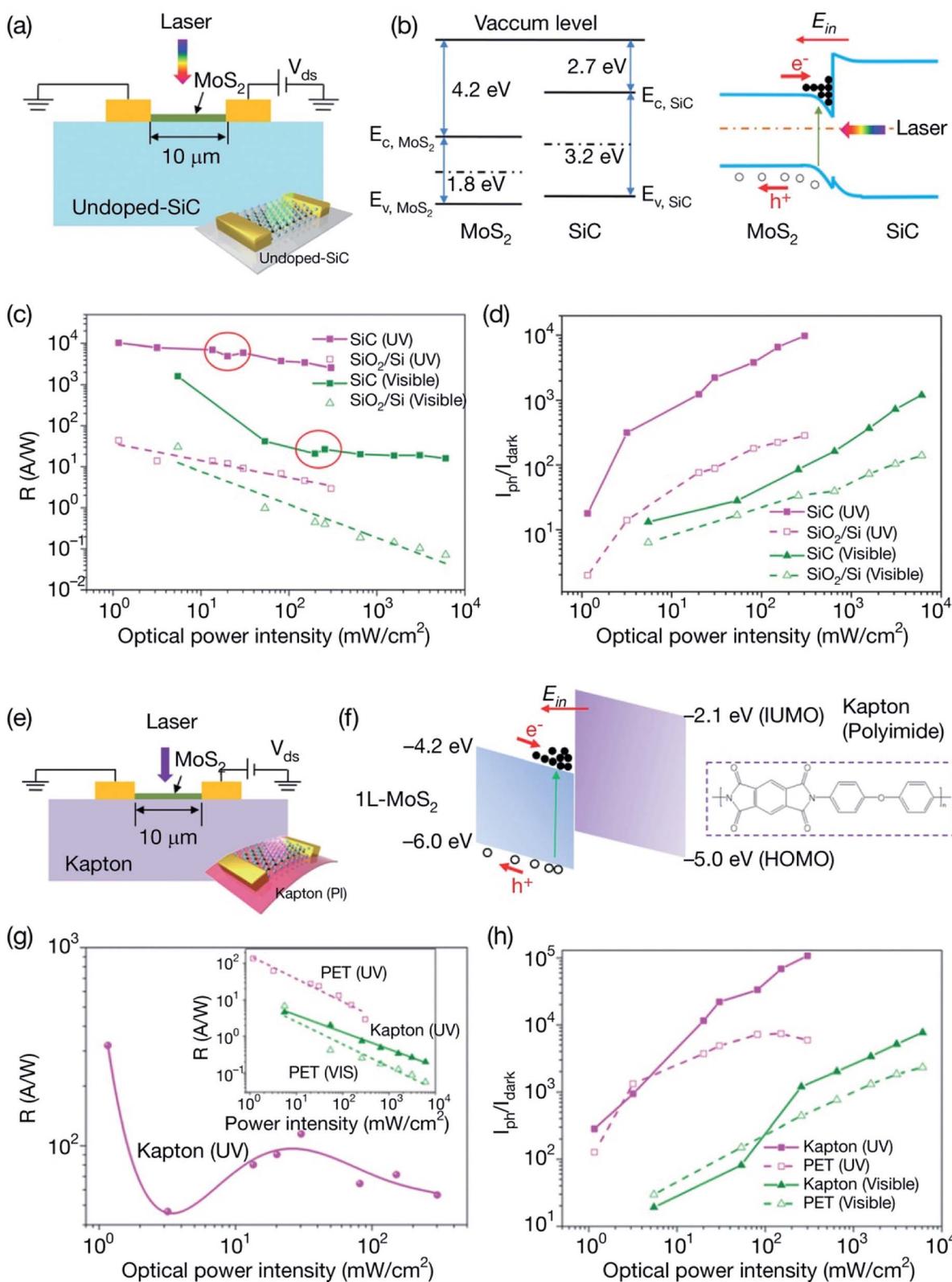


Fig. 12 (a) Schematic illustration of MoS₂ photodetector on a SiC substrate. (b) Energy level diagrams of the MoS₂ and SiC showing electron affinity and bandgap of MoS₂ and SiC. MoS₂/SiC interface showing a band structure. (c) Variation of photoresponsivity (R) and (d) on/off ratio (I_{photo}/I_{dark}) of rigid MoS₂/SiC and MoS₂/SiO₂/Si photodetectors as a function of optical power intensity in the UV and visible wavelengths. (e) Schematic illustration of MoS₂ photodetector on a flexible Kapton substrate. (f) Energy band diagram of a flexible MoS₂/Kapton photodetector. (g) Variation of photoresponsivity (R) and (h) on/off ratio (I_{photo}/I_{dark}) of flexible MoS₂/Kapton and MoS₂/PET photodetectors as a function of optical power intensity in the UV and visible regions. Reprinted with permission from ref. 217, copyright © Royal Society of Chemistry.



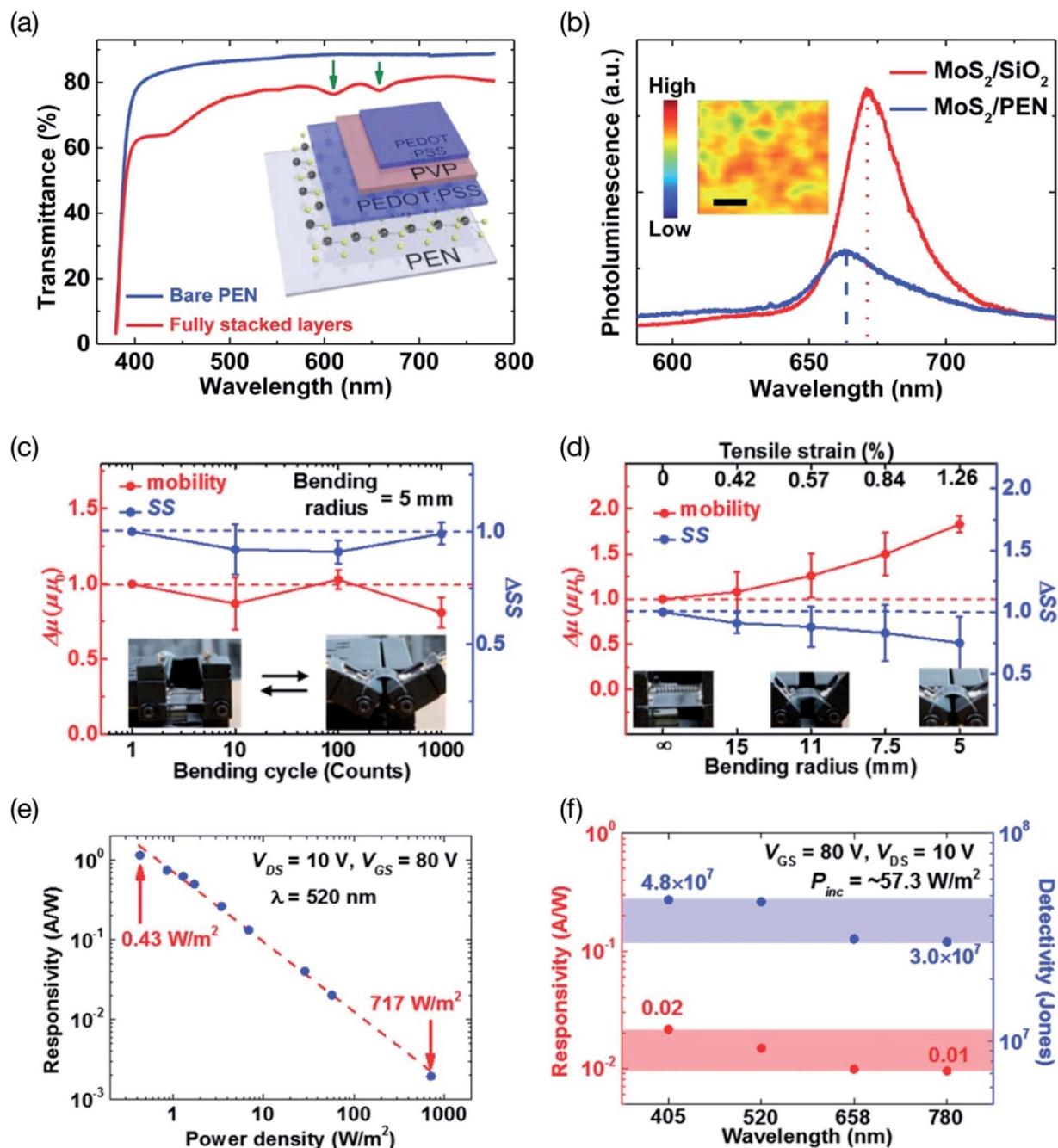


Fig. 13 (a) Transmittance spectra of the flexible bare PEN substrate, denoted by the blue line, and fully stacked PEDOT:PSS/PVP/PEDOT:PSS/MoS₂/PEN layers, denoted by the red line. The inset is a schematic of the phototransistors with stacked layers. (b) PL spectra of CVD-grown monolayer MoS₂/SiO₂/Si and MoS₂/PEN. The inset is PL intensity mapping at 670 nm (1.85 eV). (c) The variation in mobility and SS values as a function of the number of bending cycles for a 5 mm bending radius, and (d) mobility and SS values at different bending radii of 5, 7.5, 11, 15 mm, and ∞. (e) Variation of photoresponsivity MoS₂ phototransistors as a function of power density at 520 nm laser wavelength. (f) Photoresponsivity and photodetectivity of transparent MoS₂ phototransistors at different laser wavelengths. Reprinted with permission from ref. 219, copyright © American Chemical Society.

fastest current I_{on}/I_{off} ratio of $\sim 10^5$ for MoS₂/Kapton photodetector were measured at 325 nm laser illumination due to the dual-photogating effect generated at the MoS₂/substrate interface. SiC and Kapton substrates improve the photoresponse of MoS₂ photodetectors due to the light absorption.

Transparent poly(4-vinylphenol) (PVP) as the encapsulation layer, as well as a gate dielectric was used by Ma *et al.*²¹⁸ for

developing flexible MoS₂ phototransistors on a polyarylate substrate. PVP thin film having $\geq 80\%$ transmittance between 400 nm to 900 nm was used. The PVP encapsulation not only increased the mobility but also improved the current I_{on}/I_{off} ratio and subthreshold slope. The photoresponsivity (2 A W^{-1}), as well as the detectivity (3.1×10^{12} Jones) of MoS₂ phototransistors, was increased by two orders of magnitude over the



visible to IR range due to the doping effect of hydroxyl groups in PVP. For a comparison, the transparent PVP polymer improved the photoresponsivity of MoS₂-based flexible phototransistor devices by ten fold over that of PMMA encapsulated devices. The photodetectors developed from monolayer and bilayer MoS₂ encapsulated with a HfO₂ layer showed photoresponsivity in the wide range of 10–10⁴ A W⁻¹ and a response time from 10 ms to 10 s.¹³⁵ These studies show the role of encapsulation in increasing the photoresponse of MoS₂-based photodetectors.

Transparent and flexible MoS₂ phototransistor arrays using all-organic components with inkjet-printing technology were developed by Kim *et al.*²¹⁹ The CVD-grown monolayer MoS₂ channel layers were transferred onto a flexible PEN substrate. The all-organic components, including poly(3,4-ethylenedioxythiophene)polystyrenesulfonate (PEDOT:PSS)-based source/drain electrodes, PVP as a gate dielectric layer, and PEDOT:PSS as a top-gate electrode, were consecutively inkjet printed on the monolayer MoS₂/PEN substrate. The formation of CVD-grown monolayer MoS₂ was confirmed by Raman spectroscopy, XPS, scanning transmission electron microscopy (STEM) coupled with energy dispersive X-ray spectroscopy (EDS), PL spectroscopy, and optical transmittance spectroscopy as demonstrated in Fig. 13(a and b). The optical transmittance of the inkjet-printed MoS₂ phototransistors was 76%, compared with 87% for the PEN substrate in the same visible wavelength range. The performance of printed MoS₂ phototransistors was not degraded under 1000 repeated bending cycles, as shown by the mobility and SS values for a fixed 5 mm bending radius, which corresponds to 1.26% uniaxial tensile strain along the MoS₂ channel length, demonstrating good mechanical stability as shown in Fig. 13(c and d). The mobility and SS values of MoS₂ phototransistors measured at bending radii of 15, 11, 7.5, and 5 mm increased as the bending radius decreased. Fig. 13(e and f) shows the photoresponsivity and photodetectivity of transparent MoS₂ phototransistors as a function of wavelength over the 405–780 nm region at a fixed laser power density of 57.3 W m⁻², a V_{gs} of 80 V, and a V_{ds} of 10 V. The photoresistivity, detectivity and EQE of MoS₂ phototransistors showed wavelength dependence, varying between 0.01 and 0.02 A W⁻¹, 3.0 and 4.8 × 10⁷ Jones, and 1.5 and 6.6%, respectively.

Flexible photodetectors obtained by depositing MoS₂ layers on a PI substrate were developed by Lim *et al.*²²⁰ The MoS₂ layers were characterized in terms of their uniformity, stoichiometry and structural homogeneity by Raman, AFM, and XPS techniques. The frequency difference of 23.4 cm⁻¹ between the E_{2g}⁻¹ and A_{1g} Raman vibration modes of the MoS₂ layers indicated few-layer MoS₂ thin films. TEM showed the formation of five-layer MoS₂ films and an interlayer spacing of 6 Å. The optical transmittance was 90.8% at 550 nm for the MoS₂ films deposited on a glass substrate. MoS₂ FETs showed a mobility of 14 cm² V⁻¹ s⁻¹ and an on-off current ratio of 5 × 10². The MoS₂ photodetector arrays were also fabricated on a 4-inch SiO₂/Si wafer. Fig. 14 shows a photograph of the MoS₂ layer on a flexible PI film, XPS spectra, time-dependent photocurrents before and after bending up to 10⁵ bending cycles, and the change in the photocurrent of MoS₂-based flexible photodetectors as a function of the bending cycle at 20 V. The visible-light

photodetectors exhibited a response time of 13 s and a recovery time of 30 s. MoS₂ photodetectors were formed on a flexible PI substrate, and the photocurrent decreased by 5.6% after 10⁵ bending cycles at a bending radius of 5 mm.

Enhancement of the photocurrent and sensitivity has been observed for flexible p-CuO/n-MoS₂ heterojunction-based photodetectors.²²¹ The dark current of CuO/MoS₂ heterojunction increased from 0.039 nA to 0.12 nA as the tensile strain was increased from 0% to 0.65%, whereas the photocurrent increased to 108 nA under 0.65% tensile strain with 1656 mW cm⁻² light intensity, which is 2770-fold increase in the photocurrent over that of the dark current. The photocurrent of the p-CuO/n-MoS₂ heterojunction increased 27 times under 0.65% tensile strain compared with that of the strain-free heterojunction at 532 nm, and the sensitivity reached 3.27 × 10⁸ Jones. The photoresponse of a flexible CuO/n-MoS₂ heterojunction based photodetector was increased by the piezophototronic effect. Zhang *et al.*²²² fabricated a MoS₂ phototransistor on a transparent and flexible biodegradable paper substrate using a gel electrolyte gate dielectric. The gel electrolyte thin layer was coated on top of mechanically exfoliated MoS₂ flakes as the gate dielectric. The optical transmittance of the MoS₂ phototransistor at 550 nm was 82%, compared with 85% for the bare nanopaper, due to the added passivation layer and gel electrolyte gate dielectric. The MoS₂ phototransistor exhibited a photoresponsivity of 1.5 kA W⁻¹ at an illumination power of 10 nW. The MoS₂ phototransistor showed high flexibility and optical transparency with great photoresponsivity.

The solution-processed few-layer MoS₂ thin films on several flexible substrates, including PI, Al, and Cu foils, cotton thread, and cellulose, carbon, and ceramic paper were developed by Sahatiya *et al.*²²³ The growth of layers and morphologies of the MoS₂ nanosheets varied depending on the substrate. The photodetectors fabricated from these MoS₂ nanosheets showed that the defects introduced during growth control the photocurrent response. Sahatiya *et al.*²²⁴ also fabricated a broadband flexible photodetector using 1D V₂O₅ NWs and 2D MoS₂ flakes covering the UV (365 nm) to NIR (780 nm) region, wherein V₂O₅ NWs absorb in the UV-visible, and MoS₂ absorbs in the visible–NIR regions. The V₂O₅/MoS₂ hybrid device showed a responsivity of 41.5 mA W⁻¹ in the UV, 65.1 mA W⁻¹ in the visible, and 29.4 mA W⁻¹ in the NIR. The same research team²²⁵ also developed a flexible photodetector using MoS₂/C QD hybrid films. The wide UV-vis-NIR absorbance range originated from the UV absorbance of the C QDs and the visible–NIR region absorbance of MoS₂. The MoS₂/C QD sensor showed photoresponsivities of 2.62, 8.4, and 18.12 mA W⁻¹ in the NIR, UV, and visible regions, respectively. The MoS₂/C QD sensor showed almost no change in photoresponse up to 500 bending cycles. These flexible MoS₂/C QD broadband photodetectors can be used for wearable electronics for surveillance purposes.

4.1 MoS₂/graphene hybrid heterostructures for flexible photodetectors

The indirect bandgap restricts the photodetection ability of multilayer MoS₂. Hybrids of MoS₂ atomic layers with gapless



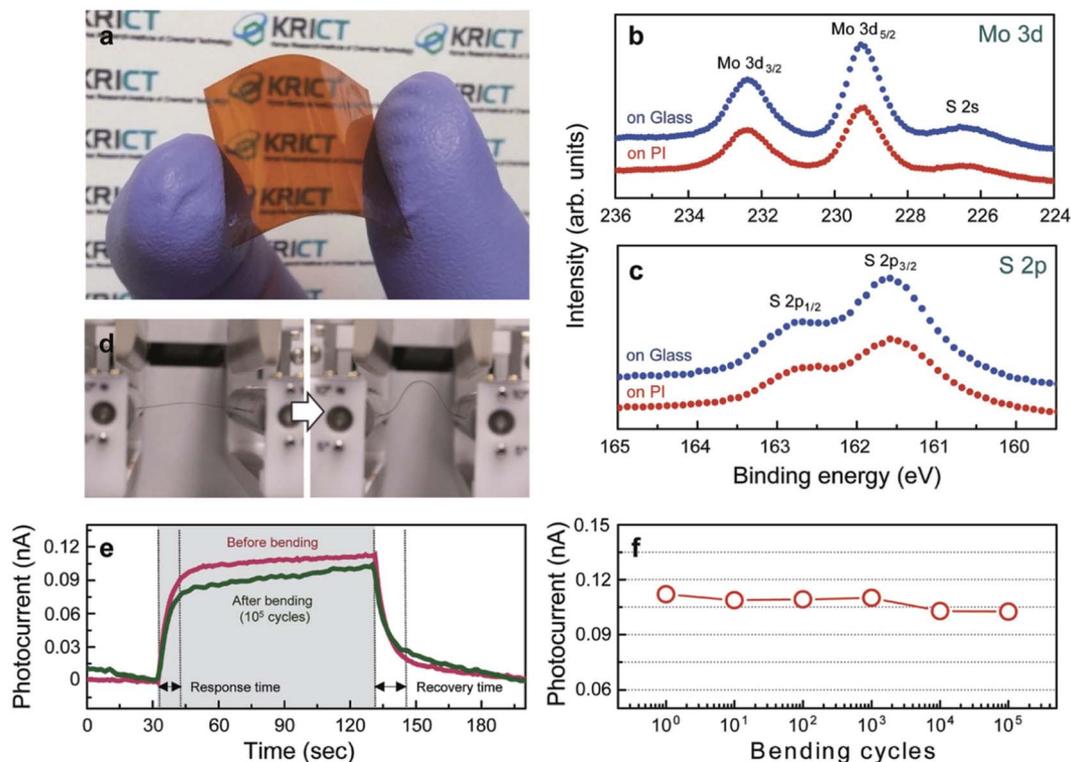


Fig. 14 (a) Photograph of the MoS₂ layer on a flexible PI film. (b) XPS spectra showing the Mo 3d core level and (c) S 2p core level spectra of MoS₂ layers on glass and PI substrates. (d) Bending of a flexible MoS₂ layer deposited on a PI substrate at a 5 mm bending radius. (e) Time-dependent photocurrents of MoS₂-based flexible photodetectors measured at an incident power of 12.5 mW cm⁻² and $V = 20$ V, before and after bending up to 10⁵ bending cycles. (f) The variation in the photocurrent of the MoS₂ photodetector as a function of the bending cycle. Reprinted with permission from ref. 220, copyright © 2016 Wiley-VCH.

graphene have been used to improve the optoelectronic properties by utilizing the high carrier mobility, ultrafast response and broadband absorption of graphene and the generation of electron-hole pairs by MoS₂. Ross *et al.*¹²⁶ obtained a high carrier mobility (1×10^4 cm² V⁻¹ s⁻¹) in a graphene/MoS₂ hybrid structure by placing monolayer graphene on a multilayer MoS₂ nanosheet on a Si/SiO₂ substrate. The graphene/MoS₂ hybrid-based photodetector showed high photoresponsivities of 1×10^{10} A W⁻¹ at 130 K under an LED power of 1 fW μm⁻² and of 5×10^8 A W⁻¹ at room temperature under an LED power of 6.4 fW μm⁻² and gate-tunable photoconductivity with a high photodetection capability. Zhang *et al.*²²⁶ used CVD-grown monolayer MoS₂ to develop a graphene/MoS₂ hybrid structure-based phototransistor that showed a photoresponsivity of 1.2×10^7 A W⁻¹ at 650 nm under a light power of 0.01 W m⁻² and a high photogain of 10⁸. A photoresponsivity of 3.34×10^3 A W⁻¹ and a detectivity of 1.004×10^{12} Jones have been observed for graphene/MoS₂ hybrid structure-based FETs.²²⁷ The phototransistor fabricated from multilayer MoS₂ using graphene source/drain electrodes showed a high photoresponsivity of 1×10^4 A W⁻¹ in the wavelength range of 470–600 nm.²²⁸

Chen *et al.*²²⁹ developed n-n-type vdWHs using multilayer MoS₂ coated with a layer of nitrogen-doped graphene QDs (N-GQDs) to complement each other with their strong characteristics to improve the carrier mobility. In this hybrid structure,

MoS₂ was used to generate photoexcited holes, while the GQDs working as a gain material contributed to the recirculation of photoexcited carriers at the heterojunction interface. Additional photocarriers were also created by the reabsorption of light emitted from the GQDs by MoS₂. Fig. 15 shows a schematic of the MoS₂/GQD heterostructure-based phototransistor, a comparison of the drain current (I_d) versus source-drain voltage (V_{ds}) curves of pristine MoS₂ and MoS₂/GQD hybrid phototransistor devices in the dark and under light illumination, and the photoresponse speed and incident light power-dependent photoresponsivity of MoS₂/GQD vdWH-based phototransistors. In the case of the MoS₂ phototransistor, the drain current increased to 10.15 μA at a 405 nm wavelength under an incident laser power of 30.1 μW, with a source-drain voltage (V_{ds}) of 1.68 V; this drain current was 1000 times larger compared with the 11 nA current recorded in the dark state. The MoS₂ phototransistor showed a sensitivity up to 5 nW and rise/fall times longer than 50 s. The MoS₂ phototransistor showed the highest photocurrent of 16.2 μA at 50 nW and photoresponsivity of 800 A W⁻¹ at 70 V. The MoS₂/GQD hybrid phototransistor showed a significant change in the drain current (I_d) versus source-drain voltage (V_{ds}) curves, a higher drain current and a faster photoresponse speed compared with the pristine MoS₂ phototransistor. The photocurrent of the MoS₂/GQD hybrid phototransistor reached 0.55 mA at a gate bias of



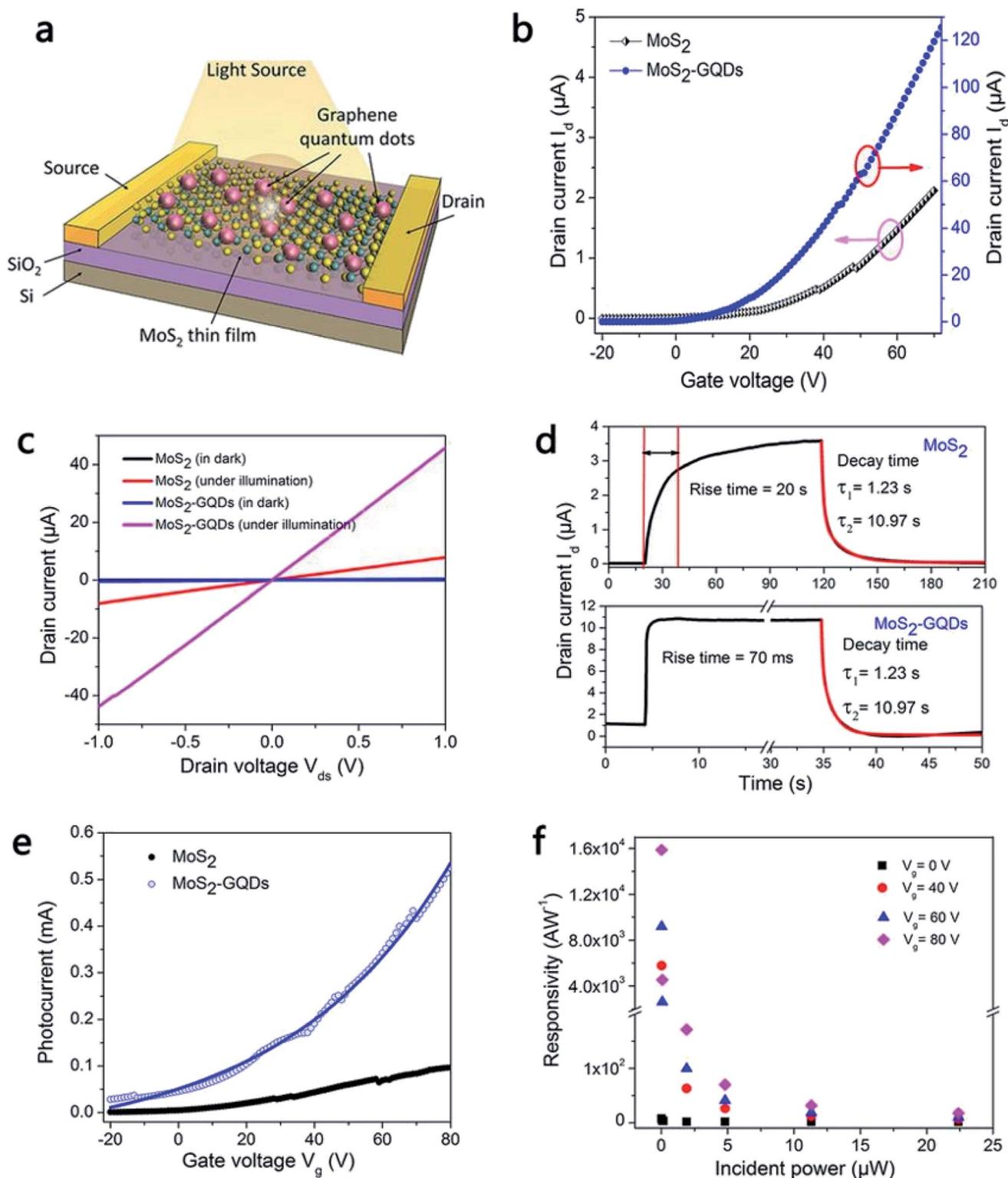


Fig. 15 (a) Schematic of the MoS₂/GQD heterostructure-based phototransistor. (b) Current vs. voltage curves of MoS₂ and MoS₂/GQD phototransistor devices. Here, I_d is the drain current, and V_{ds} is the source–drain voltage = 1 V. (c) Drain current versus source–drain voltage curves in the dark and under light illumination for MoS₂ and MoS₂/GQD phototransistors, measured at a 405 nm wavelength under an incident light power of 17 μ W. (d) Photoresponse times of MoS₂ (top) and MoS₂/GQD (bottom) phototransistors. (e) Photocurrent versus back gate voltage curves for MoS₂ and MoS₂/GQD phototransistors, measured under an incident light power of 30.1 μ W. (f) Incident light power-dependent photoresponsivity of MoS₂/GQD phototransistors. Reprinted with permission from ref. 229, copyright © Springer Nature Publishing.



80 V. The MoS₂/GQD heterostructure-based photodetector showed a photoresponsivity of $1.6 \times 10^4 \text{ A W}^{-1}$ at 80 V under an incident power of 50 nW and a photoconductive gain of 2.4×10^7 electrons per photon. The high performance of MoS₂/GQD heterostructure-based photodetectors occurs due to their increased light absorption and the effective generation of electron–hole pairs between the GQDs and MoS₂.

Large-area MoS₂ films on monolayer graphene were developed by Chen *et al.*²³⁰ by adding hydrogen gas during the CVD growth process, which resulted in centimeter-long monolayer MoS₂ continuous films on graphene. Hydrogen addition prevented the degradation of monolayer graphene by decreasing the oxidation. The MoS₂/graphene-based photodetector showed a photoresponsivity as high as 2.4 A W^{-1} under 532 nm illumination and a 135 μW power density. Flexible photodetectors, consisting of an rGO fiber and MoS₂ nanosheet hybrid composite, showed significant enhancement of the photocurrent.^{231a} The Zn-doped MoS₂/rGO hybrid fiber-based flexible photodetectors showed a photoresponsivity of 5.73 A W^{-1} at laser power density of 125.2 W m^{-2} using a bias voltage of 2 V. The same research group also reported one-step synthesis for preparing Zn-doped MoS₂ photodetector using Zn(II)-tetrakis(4-hydroxyphenyl)-porphyrin [Zn(II)THPP] as a dopant loaded seeding promoter.^{231b} The Zn-doped MoS₂ thin films were transferred onto a PET substrate to fabricate flexible photodetectors. The electrical resistance of the flexible photodetector increased 13% after 10 000 bending cycles at a bending radius of 3 cm while resistance increased 10% at a bending radius of 1 cm, demonstrating good mechanical stability of the flexible photodetectors. The photocurrent of Zn-doped MoS₂ photodetectors was found to increase from 0.05 μA to 2.62 μA as the applied voltage was raised from 1 V to 40 V at a power density of 125.2 W m^{-2} and a similar trend was observed for the photoresponsivity. The photocurrent and photoresponsivity of the Zn-doped MoS₂ photodetectors can be controlled by adjusting the Zn doping concentration. Asad *et al.*²³² used MoS₂ nanoparticle (MoS₂ NP)-decorated graphene nanoribbons (GNRs) to develop flexible phototransistors. In the GNR–MoS₂NP hybrid, the GNRs function as carrier transport channels, whereas the MoS₂ NPs offer high gain absorption. The MoS₂ NPs also generate electron–hole pairs, which are separated at the GNR and MoS₂ NP

interface and result in electron transfer from the MoS₂ NPs to the GNRs. The flexible GNR–MoS₂ hybrid photodetector showed a photoresponsivity of 66 A W^{-1} , a fast rise time of 5 ms and a decay time of 30 ms under 385 nm illumination and a 2.1 μW power density. The photoresponsivity of the GNR–MoS₂ hybrid photodetector was 1.3×10^5 times larger than that of the pristine graphene photodetector and 10^4 times larger than that of the pristine MoS₂ phototransistor. The hybrid photodetector also showed high stability for a 6 mm bending radius. De Fazio *et al.*²³³ developed visible-light flexible photodetectors using CVD-grown single-layer graphene (SLG) and single-layer (1L) MoS₂ on flexible PET substrates. Fig. 16 is a schematic of a flexible SLG/1L MoS₂ photodetector on a flexible PET substrate and a photograph of the photodetector showing the optical transparency and flexibility. The polymer electrolyte-gated flexible SLG/1L MoS₂ photodetector showed an external photoresponsivity of 45.5 A W^{-1} and an internal photoresponsivity of 570 A W^{-1} at 642 nm, a photoconductive gain of 4×10^5 , and >82% optical transparency. The flexible photodetectors showed mechanical durability upon bending at a 1.4 cm bending radius. The photoconductive gain increased by nearly one order of magnitude from 0.1 to 1.0 V, and the photoresponsivity reached 45.5 A W^{-1} at a V_{ds} of 1 V. Therefore, the photoconductive gain of the SLG/1L MoS₂-based heterostructure photodetector being four orders of magnitude larger than that of the 1L MoS₂ photodetector developed without any SLG showed that the SLG/1L MoS₂ heterostructure is important for high photoresponsivity.

Photodetector devices using a MoS₂/graphene hybrid pattern formed by the soft-lithographic patterning technique were reported by Kang *et al.*²³⁴ The cross-stacked MoS₂/graphene patterns were transferred onto a flexible PET substrate for fabricating flexible photodetectors. Fig. 17 shows a schematic of the formation of cross-stacked MoS₂/graphene patterned nanostructures. The various fabrication steps involve the CVD growth of MoS₂ nanosheets on a 5,10,15,20-tetrakis(4-hydroxyphenyl)-21H,23H-porphyrin (p-THPP) promoter layer, transfer and patterning of the MoS₂ nanosheets using a PDMS mold, CVD growth of a graphene layer on Cu foil, transfer of the graphene layer onto a 300 nm thick SiO₂ substrate, and transfer of the graphene layer onto the top of the MoS₂ patterns by using

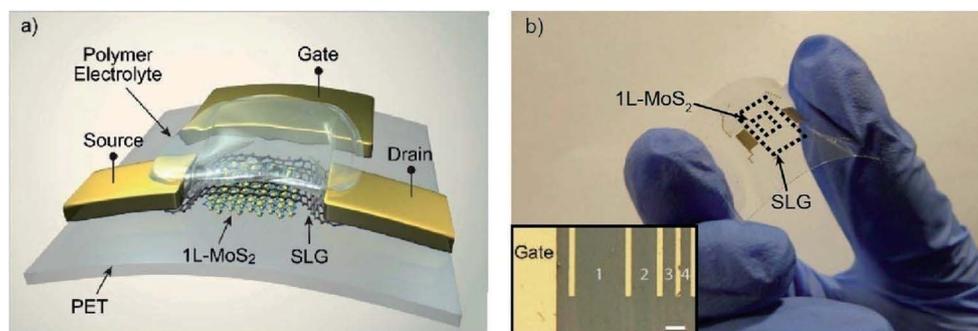


Fig. 16 (a) Schematic of the single-layer graphene (SLG)/1L MoS₂ photodetector on a flexible PET substrate. (b) Photograph of the flexible photodetector showing optical transparency. The inset is an optical image of 4 photodetectors with different channel lengths. Reprinted with permission from ref. 233, copyright © 2016 American Chemical Society.



a dimethyl sulfoxide (DMSO)-coated PDMS mold—the same process that was eventually employed for the transfer and patterning of the MoS₂ film. The SEM images confirmed the cross-stacked MoS₂/graphene line patterns with a width of 7 μm. The Raman peak difference between the E_{2g}¹ and A_{1g} vibration modes of MoS₂ was estimated to be 20 cm⁻¹, which indicated the formation of monolayer MoS₂ patterns.

The photoresponsivities for cross-stacked MoS₂/graphene hybrid photodetectors containing monolayer, bilayer, and trilayer MoS₂ films increased linearly as a function of the bias voltage from (10 to 50) V. The trilayer MoS₂ film-based hybrid photodetector showed the highest photoresponsivity compared with the monolayer and bilayer MoS₂ films, a value of 5 A W⁻¹ at a bias voltage of 2 V. Fig. 18 shows optical microscope images of cross-stacked patterns of MoS₂/graphene based on MoS₂ patterns formed by the soft-lithographic patterning technique, photographs of MoS₂/graphene hybrid patterns on a flexible PET substrate before and after the bending test, and the photoresponsivity of flexible MoS₂/graphene hybrid photodetectors as a function of the number of bending cycles at a bending radius of 9 mm and a 1.0 V bias. The mechanical stability of cross-stacked patterns of graphene/MoS₂ photodetector devices fabricated on flexible PET substrates was evaluated under various bending cycles. The photocurrent of the MoS₂/graphene hybrid photodetector with MoS₂ patterns 10 μm in width was measured between 1 and 10 000 bending cycles. The photocurrent of the device decreased from 4.3 to 3.7 A as a function of the number of bending cycles. The smallest reduction in the photoresponsivity

with increasing number of bending cycles was observed for the 10 μm wide MoS₂ patterns due to the lower compressive stress. Graphene has also been used in developing flexible photodetectors with atomic layered materials. Graphene/tin monosulfide (SnS) hybrid structure-based photodetectors further substantiated that a high photoresponsivity of 1.7 × 10⁴ A W⁻¹ under an optical light intensity of 1.2 mW m⁻² can be achieved in flexible graphene/SnSe/graphene-based photodetectors and demonstrated no change in photoresponse after 25 bending cycles, indicating high mechanical strength of the devices.⁶⁵

Polymer electrolytes have been used for developing flexible MoS₂ photodetectors. Sun *et al.*²³⁵ fabricated four phototransistor arrays with 100, 200, 500, and 1000 μm channel lengths in one flexible photodetector device based on a vertically stacked MoS₂/graphene thin film deposited on a PET substrate. A polymer electrolyte consisting of PEO : LiClO₄ in an 8 : 1 ratio was used to fabricate a flexible side-gated photodetector. The flexible photodetector based on the MoS₂/graphene film showed an external responsivity of 3.5 A W⁻¹ at -1 V (V_{gs}) under a 60 μW incident power. The MoS₂/graphene thin film had a transmittance of 10.5% at 520 nm, and the internal photoresponsivity of the photodetector reached 33.3 A W⁻¹. The photocurrent of the MoS₂/graphene film can be adjusted in the 0 to 300 μA range.

4.2 MoS₂/carbon nanotube hybrid heterostructures for flexible photodetectors

CNTs show large optical absorption compared with traditional semiconductors; therefore, photodetection can be further

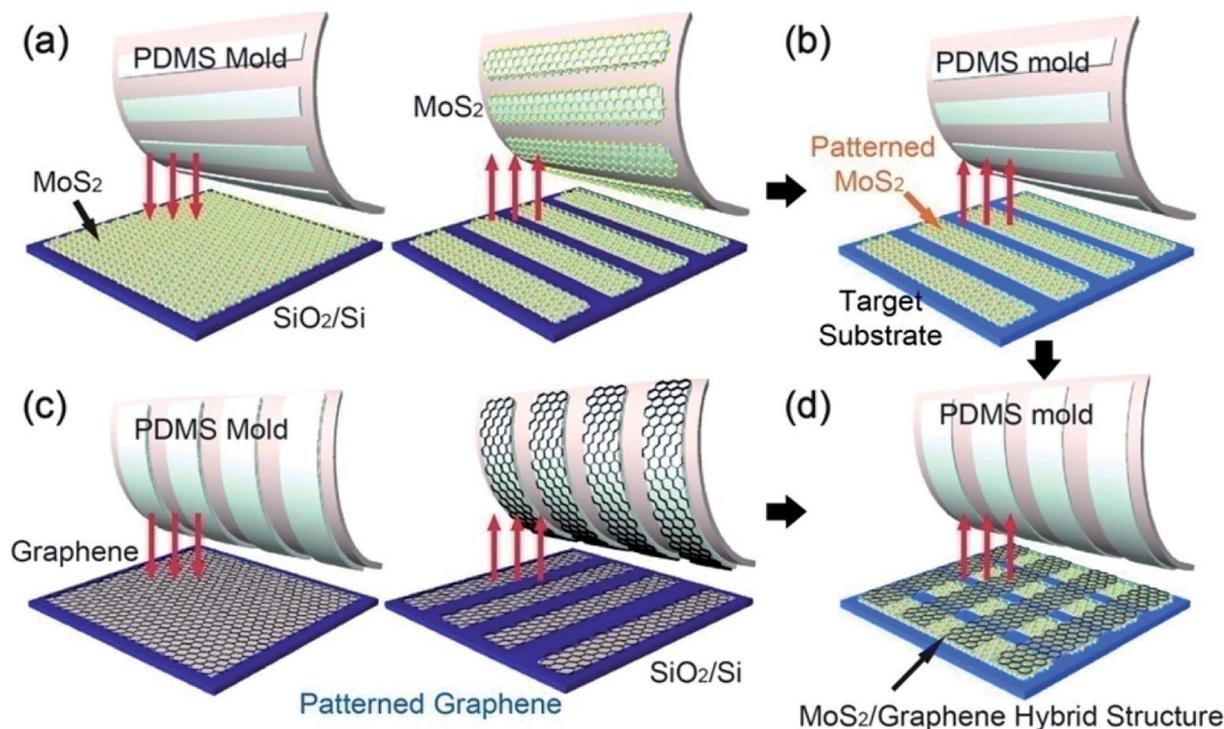


Fig. 17 Schematic of the fabrication steps for cross-stacked MoS₂/graphene patterned nanostructures. (a) CVD growth of MoS₂ nanosheets on a SiO₂ substrate (left), and pattern fabrication on the MoS₂ layer within a target area (right). (b) Transfer of MoS₂ patterns on the PDMS mold onto a target substrate. (c) CVD growth of a graphene layer. (d) Transfer of graphene patterns on the PDMS mold to the top of MoS₂ patterns on the target substrate. Reprinted with permission from ref. 234, copyright © Elsevier.



improved by using CNTs in hybrid heterostructures.^{236,237} CNT-based hybrid broadband photodetectors have been fabricated with a variety of functional materials, including Si, GaAs, fullerenes, graphene, ZnO, PbS, and perovskites.^{238–249} A negative photoresponsivity on the order of 10^8 A W⁻¹ has been reported in fullerene-sensitized aligned CNTs at room temperature.²⁵⁰ Highly flexible photodetectors based on CNTs have been demonstrated for wearable technologies.^{251–254} The outstanding electronic and optical properties have been utilized by integrating CNTs with MoS₂ atomic layers to develop flexible CNT/MoS₂ hybrid broadband photodetectors.

Photodetectors have been developed using few-layer MoS₂/SWCNT hybrids,²⁵⁵ which showed high photoresponsivities of 100 to 1000 A W⁻¹ at a 0.1 V bias voltage in the 500–700 nm visible range. Specifically, the MoS₂/SWCNT hybrid structure exhibited a photoresponsivity of 300 A W⁻¹ at 532 nm under a V_g of 0 V. Large-area MoS₂, WS₂, and MoSe₂ device arrays were fabricated by Li *et al.*²⁵⁶ MoS₂ channels and MoS₂/CNT hybrid electrodes were simultaneously deposited on a CNT-patterned substrate by the CVD method. MoS₂–MoS₂/CNT devices showed ohmic contacts between MoS₂ channels and MoS₂/CNT hybrid electrodes and exhibited better mechanical stability as well as photoresponsivity than gold-contacted devices,

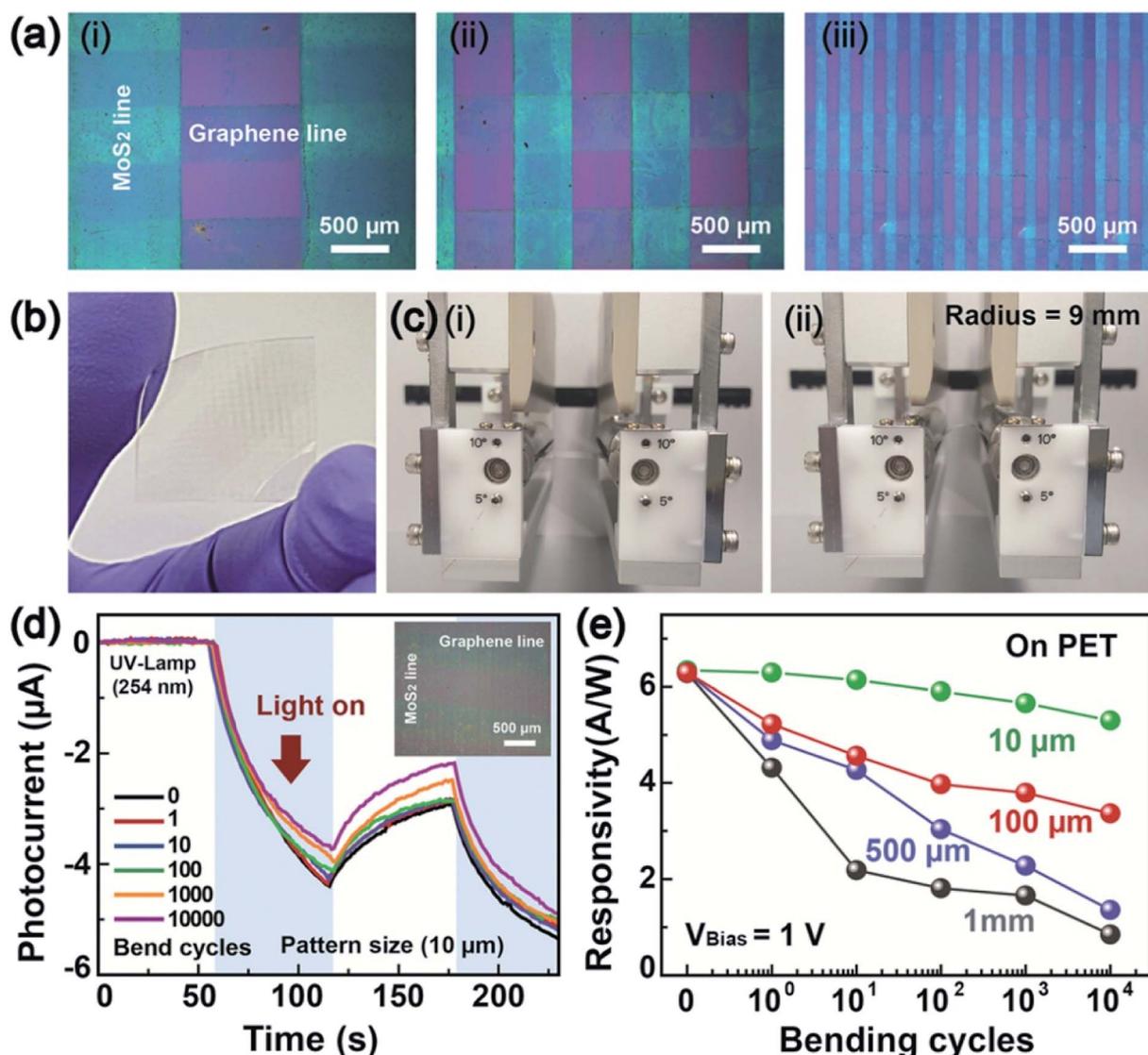


Fig. 18 (a) Optical microscope images of cross-stacked patterns of MoS₂/graphene based on MoS₂ patterns with widths of (i) 1 mm, (ii) 500 μm, and (iii) 100 μm, where the graphene patterns have a fixed width of 500 μm. (b) Photographic image of cross-linked MoS₂/graphene hybrid patterns on a flexible PET substrate developed by the soft-lithographic patterning technique. (c) Photographs of MoS₂/graphene hybrid patterns on a flexible PET substrate (i) before and (ii) after the photodetector device bending test. (d) Photocurrent as a function of time for a flexible MoS₂/graphene hybrid photodetector under 1–10 000 bending cycles. The inset shows an optical image of cross-stacked patterned MoS₂ (width = 10 μm) and graphene (width = 500 μm). (e) Photoresponsivity of flexible MoS₂/graphene hybrid photodetectors as a function of bending cycle (1–10 000), where the photodetectors have 1 mm, 500 μm, 100 μm, and 10 μm pattern sizes for the MoS₂ layers. The bending test was conducted at a bending radius of 9 mm and a 1.0 V bias voltage. Reprinted with permission from ref. 234, copyright © Elsevier.



indicating the suitability of the MoS₂-MoS₂/CNT hybrid for flexible electronic devices. Highly flexible pixel arrays based on MoS₂-MoS₂/CNT hybrid photodetectors were developed. Fig. 19 shows the flexible MoS₂-MoS₂/CNT hybrid film-based photodetector pixel array, photocurrent response at zero gate voltage, and image patterns recorded using the MoS₂-MoS₂/CNT photodetector pixel array. The flexible MoS₂-MoS₂/CNT photodetector arrays were deposited on a 3 μm thick flexible SU-8 substrate. The MoS₂-MoS₂/CNT device showed a current below 10⁻¹¹ A at a bias voltage of 0.5 V. The MoS₂-MoS₂/CNT devices showed a detectivity of 2.4 × 10¹⁰ cm Hz^{-1/2} W⁻¹. The 160 photodetector pixels formed with MoS₂-MoS₂/CNT devices were arranged in 16 columns × 10 rows. Both L- and T-shaped image patterns were recorded by the pixel array.

4.3 MoS₂/2D TMD hybrid heterostructures for flexible photodetectors

MoS₂-based vdWHs have been extensively studied for developing photodetectors, as discussed in the previous section.¹⁹⁵⁻²⁰⁶ The electronic and optoelectronic properties of MoS₂-based FETs and photodetectors have been dramatically improved by integrating atomic layered MoS₂ with other vdW structures, such as 2D graphene, h-BN and TMDs. Huo *et al.*²⁵⁷ developed a phototransistor from multilayer MoS₂/WS₂ vdWHs that

exhibited an on/off switching ratio >10⁵, an electron mobility of 65 cm² V⁻¹ s⁻¹, and a photoresponsivity of 1.42 A W⁻¹, enhanced properties compared with multilayer MoS₂ or WS₂ transistors. Xue *et al.*¹⁶⁴ used vertical heterojunction arrays of few-layer MoS₂/WS₂ to develop flexible photodetectors. HRTEM and AFM images indicated three layers of MoS₂ (~2.1 nm) and five layers of WS₂ (~3.9 nm) in the heterostructure layered films. MoS₂/WS₂ heterojunction-based FETs were fabricated on a rigid SiO₂/Si substrate. Flexible MoS₂/WS₂ heterojunction photodetector arrays were also fabricated on a PDMS substrate. Fig. 20 shows a schematic of the MoS₂/WS₂ vertical heterojunction-based phototransistor, current-voltage plot, time-dependent photocurrent of the MoS₂/WS₂ vertical heterojunction at different incident powers during laser switching on/off, photographic image of the flexible MoS₂/WS₂ vertical heterojunction photodetector arrays on a PDMS substrate and time-dependent photocurrent of the flexible MoS₂/WS₂ vertical heterojunction photodetector device on the PDMS substrate at different incident laser powers. The MoS₂/WS₂-based photodetector showed a photoresponsivity of 2.3 A W⁻¹ at a 450 nm wavelength.

In another study, Lin *et al.*²⁵⁸ fabricated flexible vdW photodiodes on flexible PET substrates using monolayer n-MoS₂/few-layer p-WSe₂ heterojunctions. The photoresponsivity of the MoS₂/WSe₂ photodiode increased by 86% at -0.62% compressive strain in the MoS₂ armchair direction due to

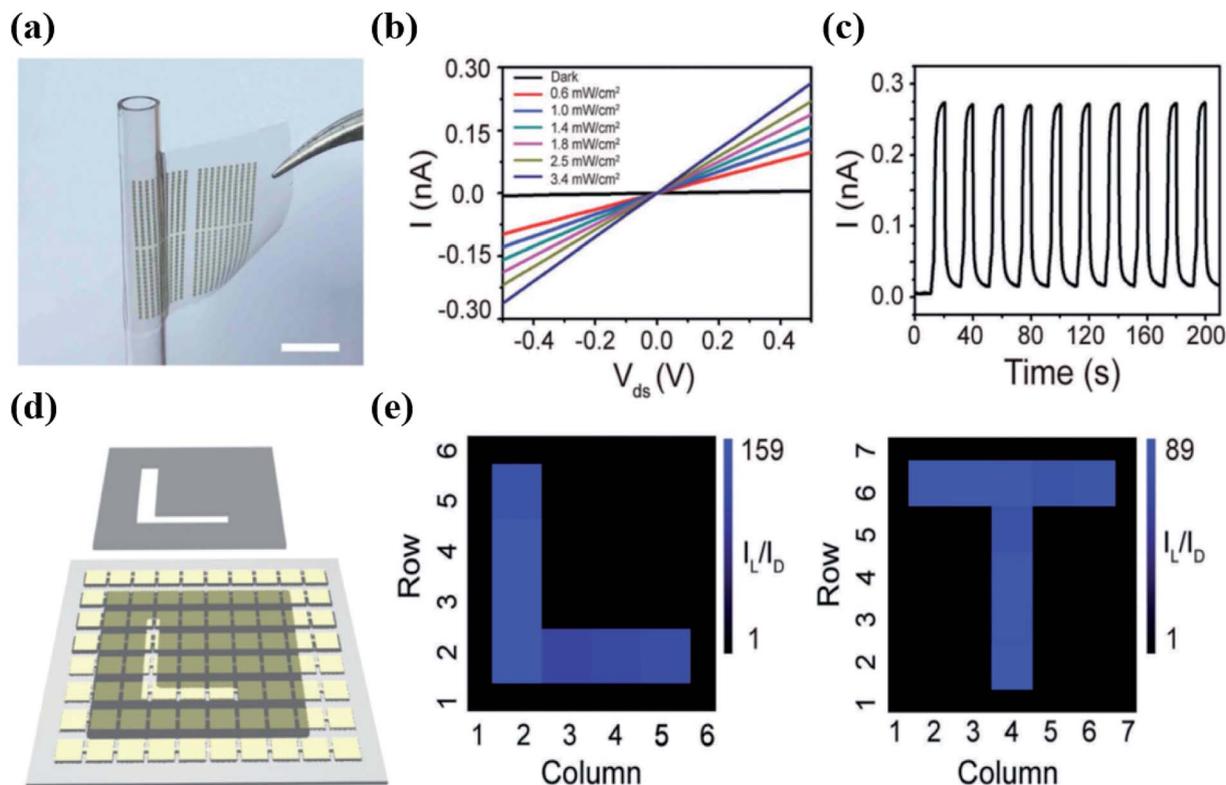


Fig. 19 Flexible MoS₂-MoS₂/CNT hybrid photodetectors. (a) Flexible MoS₂-MoS₂/CNT hybrid film-based electronic device array attached to the surface of a glass tube. (b) *I*-*V*_{ds} curves of the MoS₂-MoS₂/CNT hybrid device at 442 nm under different intensities of light illumination. (c) Photocurrent response of the MoS₂-MoS₂/CNT device as a function of time. (d) Schematic representation of a photodetector pixel array of MoS₂-MoS₂/CNT hybrid photodetectors. (e) L- and T-shaped image patterns (row vs. column) recorded using the MoS₂-MoS₂/CNT photodetector pixel array. Reprinted with permission from ref. 256, copyright © Wiley-VCH.



realignment of the energy band at the MoS₂/WSe₂ interface caused by the strain-induced piezoelectricity. The MoS₂/WSe₂ photodiode showed a photoresponsivity of 3.4 mA W⁻¹. The PL peak intensity of CVD-grown monolayer MoS₂ increased by three orders of magnitude after 15 min of TFSI chemical treatment. This study demonstrated that strain-tunable 2D TMD-based vdWHs could be used for developing optoelectronic devices. The photoresponse of flexible MoS₂/WSe₂ vdWH-based photodiodes was enhanced by the piezophototronic effect. Alloys of TMDs have been explored for developing flexible photodetectors. In another study, Wu *et al.*²⁶⁴ also demonstrated improved photoresponse of flexible monolayer MoS₂ due to the piezophototronic effect for strain-gated photodetectors. The polarization charges induced by the applied strain can change the separation of photogenerated carriers and their transport at the interface between MoS₂ and metal allowing tuning of photoresponse. The strain-induced flexible monolayer MoS₂ photodetector showed high photoresponsivity of 2.3 × 10⁴ A W⁻¹, which is 26 times larger photoresponsivity compared with

monolayer MoS₂ phototransistors¹⁷² due to the piezophototronic effect. The current (I_{ds}) monolayer MoS₂ increased by 2.5 times, from 90 nA in dark to 220 nA under 4.297 mW cm⁻² optical power intensity at a drain voltage of -2 V. The photocurrent was 40 nA at 3.4 μW cm⁻² power intensity under strain of -0.38%, which increased to 207 nA at 4.29 mW cm⁻² power intensity under increased strain of -0.45%. Zheng *et al.*²⁵⁹ developed Mo_{0.5}W_{0.5}Se₂ alloy-based photodetectors on a SiO₂/Si rigid substrate and a PI flexible substrate for photodetection over a broadband spectrum ranging from the UV to NIR region (370 nm to 808 nm). The Mo_{0.5}W_{0.5}Se₂ photodetectors on the SiO₂/Si substrate exhibited a photoresponsivity of 77.1 A W⁻¹ and a detectivity of 1.1 × 10¹² Jones with a rapid response speed of 8.3 ms, whereas the photodetector on the flexible PI substrate showed a photoresponsivity of 63.5 A W⁻¹ and a detectivity of 3.56 × 10¹² Jones. The Mo_{0.5}W_{0.5}Se₂ photodetectors showed no noticeable degradation in performance over 100 bending cycles, showing their suitability for flexible optoelectronic devices.

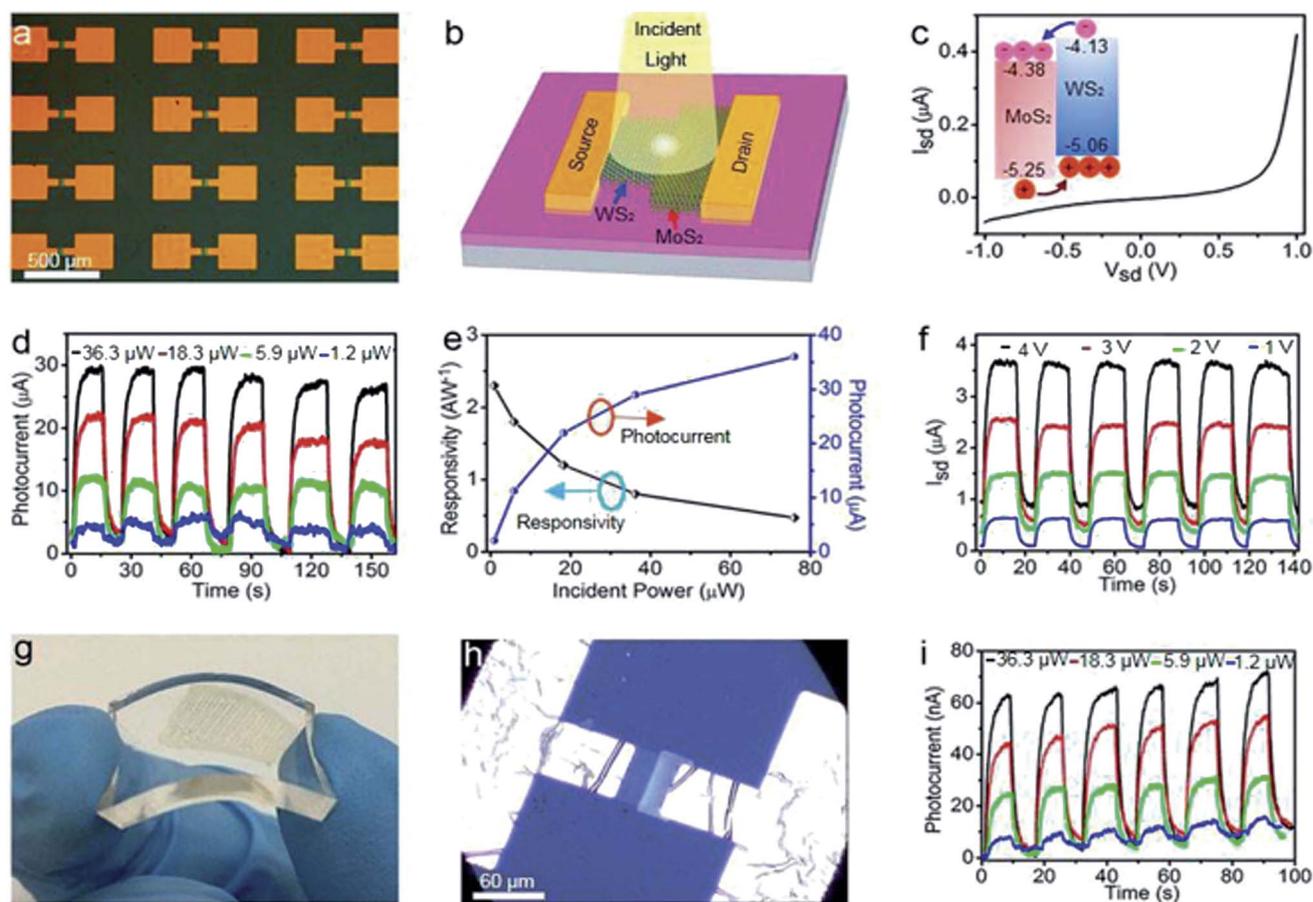


Fig. 20 (a) Vertical heterojunction arrays fabricated from few-layer MoS₂/WS₂ on a SiO₂/Si substrate. (b) Schematic illustration of the MoS₂/WS₂ vertical heterojunction-based phototransistor. (c) Current–voltage plot of a MoS₂/WS₂ vertical heterojunction-based phototransistor without illumination. The inset indicates the band alignment for few-layer MoS₂ and WS₂. (d) Time-dependent photocurrent of the MoS₂/WS₂ vertical heterojunction at different incident powers. (e) Photocurrent and photoresponsivity as a function of incident light power at a 405 nm wavelength. (f) Time dependence of the photocurrent based on the MoS₂/WS₂ vertical heterojunction photodetector during switching on/off of the laser with varying source–drain voltage (V_{sd}) from 1 to 4 V. (g) Photographic image of the flexible MoS₂/WS₂ vertical heterojunction photodetector array on a PDMS substrate. (h) Optical microscope image of a single flexible MoS₂/WS₂ vertical heterojunction photodetector device on a PDMS substrate. (i) Time-dependent photocurrent of the flexible MoS₂/WS₂ vertical heterojunction photodetector device on the PDMS substrate at different incident laser powers. Reprinted with permission from ref. 164, copyright © 2017 American Chemical Society.



4.4 MoS₂/ZnO hybrid heterostructures for flexible photodetectors

Ultra-high photoresponsivity has been observed in ZnO-based hybrid heterostructures, and flexible broadband photodetectors have been fabricated using ZnO nanostructures (NWs, nanorods, QDs, nanoparticles) combined with other functional nanomaterials, including graphene, CNTs, carbon nanodots, Au, Cu, ZnS, CdO, SnO₂, and Zn₂SnO₄, for wearable technologies.^{52–54,260–271} Hybrid heterostructures of ZnO with MoS₂ have similarly been used to develop photodetectors. The electronic and optoelectronic properties of MoS₂ atomic layers have been improved by ZnO doping. A photoresponsivity of $3.18 \times 10^3 \text{ A W}^{-1}$ and a detectivity of 5.94×10^{12} Jones have been observed for the ZnO-doped MoS₂ photodetector at a wavelength of 520 nm due to the suppression of the recombination rate of photocarriers induced by ZnO doping.²⁷² Nazir *et al.*²⁷³ used ZnO QD-decorated multilayer MoS₂ nanosheets to develop ZnO QD/MoS₂ heterostructure-based photodetectors. The field-effect mobility of ML MoS₂ increased from 5.75 to 25.09 cm² V⁻¹ s⁻¹ after decorating ML MoS₂ with ZnO QDs due to the charge transfer from the deposited ZnO QD thin layer to the surface of the pristine ML MoS₂ nanosheet. The ML MoS₂ photodetector showed a photoresponsivity of 1913 A W⁻¹ at a V_{BG} of 30 V and a V_{ds} of 1 V at 220 nm under an incident laser power of 11 mW cm⁻², which increased to 2267 A W⁻¹ under similar conditions after decorating the ML with a ZnO QD thin layer. The flexible MoS₂/ZnO/PEN photodetectors showed an I_{on}/I_{off} current ratio of 10⁴, compared with 10³ for the pure ZnO/PEN photodetector.²⁷⁴ The highest photocurrent was obtained for the device with a 5 wt% MoS₂ layer. The I_{on}/I_{off} current ratio increased from 8840 to 17 800 for 0 to 5 wt% MoS₂ in the MoS₂/ZnO hybrid structure along with the highest photocurrent and photoresponsivity values for the photodetectors. Pak *et al.*²⁷⁵ reported a photoresponsivity of 14.9 mA W⁻¹ at 1100 nm for the α-IGZO photodetector capped by a MoS₂ layer, which was five times higher compared with the pure α-IGZO photodetector with no MoS₂ layer. Yang *et al.*²⁷⁶ used few-layer MoS₂ (bandgap of 1.7 eV) with amorphous indium–gallium–zinc oxide (InGaZnO: α-IGZO) (bandgap of 3.0 eV) to develop a hybrid heterostructure-based phototransistor. The MoS₂/α-IGZO hybrid formation imparted enhanced visible light absorption and a high carrier mobility. The band alignment at the heterostructure junction facilitated the transfer of electrons generated by visible light within the upper few-layer MoS₂ to the underlying α-IGZO layer. The photocurrent of the MoS₂/α-IGZO heterostructure-based phototransistors was four orders of magnitude higher than that of the MoS₂ phototransistor at an incident laser power of 1.0 μW; the photoresponsivity was 1.7 A W⁻¹ at a wavelength of 520 nm, and the extrapolated photoresponsivity exceeded 10³ A W⁻¹ under an incident laser power of 1.0 pW. The MoS₂/α-IGZO phototransistors showed an I_{on}/I_{off} current ratio of 10⁵ and photoresponse times of 2.6 s and 1.7 s.

The morphology of nanomaterials also plays an important role in controlling the electrical properties. Lee *et al.*²⁷⁷ reported different ZnO morphological nanostructures, such as NWs, nanostars (NSs), and nanoflowers (NFs), which were

hydrothermally grown on graphene monolayer and MoS₂ multilayer films by adjusting the pH of the solution, and used these ZnO hybrid nanostructures to fabricate photodetectors. ZnO-based NWs, NSs, and NFs were obtained at pH values of 6.53, 8.18, and 9.18, respectively, and characterized by field-emission (FE)-SEM, resonant Raman spectroscopy, and XPS methods. The photodetectors fabricated from ZnO NWs/graphene, ZnO NSs/graphene, and ZnO NFs/graphene showed high photoresponsivity values of 145, 302, and 350 A W⁻¹, respectively. However, the photodetectors fabricated from ZnO NW/MoS₂, ZnO NS/MoS₂, and ZnO NF/MoS₂ hybrids showed very low photoresponsivities of 7.91×10^{-6} , 1.02×10^{-4} , and 8.99×10^{-4} A W⁻¹, respectively. The EQE of photodetectors developed from ZnO NWs, NSs, and NFs in combination with graphene were 5.14×10^4 , 1.07×10^5 , and $1.24 \times 10^5\%$, compared with 2.81×10^{-3} , 3.63×10^{-2} , and $3.19 \times 10^{-1}\%$ for ZnO NWs, NSs, and NFs with MoS₂, respectively. Fig. 21 shows a photograph of the ZnO NF/MoS₂ photodetector fabricated on a flexible PI substrate, time-dependent photocurrents of hybrid photodetectors developed from ZnO NWs, ZnO NSs, and ZnO NFs along with graphene and MoS₂, and the variation in the photocurrent of the ZnO NF/graphene hybrid and ZnO NF/MoS₂ hybrid as a function of the bending cycle. The photocurrents of the ZnO NW, NS, and NF hybrids with graphene decrease, whereas the photocurrents of the ZnO NW, NS, and NF hybrids with MoS₂ increase under UV illumination at 256 nm owing to the difference in the electronic structures of graphene and MoS₂. The photocurrent of the ZnO hybrid photodetectors shows an increasing trend of ZnO NWs < NSs < NFs, regardless of whether graphene or MoS₂ is used, because of the increased oxygen vacancies induced by adjusting the pH. The response and decay times were 10 and 67 s for the ZnO NF/graphene hybrid and 61 and 90 s for the ZnO NF/MoS₂ hybrid-based photodetector devices, respectively. The photocurrent decreased by 74% for the ZnO NF/graphene hybrid and by 76% for the ZnO NF/MoS₂ hybrid-based devices from the initial values after 10 000 bending cycles at a bending radius of 6 mm.

Flexible photodetectors have been developed using MoS₂ and graphitic carbon nitride (g-C₃N₄) hybrid thin films on a paper substrate, as reported by Velusamy *et al.*²⁷⁸ The dark current of MoS₂ decreased as the amount of g-C₃N₄ increased in the hybrid films because of the resistivity of g-C₃N₄, although the photocurrent was significantly improved due to the photocarriers generated under light illumination. The photocurrent of the MoS₂/g-C₃N₄ hybrid increased linearly as a function of increasing laser power density, yielding I_{on}/I_{off} current ratios of 4×10^3 and 1×10^4 , photoresponsivities of 700 mA W⁻¹ and 4 A W⁻¹, and detectivities of 8×10^{10} Jones and 4×10^{11} Jones at 532 nm and 365 nm wavelengths under laser power densities of 0.225 W cm⁻² and 0.5 W cm⁻² for MoS₂/g-C₃N₄ (5 : 5) hybrid thin film, respectively. The photodetectors showed stability, retaining 90% of their initial photocurrent after a time period of 3000 s. The MoS₂/g-C₃N₄ (5 : 5) hybrid film-based photodetectors showed no significant decrease in the photocurrent after bending the thin films to radii of 10, 7, 5, 4, 3, and 2 mm; and no deformation up to 400 bending cycles at a 4 mm bending radius under the measurement conditions described above. Yu *et al.*²⁷⁹



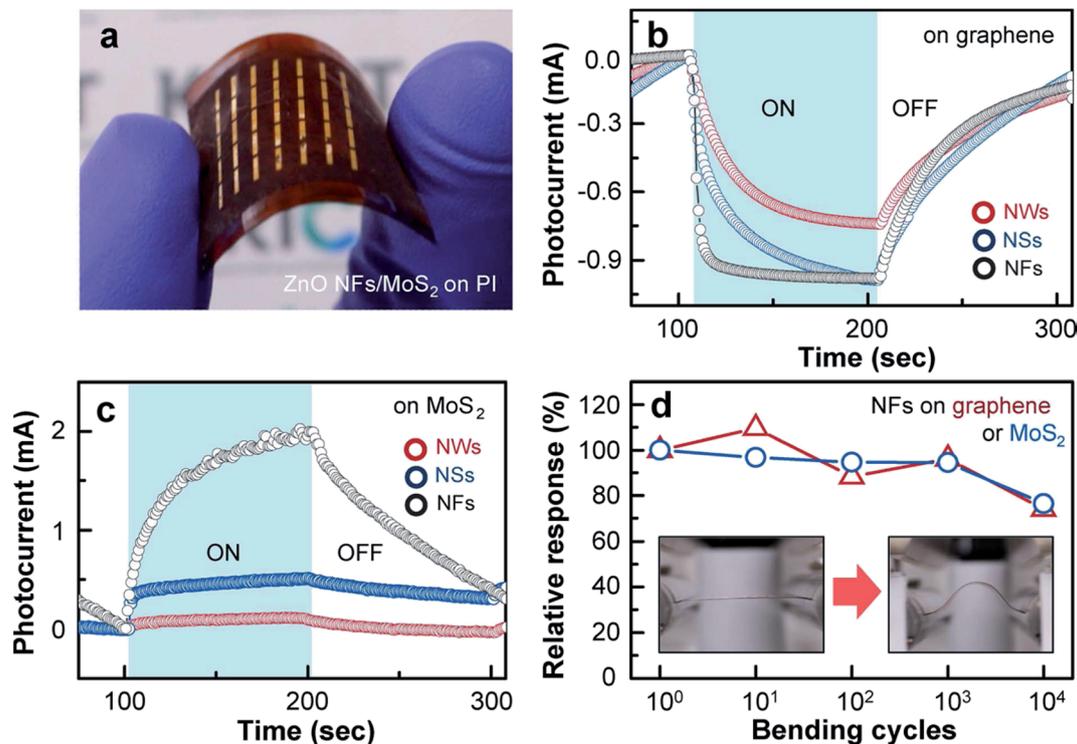


Fig. 21 (a) Photograph of the ZnO NF/MoS₂ photodetector fabricated on a flexible PI substrate. (b) Time-dependent photocurrents for ZnO NW/graphene, ZnO NS/graphene, and ZnO NF/graphene hybrids measured at 1 V. (c) Time-dependent photocurrents for ZnO NW/MoS₂, ZnO NS/MoS₂, and ZnO NF/MoS₂ hybrids measured at 1 V. (d) The change in the photocurrent of the ZnO NF/graphene hybrid (red) and ZnO NF/MoS₂ hybrid (blue) as a function of the bending cycle. Insets show the bending process of photodetectors at a bending radius of 6 mm. Reprinted with permission from ref. 277, copyright © 2017 American Chemical Society.

developed flexible photodetectors by transferring MoS₂ layers onto interdigital electrodes patterned on a PET substrate which showed a photoresponsivity of 20 mA W⁻¹, with response rise/fall times of 12 s and 19 s. Seo *et al.*²⁸⁰ developed inkjet printed flexible photodetector devices using MoS₂ nanosheets and graphene electrodes. MoS₂/graphene photodetectors were fabricated on a rigid glass substrate applying thermal annealing and a flexible polyimide (PI) substrate using photonic annealing. Thermally annealed MoS₂/graphene photodetectors showed photoresponsivity of 1 mA W⁻¹ and detectivity of 4.37 × 10⁷ Jones at 515.6 nm, which increased many folds for the photonic annealed photodetectors because more charge carriers were generated by the photothermal effect associated with lower thermal conductivity and larger heat capacity of polyimide thin film substrate than that of a rigid glass substrate. The photonic annealed flexible MoS₂/graphene photodetectors also showed retained mechanical stability over 500 bending cycles at 8.1 mm radius of curvature.

4.5 Surface functionalized MoS₂ layers for flexible photodetectors

The electrical, optical, mechanical, thermal, chemical, and biological properties of MoS₂ atomic layers can be altered by covalent surface functionalization. Different organic functional groups can be covalently attached to the MoS₂ atomic layers by exploiting vacancies mainly on surface atoms and consequently

their physical and chemical properties can be tuned.^{281–284} Tang *et al.*²⁸⁵ reported that the bandgap of 1T-phase MoS₂ monolayer can be tuned between zero to 1.0 eV depending upon the chemical bonding of CH₃, CF₃, OCH₃, NH₂ and H functional groups. Ding *et al.*²⁸⁶ demonstrated covalent functionalization of 2H-phase MoS₂ monolayers can be achieved and tuned by thiol (SH) group functionalization and controlling the vacancies of sulfur atoms on the basal planes. The covalent functionalization by thiol groups can modulate both photoluminescence and catalytic behavior of functionalized MoS₂ monolayers. The applications of PEGylated MoS₂ atomic layers and carboxyl (–COOH) functionalized MoS₂ nanosheets have been reported in cancer therapy²⁸⁷ and surface plasmon resonance (SPR) immunosensors.²⁸⁸ Various studies have shown that the covalent surface functionalization can be used for controlling the phase stability, surface characteristics, chemical reactivity, mechanical, and electrical properties of MoS₂.^{289–292}

Flexible MoS₂ photodetectors have been fabricated using surface-functionalized monolayer MoS₂. Pak *et al.*²⁹³ developed flexible photodetectors based on CVD-grown monolayer MoS₂ surface-functionalized with electron accepting (p-doping) octadecyltrichlorosilane (ODTS, –CH₃ groups) or electron donating (n-doping) (3-aminopropyl)triethoxysilane (APTES, –NH₂ groups) organic molecules. The PL spectrum of ODTS–MoS₂ layer was found to increase in the intensity and blue-shifted by 11 meV whereas the PL intensity of APTES–MoS₂ layer decreased and red-



shifted by 15 meV, compared with the pristine MoS₂ layer due to the different process of exciton recombination. Fig. 22 shows the schematics of a flexible MoS₂ based photodetector, ODTS functionalized monolayer MoS₂, and mechanical stability test and then compares the incident power dependent photoresponsivity and detectivity of the pristine MoS₂ with APTES–MoS₂ and ODTS–MoS₂ photodetector devices. The lower dark and photo current values of 475 pA and 202 nA for ODTS–MoS₂ device and higher values 1.2 μA and 4.6 μA for APTES–MoS₂ device were associated with their charge carrier concentrations, respectively. The photoresponsivity of the ODTS–MoS₂ device increased from 2.5 A W⁻¹ to 37.5 A W⁻¹ and from 25.2 A W⁻¹ to 1500 A W⁻¹ for the APTES–MoS₂ device at the lowest incident power. The functionalized MoS₂ channel assisted in transporting more excitons that yielded higher photoresponsivity for the APTES–MoS₂ device. The ODTS–MoS₂ photodetector showed highest detectivity of 10¹¹ Jones under the lowest incident power, which may be associated with the dropping of the dark level due to the electrons withdrawal from the channel. The photoresponse decay time of flexible APTES–MoS₂ photodetector was decreased to 0.7 s compared with 1.45 s for the pristine 1L-MoS₂ photodetector. The mechanical stability of the flexible ODTS–MoS₂/PET device was studied as a function of bending radius and bending cycles. At 2 mm bending radius, less than 10% degradation in photocurrent occurred while 20% degradation was noticed at bending radius of 4 mm up to 1000 bending cycles. This study demonstrated that a flexible MoS₂ photodetector can be fabricated with surface-functionalization of a MoS₂ monolayer. Kang *et al.*¹⁸⁰ also observed the similar phenomenon for the MoS₂

photodetector devices n-doped by APTES organic molecules where photoresponsivity was improved from 219 A W⁻¹ to 5.75 × 10³ A W⁻¹ for the APTES–MoS₂ devices.

Several other studies have been reported on MoS₂ hybrid based flexible photodetectors. In-plane lateral graphene/MoS₂ heterostructure showed photoresponsivity of 1.1 × 10⁵ A W⁻¹, detectivity of 1.4 × 10¹⁴ Jones and I_{on}/I_{off} current ratio of 10⁶ due to the strong absorption and increased separation of the photoexcited charge carriers and the charge transportation.²⁹⁴ MoS₂ phototransistors have been prepared on flexible poly-arylate substrate where poly(4-vinylphenol) (PVP) was used both as a gate dielectric and an encapsulating layer material.²⁹⁵ The encapsulation with PVP improved the field-effect mobility (μ_{FE}) and the current I_{on}/I_{off} ratio, which resulted in two orders of magnitude enhancement in photoresponsivity as well as in detectivity from visible to infrared wavelength. The hydroxyl functional groups in PVP improved the performance of MoS₂ phototransistors due to the n-doping effect and decreased the recombination of photoexcited carriers. Kang *et al.*²⁹⁶ developed MoS₂–ZnO hybrid based flexible photodetectors using MoS₂ nanosheets with atomic layer deposition (ALD) deposited ZnO nanopatches. The photocurrent was found to increase with an increasing number of ALD cycles where the highest photocurrent was measured for the MoS₂ nanosheets having 40 cycles ZnO nanopatches. The sulfur vacancies in defected MoS₂ structure disappeared with increasing ZnO cycles and the recombination of photoexcited carriers decreased. The photocurrent of pristine MoS₂ and flexible

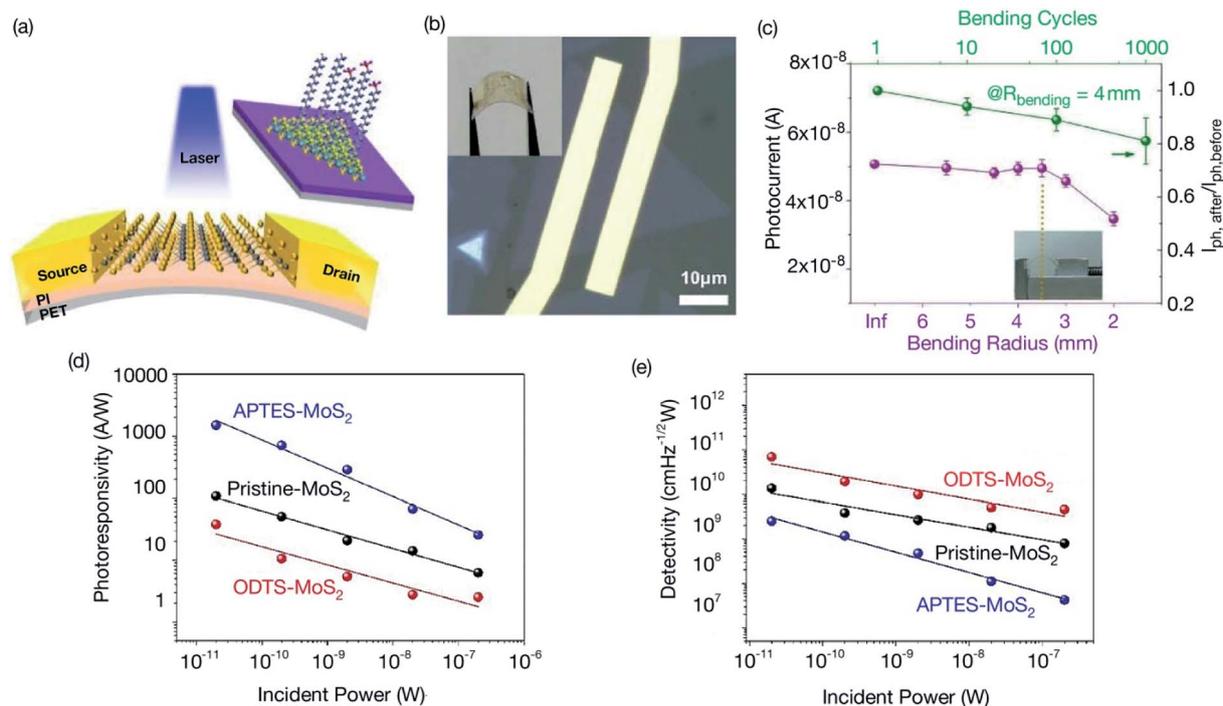


Fig. 22 (a) Schematics of a flexible MoS₂ based photodetector on a PET substrate and the surface-functionalized monolayer MoS₂ with ODTS (–CH₃ groups). (b) Schematics and optical image of flexible MoS₂ photodetector device developed using e-beam lithography. (c) Mechanical stability of flexible MoS₂ devices in terms of their photocurrent at different bending radius as a function of bending cycles. A comparison of the photoresponsivity (d) and detectivity (e) of the pristine MoS₂ with APTES–MoS₂ and ODTS–MoS₂ photodetector devices as a function of incident power intensity. Reprinted with permission from ref. 293, copyright © The Royal Society of Chemistry.



Table 1 The figure of merit of different MoS₂ based photodetectors. The performance of photodetectors is compared in terms of their photoresponsivity, detectivity and response/recovery time ($\tau_{\text{rise}}/\tau_{\text{decay}}$) along with their measurement wavelength and incident laser power (Jones = cm Hz^{1/2} W⁻¹)^a

MoS ₂ photodetector	Conditions (wavelength, incident power)	Photoresponsivity (A W ⁻¹)	Detectivity (Jones)	Response time ($\tau_{\text{rise}}/\tau_{\text{decay}}$)	Ref.
(i) Pristine MoS₂ based photodetectors					
1L MoS ₂	($\lambda = 561$ nm, 150 pW)	880	2×10^9	4 s/9 s	171
2L MoS ₂	($\lambda = 532$ nm, 35 pW)	1.1×10^5	—	232 s	306
	($\lambda = 1070$ nm, 150 nW)	5.2	—	216.5 s	308
3L MoS ₂	($\lambda = 532$ nm)	0.57	—	70 μ s/110 μ s	307
ML MoS ₂	($\lambda = 633$ nm, 10 nW)	0.12	10^{10} to 10^{11}	—	308
FL MoS ₂	($\lambda = 520$ nm)	6.3×10^{-5}	4.2×10^8	20 ms	304
5L MoS ₂	($\lambda = 850$ nm)	1.80	5.0×10^8	0.3 s/0.36 s	139
Pd/MoS ₂ /Cr–Au	($\lambda = 500$ nm)	5.07	3×10^{10}	100 ms/200 ms	197
1–2L MoS ₂ /HfO ₂	($\lambda = 635$ nm, 3.75 pW)	1×10^4	7.7×10^{11}	10 ms	136
1L MoS ₂ /Al ₂ O ₃	($\lambda = 635$ nm)	406	3.8×10^{11}	55 ms	136
MoS ₂ /SiC	($\lambda = 325$ nm)	1.02×10^4	6.4×10^{11}	—	217
	($\lambda = 532$ nm)	1.6×10^3	1.0×10^{11}	—	217
MoS ₂ /SiC/Si	($\lambda = 325$ nm)	44	1.4×10^{10}	—	217
	($\lambda = 532$ nm)	30	9.5×10^9	—	217
2L MoS ₂ (CVD)	($\lambda = 532$ nm, 0.377 mW cm ⁻²)	7160	6.62×10^{10}	97 ms/291 ms	351
MoS ₂ layers (PLD)	($\lambda = 365$ nm)	3.0×10^4	1.81×10^{14}	32 ms	369
1L/6L MoS ₂ (serial multi-heterojunction)	($\lambda = 520$ nm, 1 nW)	9.26×10^4	2.38×10^{13}	20 ms/25 ms	348
	($\lambda = 520$ nm, 5 pW)	2.67×10^6	—	5 ms/5 ms	—
	($\lambda = 785$ nm, 1 nW)	1.86×10^4	—	15 ms/20 ms	—
	($\lambda = 850$ nm, 1 nW)	1.08×10^4	—	10 ms/15 ms	—
	($\lambda = 1064$ nm, 1 nW)	1.07×10^3	—	5 ms/10 ms	—
	($\lambda = 1064$ nm, 5 pW)	1.65×10^4	—	1.5 ms/2.5 ms	—
	($\lambda = 520$ nm, 1 nW)	8.74×10^3	—	465 ms/315 ms	348
MoS ₂ microspheres	($\lambda = 405$ nm, 1.77 mW cm ⁻²)	0.96	2.9×10^{10}	—	384
(ii) MoS₂ based flexible photodetectors					
MoS ₂ /Kapton (flexible)	($\lambda = 325$ nm)	3.19×10^2	4.5×10^{11}	—	217
	($\lambda = 532$ nm)	5.0	6.4×10^9	—	217
MoS ₂ /PET (flexible)	($\lambda = 325$ nm)	1.36×10^2	2.0×10^{11}	—	217
	($\lambda = 532$ nm)	9.0	9.7×10^9	—	217
FL MoS ₂ /graphene/PET (flexible)	($\lambda = 632.8$ nm, 0.645 μ W)	10	—	1.5 s	309
g-C ₃ N ₄ /MoS ₂ (flexible)	($\lambda = 365$ nm, 0.5 W cm ⁻²)	4.0	4×10^{11}	60 ms/95 ms	278
MoS ₂ /graphene/polyimide (flexible) (photonic annealing at 2.8 kV for 1.36 ms)	($\lambda = 515.6$ nm, 0.6 mA W ⁻¹)	50 mA W ⁻¹	3.18×10^9	5 ms	280
MoS ₂ /Kapton (flexible)	($\lambda = 325$ nm)	319	4.5×10^{11}	—	303
	($\lambda = 532$ nm)	5.0	6.4×10^9	—	303
MoS ₂ /PET (flexible)	($\lambda = 325$ nm)	136	2.0×10^{11}	—	303
	($\lambda = 532$ nm)	9.0	9.7×10^9	—	303
MoS ₂ /SiO ₂ /Si	($\lambda = 325$ nm)	44	1.4×10^{10}	—	303
	($\lambda = 532$ nm)	30	9.5×10^9	—	303
MoS ₂ /SiC	($\lambda = 325$ nm)	1.02×10^4	6.4×10^{11}	—	303
	($\lambda = 532$ nm)	1.6×10^3	1.0×10^{11}	—	303
ZnO NFs/MoS ₂ (flexible)	($\lambda = 350$ nm, 1.2 mW cm ⁻²)	8.99×10^{-4}	—	61 s/90 s	277
(iii) MoS₂/2D van der Waals heterostructure-based photodetectors					
1L MoS ₂ /1L graphene	($\lambda = 532$ nm, 0.01 W m ⁻²)	1.2×10^7	—	2.5 s	226
MoS ₂ /graphene nanoribbon	($\lambda = 385$, 2.1 μ W)	66	—	5 ms/30 ms	232
Graphene/MoS ₂ /graphene	($\lambda = 633$ nm, 1 μ W cm ⁻²)	1.14×10^5	$9. \times 10^{15}$	25.7 s/41.4 s	300
Au/MoS ₂ /Au	($\lambda = 633$ nm, 1 μ W cm ⁻²)	4.8×10^3	2.7×10^{14}	7.1 s/31.8 s	300
Graphene/MoS ₂ /graphene	($\lambda = 432$ nm)	2.2×10^5	3.5×10^{13}	1.7 ms/2.8 ms	301a
FL MoS ₂ /rGO NPs	($\lambda = 460$ nm, 8 mW cm ⁻²)	2.10	5×10^{11}	18 ms	310
ML graphene–MoS ₂ –WS ₂ /fiber	($\lambda = 400$ nm, 6.35 nW cm ⁻²)	6.6×10^7	—	7 ms/21.86 ms	199
MoS ₂ /h-BN/graphene	($\lambda = 532$ nm)	180	2.6×10^{13}	0.23 s/0.25 s	196
5L 2H-SnSe ₂ /1L 2H-MoS ₂	($\lambda = 500$ nm)	9.1×10^3	9.3×10^{10}	0.2 s/0.6 s	329
1L ML MoS ₂	($\lambda = 500$ nm)	37.3	1.4×10^9	—	329
MoS ₂ /SnS ₂	($\lambda = 450$ nm)	2.3	—	—	164



Table 1 (Contd.)

MoS ₂ photodetector	Conditions (wavelength, incident power)	Photoresponsivity (A W ⁻¹)	Detectivity (Jones)	Response time ($\tau_{\text{rise}}/\tau_{\text{decay}}$)	Ref.
ML PdSe ₂ /MoS ₂	($\lambda = 10.6 \mu\text{m}$, 435.9 nW)	42.1	8.21×10^9	74.5 ms/93.1 ms	168
	($\lambda = 4.012 \mu\text{m}$)	28.83	6.09×10^{10}	—	168
FL MoS ₂ /FL SnSe	($\lambda = 532$, 0.65 nW)	100	—	4 ms/6 ms	201
MoS ₂ /WS ₂	($\lambda = 638 \text{ nm}$)	1.36	—	—	204
ML MoS ₂ /MoTe ₂	($\lambda = 532 \text{ nm}$, 100 mW cm ⁻²)	0.62	10^{10}	0.01 ms	205a
ML MoS ₂ /MoTe ₂	($\lambda = 473 \text{ nm}$, 46.8 mW cm ⁻²)	0.15	—	68 ms/68 ms	205b
MoS ₂ /MoTe ₂	($\lambda = 1200 \text{ nm}$)	0.046	—	60 μs /25 μs	205c
ML MoS ₂ /MoTe ₂	($\lambda = 473 \text{ nm}$)	0.064	1.6×10^{10}	385 ms/453 ms	205d
1L MoS ₂ /GaSe	($\lambda = 300 \text{ nm}$, 6.41 mW cm ⁻²)	0.063	—	80 ms/20 ms	167 and 202
FL MoS ₂ /FL black phosphorus	($\lambda = 532 \text{ nm}$, 1 nW)	22.3	3.1×10^{11}	—	170
	($\lambda = 1550 \text{ nm}$, 1 nW)	0.1534	2.13×10^9	15 μs /70 μs	
MoS ₂ /FL black phosphorus	($\lambda = 520 \text{ nm}$)	5000	1.6×10^{10}	0.02 s	332
FL WSe ₂ /FL BP/FL MoS ₂	($\lambda = 532 \text{ nm}$, 13.5 nW)	6.32	1.25×10^{11}	—	333
	($\lambda = 1550 \text{ nm}$, 13.5 nW)	1.12	2.21×10^{10}	—	333
MAPbI ₃ /FL BP/FL MoS ₂	($\lambda = 457 \text{ nm}$, 0.02 mW cm ⁻²)	11	1.3×10^{12}	0.15 ms/0.24 ms	341
MoS ₂ /graphene/WSe ₂	($\lambda = 532 \text{ nm}$, 0.2 nW)	4250	2.2×10^{12}	53.6 μs /30.3 μs	198
GaTe/MoS ₂	($\lambda = 633 \text{ nm}$, 100 mW cm ⁻²)	1.365	—	10 ms	167
GaSe/MoS ₂	($\lambda = 300 \text{ nm}$)	0.06	—	80 ms/20 ms	207a
FL graphene/GaSe/MoS ₂	($\lambda = 532 \text{ nm}$)	3.0	10^{10}	50 ms	207b
FL GaTe/FL MoS ₂	($\lambda = 473 \text{ nm}$, 0.04 mW)	21.83	8.4×10^{13}	7 ms	334
1L MoS ₂ /ML MoSe ₂	($\lambda = 610 \text{ nm}$, 0.29 mW cm ⁻²)	1.3	2.6×10^{11}	0.6 s/0.5 s	335
BL MoS ₂ /BLWS ₂ (CVD)	($\lambda = 457 \text{ nm}$, 3.57 $\mu\text{W cm}^{-2}$)	6.7×10^3	3.09×10^{13}	—	344
	($\lambda = 532 \text{ nm}$, 3.57 $\mu\text{W cm}^{-2}$)	3.0×10^3	1.24×10^{13}	—	344
FL β -In ₂ Se ₃ /1L MoS ₂	($\lambda = 532 \text{ nm}$, 4 mW cm ⁻²)	23.7	5×10^{11}	4 ms/7 ms	354
WS ₂ /MoS ₂ vdWHs	(405 nm, 3.25 mW mm ⁻²)	7×10^{-5}	—	1.1 s/2.7 s	382
WS ₂ /MoS ₂ vdWHs nanoscroll	(405 nm)	0.647	—	0.7 s/2/4 s	382
(iv) MoS₂/perovskite heterostructure-based photodetectors					
Perovskite/MoS ₂ -rGO	($\lambda = 660 \text{ nm}$)	1.08×10^4	4.28×10^{13}	45 ms	179
ML 1T-MoS ₂ /CH ₃ NH ₃ PbI ₃	($\lambda = 500 \text{ nm}$, 0.14 mW)	3.3×10^5	7×10^{11}	0.45 s/0.75 s	325c
ML 2H-MoS ₂ /CH ₃ NH ₃ PbI ₃	($\lambda = 500 \text{ nm}$, 0.14 mW)	142	2.6×10^{11}	25 ms/50 ms	325c
2H-MoS ₂ /1T@2H-MoS ₂	($\lambda = 532 \text{ nm}$, 2.35 mW cm ⁻²)	1917	7.55×10^{11}	—	325d
1L 2H-MoS ₂	($\lambda = 532 \text{ nm}$, 2.35 mW cm ⁻²)	56	1.3×10^{11}	—	325d
1L MoS ₂ /CsPbI _{3-x} Br _x QDs	($\lambda = 532 \text{ nm}$, 1.5 μW)	7.7×10^4	5.6×10^{11}	0.59 s/0.32 s	190
1L MoS ₂ /CsPbBr ₃	($\lambda = 442 \text{ nm}$, 20 $\mu\text{W cm}^{-2}$)	4.4	2.5×10^{10}	0.72 ms/1.01 ms	191a
MoS ₂ /CsPbBr ₃ QDs	($\lambda = 405 \text{ nm}$, 12.8 $\mu\text{W cm}^{-2}$)	4.68×10^4	—	7.5 ms/8 ms	191b
1L MoS ₂ /CsPbBr ₃ NCs	($\lambda = 405 \text{ nm}$, 0.6 mW cm ⁻²)	24.34	3.93×10^{12}	5.5 μs /24 μs	191c
1L MoS ₂ /CsPbBr ₃ NCs	($\lambda = 532 \text{ nm}$)	6.4×10^2	3.38×10^{11}	—	191d
ML MoS ₂ /CH ₃ NH ₃ PbI ₃ /APTES	($\lambda = 520 \text{ nm}$, 4.63 pW)	2.12×10^4	1.38×10^{10}	6.17 s/4.5 s	192
ML MoS ₂ /APTES	($\lambda = 520 \text{ nm}$)	2.38×10^3	4.23×10^9	—	192
ML MoS ₂	($\lambda = 520 \text{ nm}$)	8.16×10^2	3.93×10^9	10.7 s/6.2 s	192
FL MoS ₂ /MA ₃ Bi ₂ Br ₉	($\lambda = 530 \text{ nm}$, 18 $\mu\text{W cm}^{-2}$)	112	3.8×10^{12}	0.3 ms/0.3 ms	326
MoS ₂ /CH ₃ NH ₃ PbI ₃ (vertical)	—	68.11	—	205 ms/206 ms	327
MoS ₂ /CH ₃ NH ₃ PbI ₃ (planar)	—	28	—	356 ms/204 ms	327
(PEA) ₂ SnI ₄ /FL MoS ₂ /graphene	($\lambda = 451 \text{ nm}$, 36 pW)	1100	8.09×10^9	34 ms/38 ms	328
(PEA) ₂ PI ₄ /ML MoS ₂	($\lambda = 637 \text{ nm}$, 0.1 nW)	16.8	1.06×10^{13}	6 ms/4 ms	322b
(v) MoS₂/inorganic semiconductor heterostructure-based photodetectors					
ML MoS ₂ /Si	($\lambda = 808 \text{ nm}$, 1 mW cm ⁻²)	300	10^{13}	4 μs /42 μs	162
ML MoS ₂ /Si	($\lambda = 780 \text{ nm}$, 45 $\mu\text{W cm}^{-2}$)	23.1	1.63×10^{12}	21.6 μs /65.5 μs	311
ML MoS ₂ /Si	($\lambda = 580 \text{ nm}$, 5 mW cm ⁻²)	8.75	1.4×10^{12}	10 μs /19 μs	312a
MoS ₂ /Si (porous)	($\lambda = 550 \text{ nm}$)	9	8×10^{12}	9 μs /7 μs	312b
V-ML MoS ₂ /Si	($\lambda = 650 \text{ nm}$, 90 mW cm ⁻²)	11.9	2.1×10^{10}	30.5 μs /71.6 μs	313
V-FL MoS ₂ /Si	($\lambda = 808 \text{ nm}$, 1.6 mW cm ⁻²)	908.2	1.88×10^{13}	56 ns/825 ns	305
V-ML MoS ₂ /Si	($\lambda = 660 \text{ nm}$, 12 nW)	76.1	1.6×10^{12}	48.9 s	175
V-FL MoS ₂ /Si	($\lambda = 808 \text{ nm}$, 5 mW cm ⁻²)	0.746	6.03×10^{11}	178 μs /198 μs	356
FL MoS ₂ /Si FET	($\lambda = 850 \text{ nm}$, 50 $\mu\text{W cm}^{-2}$)	1.78×10^4	3.0×10^{13}	1.44 ms/1.45 ms	315
MoS ₂ /SiNWA	($\lambda = 650 \text{ nm}$, 6.3 $\mu\text{W cm}^{-2}$)	53.5	2.8×10^{13}	2.9 μs /7.3 μs	366



Table 1 (Contd.)

MoS ₂ photodetector	Conditions (wavelength, incident power)	Photoresponsivity (A W ⁻¹)	Detectivity (Jones)	Response time ($\tau_{\text{rise}}/\tau_{\text{decay}}$)	Ref.
Si/MoS ₂	($\lambda = 660$ nm)	1.1	—	0.38 s/0.1 s	138
Si-(Au@MoS ₂)	($\lambda = 800$ nm)	30	—	0.02 s	138
Au@MoS ₂ core-shell	($\lambda = 660$ nm, 50 μW)	0.5	—	3.2 s/0.28 s	138
ML MoS ₂ /GaN	($\lambda = 405$ nm, 2 mW)	17.2	1.8×10^{12}	0.1 s/9 s	317a
ML MoS ₂ /GaN	($\lambda = 365$ nm, 4.351 μW)	11.3	1.34×10^{10}	21.1 s/19.7 s	317b
Al ₂ O ₃ /MoS ₂ /GaN	($\lambda = 365$ nm, 3.141 μW)	24.62	7.2×10^9	12.8 s/26.4 s	317b
MoS ₂ /GaAs (CVD)	($\lambda = 635$ nm)	0.321	3.5×10^{13}	17 μs /31 μs	318
SiQDs/MoS ₂ /h-BN/GaAs	($\lambda = 635$ nm)	0.419	1.9×10^{14}	—	318
FL MoS ₂ /CdSe	($\lambda = 405$ nm, 8.52 $\mu\text{W cm}^{-2}$)	2.5×10^5	1.24×10^{14}	60 ms/60 ms	323
MoS ₂ /ZnCdSe/ZnS QDs	($\lambda = 450$ nm, 400 nW)	3.7×10^4	1×10^{12}	0.3 s/1.2 s	194
FL MoS ₂ /TiO ₂ /HgTe QDs	($\lambda = 635$ nm, 0.35 $\mu\text{W cm}^{-2}$)	5×10^5	6.4×10^{12}	—	193
	($\lambda = 1310$ nm, 53 mW cm^{-2})	2×10^4	3×10^{11}	4 ms	
ML MoS ₂ /TiO ₂ /PbS QDs	($\lambda = 635$ nm, 10 nW cm^{-2})	10^5	5×10^{12}	12 ms	319
MoS ₂ /PbS	($\lambda = 800$ nm, 0.15 mW cm^{-2})	4.5×10^4	3×10^{13}	7.8 ms	321
FL MoS ₂ /PbS QDs	($\lambda = 635$ nm, 1 mW)	6.0×10^5	7×10^{14}	0.35 s	322
1L MoS ₂ /PbS QDs	($\lambda = 850$ nm)	5.4×10^4	1×10^{11}	950 μs /1 ms	188
MoS ₂ -ZnO/PET	(254 nm, 2.65 W m^{-2})	2.7	—	13.96 s/55.33 s	296
p-MoS ₂ /n-ZnO	(365 nm, 5.7 mW cm^{-2})	24.36	—	0.9 s/1.04 s	360
n-MoS ₂ /n-ZnO	(532 nm)	0.35	—	1.3 s/2.2 s	360
1L MoS ₂ /TiO ₂ nanoflowers	($\lambda = 370$ nm)	35.9	1.98×10^{13}	33.7 ms/28.3 ms	320
	($\lambda = 570$ nm)	18.5	1.09×10^{13}	—	320
1L MoS ₂ /Ti	($\lambda = 570$ nm)	8.6	5.7×10^{12}	741 ms/128 ms	320
1L MoS ₂ /SiO ₂	($\lambda = 570$ nm)	6.2	3.49×10^{12}	642 ms/214 ms	320
V-MoS ₂ /TiO ₂	($\lambda = 532$ nm, 856 nW)	133	3.325×10^{11}	27.8 ms/48 ms	346b
MoS ₂ /CuO nanowires (strain)	($\lambda = 600$ nm, 55 μW)	157.6	3.3×10^8	34.6 ms/51.9 ms	331
V-ITO/MoS ₂ /Cu ₂ O/Au	($\lambda = 500$ nm, 0.26 W m^{-2})	5.77×10^4	3.2×10^{14}	70 ms	302
(vi) Chemically doped MoS₂ photodetectors					
1L MoS ₂ /methylene blue doping	($\lambda = 610$ nm)	9.09	2.2×10^{11}	27.1 s/39.2 s	140
1L MoS ₂ /R6G doping	($\lambda = 520$ nm, 1 μW)	1.17	1.5×10^7	5.1 μs /2.3 s	181
ML MoS ₂ /PPh ₃ doping	($\lambda = 520$ nm, 5 pW)	3.92×10^5	2.36×10^{10}	13.2 ms/17.2 ms	187
1L MoS ₂ /HAuCl ₄ doping	($\lambda = 532$ nm, 0.15 mW cm^{-2})	99.9	9.4×10^{12}	16.6 s/5.2 s	337
1L MoS ₂ /pentacene	($\lambda = 655$ nm, 28.18 mW cm^{-2})	0.31	1.55×10^{13}	—	330
1L MoS ₂ /ZnPc/Al ₂ O ₃	($\lambda = 532$ nm, 0.07 mW cm^{-2})	1.4×10^4	3×10^{11}	—	182
	($\lambda = 532$ nm, 3.64 mW cm^{-2})	4.3×10^2	—	—	
1L MoS ₂ /ZnPc	($\lambda = 532$ nm, 3.64 mW cm^{-2})	7.84	—	100 ms/10 ms	182
3L MoS ₂ /P(VDF-TrFE)	($\lambda = 635$ nm, 1 nW)	2.579×10^3	2.2×10^{12}	1.8 ms/2 ms	137
MoS ₂ /P(VDF-TrFE-CFE)	($\lambda = 450$ nm, 20 nW)	346.24	—	—	324
MoS ₂ /P(VDF-TrFE)	($\lambda = 637$ nm, 1 nW)	3.26×10^3	9.0×10^{14}	480 μs /320 μs	340
MoS ₂ /polyaniline (PANI)	($\lambda = 785$ nm, 1.4 mW mm^{-2})	25	—	—	338
MoS ₂ -MoO _x	($\lambda = 405$ nm, 1 mW cm^{-2})	1.09	2.08×10^{11}	9.8 s/12.6 s	339
MoS ₂ /UCNPs	($\lambda = 325$ nm, 50 W m^{-2})	192	1.61×10^{14}	—	364
	($\lambda = 532$ nm, 50 W m^{-2})	81	6.80×10^{13}	16 s/18 s	364
	($\lambda = 980$ nm, 50 W m^{-2})	1254	1.05×10^{15}	11 s/17 s	364
	($\lambda = 1064$ nm, 50 W m^{-2})	127.5	1.05×10^{14}	—	364
MoS ₂ /Mo ₂ C (multiple grating)	($\lambda = 665$ nm)	3.7×10^3	—	0.13 s/0.95 s	387
Sb ₂ O ₃ /MoS ₂	($\lambda = 532$ nm, 0.057 W cm^{-2})	4.5×10^4	1.0×10^{15}	63 ms/71 ms	372
BaTiO ₃ /MoS ₂	(365 nm, 1.0 mW cm^{-2})	120	1.1×10^{11}	0.7 s/2 s	375
MoSSe	(660 nm, 1.75 mW mm^{-2})	1.75×10^{-3}	—	4.7 ms	385
MoS _{1.15} Se _{0.85}	(532 nm)	2.06	—	18 ms/35 ms	386
MoS _{2(1-x)} Se _{2x}	(650 nm)	191.5	10^{12}	51 ms	435

^a 1L: single-layer, BL: bi-layer, FL: few-layer, ML: multilayer, V: vertically oriented, PET: poly(ethylene terephthalate), PPh₃: triphenylphosphine, QDs: quantum dots, SiNWA: Si nanowire array, P(VDF-TrFE): poly(vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE)] ferroelectric polymer, P(VDF-TrFE-CFE): poly(vinylidene fluoride-trifluoroethylene-chlorofluoroethylene), APTES: (3-aminopropyl)triethoxysilane, ZnPc: zinc phthalocyanine, Jones = $\text{cm Hz}^{1/2} \text{W}^{-1}$, polymer stabilizer ethyl cellulose (EC), 2D Ruddlesden-Popper perovskites: $(\text{C}_6\text{H}_5\text{C}_2\text{H}_4\text{NH}_3 = \text{PEA})_2\text{SnI}_4$, PANI: polyaniline, Mo₂C: molybdenum carbide.



MoS₂-ZnO/PET hybrid photodetectors showed no significant degradation after bending at 3 mm radius; up to 10 000 cycles were examined.

0D PbS QDs were deposited onto few layered 2H-MoS₂ to develop flexible photodetectors on a PET substrate.²⁹⁷ The 0D/2D PbS/MoS₂ hybrid photodetectors showed the photoresponsivity of 0.398 to 0.543 A W⁻¹ and the detectivity of 2 × 10¹² to 2.68 × 10¹² Jones between 480 to 1200 nm at a 2 V bias. The 2H-MoS₂ based stretchable photodetectors were fabricated on polydimethylsiloxane (PDMS) substrates.²⁹⁸ The photoresponsivity of 2.52 μA W⁻¹ and response times of 122 ms/120 ms were recorded at 515.6 nm. The stretchable photodetectors retained the photoconductivity up to 5.72% tensile strain and over 1000 stretching cycles. MoS₂ was deposited on a flexible cellulose ester paper and the inkjet-printed PEDOT:PSS electrodes were used to develop MoS₂ paper based photodetectors, which showed internal quantum efficiency of 0.063%, photoresponsivity of 0.134 mA W⁻¹ and photoresponse/recovery times of 17.5 s/15.3 s at 405 nm.²⁹⁹

5. Analysis and factors affecting the figure of merit of MoS₂ photodetectors

All these studies show that MoS₂ atomic layers can be used to develop high-performance photodetectors. The analysis and factors affecting the parameters such as photoresponsivity, specific detectivity, NEP, photogain, EQE, LDR, and response time of the MoS₂ photodetectors have been further discussed in this section. Table 1 summarizes the research data collected on the figure-of-merit of a wide variety of MoS₂ based photodetectors in terms of their key parameters such as photoresponsivity, detectivity and response/recovery time ($\tau_{\text{rise}}/\tau_{\text{decay}}$) of photodetectors and their performance measured at various laser wavelengths under different incident laser powers/intensities.^{138,171,175,179,190-199,217,226,300-435} The MoS₂ based photodetectors have been analyzed taking into account the factors affecting their performance. MoS₂ based photodetectors have also been discussed and categorized as (i) pristine MoS₂ based photodetectors, (ii) flexible MoS₂ photodetectors, (iii) MoS₂/2D van der Waals heterostructure-based photodetectors, (iv) MoS₂/perovskite heterostructure-based photodetectors, (v) MoS₂/inorganic semiconductor heterostructure-based photodetectors, (vi) chemically doped MoS₂ photodetectors, and (vii) self-powered MoS₂ photodetectors.

The influence of different types of substrate materials on the performance of MoS₂ photodetectors has been extensively studied.^{164,178,184,217-220,222,223,234,256,259,278,298,311-313} MoS₂@TiO₂ core-shell heterojunction-based photodetectors were prepared by Paul *et al.*³²⁰ where 1L MoS₂ was used as the shell and TiO₂ nanoflowers as the core. The 83 and 30 times increase in PL intensity was observed from the 1L MoS₂ shell/TiO₂ nanoflowers p-n heterojunction more than that of 1L MoS₂/Ti and 1L MoS₂/sapphire substrates, respectively. The 1L MoS₂@TiO₂ heterojunction-based photodetectors showed photoresponsivity of 35.9 A W⁻¹ and detectivity of 1.98 × 10¹³ Jones in the UV region and 18.5 A W⁻¹ and 1.09 × 10¹³ Jones in

the visible region. 1L MoS₂@TiO₂ heterojunction also exhibited an order of magnitude faster photoresponse compared to the 1L MoS₂@Ti and 1L MoS₂@SiO₂ devices due to the fast photoexcited carrier transport at the p-n heterojunction associated with the substantial built-in electric field. In another study, MoS₂ nanosheets were placed on top of a CuO nanowire by wet transfer printing method.³³¹ MoS₂/CuO heterojunction-based photodetectors showed the photoresponsivity 157.6 A W⁻¹, rectification ratio of 6000, and low dark current of 38 fA at -2 V.

5.1 0D/2D quantum dots/MoS₂ hybrid heterostructures

The performance of MoS₂ photodetectors can be significantly enhanced by developing hybrid heterostructures as discussed in the earlier sections. Kufer *et al.*^{322a} showed significantly increased light absorption and carrier mobility in the 2D-0D MoS₂/PbS QDs hybrid phototransistors. The photoresponsivity of pristine bilayer MoS₂ with MoS₂/PbS hybrid were compared as a function of wavelength between 600 nm to 1200 nm. The bilayer MoS₂ absorbs up to 700 nm whereas the MoS₂/PbS hybrid extends the absorption up to 1200 nm with an excitation peak at 980 nm. Bilayer MoS₂ phototransistors showed field-effect mobility of 10-20 cm² V⁻¹ s⁻¹ and current $I_{\text{on}}/I_{\text{off}}$ ratios of 10⁵ to 10⁶ which is further increased after PbS QDs doping. The photoresponsivity of pristine bilayer MoS₂ is 5 A W⁻¹ at 1.8 eV, which is dramatically enhanced to 10⁵ to 10⁶ A W⁻¹ for the MoS₂/PbS hybrid phototransistor due to the PbS DQs. The multilayer MoS₂/PbS hybrid photodetector showed photoresponsivity up to 10⁶ A W⁻¹ at the lowest laser power intensity. The photoresponsivity of the hybrid photodetector varies by six orders of magnitude as a function of applied laser power intensity, which decreased as the laser intensity increases. The photoresponse of hybrid photodetector to light can be tailored by adjusting the size of PbS QDs. The photoresponsivity of the hybrid photodetector (6 × 10⁵ A W⁻¹) was found to be three orders of magnitude higher than that of PbS QDs (4.3 × 10² A W⁻¹). The NEP were found to be 2 × 10⁻¹⁵ W Hz^{-1/2} for the bilayer MoS₂ and 7 × 10⁻¹⁶ W Hz^{-1/2} for few-layer MoS₂/PbS hybrids at 1 Hz frequency, yielding corresponding detectivity of 2 × 10¹¹ Jones and 5 × 10¹¹ Jones, respectively. The MoS₂/PbS hybrid photodetector reached detectivity as high as 7 × 10¹⁴ Jones at applied negative back-gate bias V_g of -100 V. The study showed that MoS₂/PbS hybrid photodetector can achieve several orders of magnitude higher photoresponsivity than those of individual bilayer MoS₂ and PbS QD-based photodetectors. In another study, Pak *et al.*^{322b} fabricated hybrid photodetectors by decorating n-type MoS₂ with p-type manganese oxide QDs (MnO QDs), which showed suppressed dark current (5 nA) and an enhancement of photocurrent (52 nA) compared to a pristine MoS₂ photodetector (10 nA and 32 nA) at under 100 mW cm⁻² incident light intensity. MoS₂/MnO QDs hybrid photodetectors showed the photoresponsivity of 20 A W⁻¹ and detectivity of 5 × 10¹¹ Jones at 260 nm light illumination where increased detection was noticed in DUV range below 300 nm. The absorption of MnO QDs on MoS₂ surface suppressed dark current and increased photogeneration of charge carriers.



Ferroelectric polymers have been used with MoS₂ to improve the performance of photodetectors. Wang *et al.*³⁴⁰ developed an ultrabroad band photodetector integrating unique pyroelectric properties of organic ferroelectric copolymer P(VDF-TrFE) with MoS₂, which operated from 375 nm (ultraviolet) to 10 μm (LWIR) spectral range. The photocurrent of MoS₂ was improved in the 2.76 μm to 10 μm spectral range due to the pyroelectricity of P(VDF-TrFE). The photoresponsivity of 140 mA W⁻¹, $I_{\text{on}}/I_{\text{off}}$ current ratio of 10³ and a response time of 5.5 ms was achieved for the MoS₂/P(VDF-TrFE) hybrid photodetector. Though the highest photoresponsivity of 3.26×10^3 and detectivity of 9.0×10^{14} Jones were observed at 637 nm illumination at the drain bias of 5 V, the ferroelectric polarization field of P(VDF-TrFE) significantly improved the photoconductivity of MoS₂ and curtailed the dark current as well as the noise. The single-pixel imaging was also recorded by the photodetector. In a recent study, Li *et al.*³³⁷ showed that the photoresponsivity and detectivity of n-doped monolayer MoS₂ photodetector increased by 14.6 and 4.8 times compared with pristine MoS₂ photodetector, after *in situ* chemical doping with gold chloride hydrate (HAuCl₄·xH₂O). The *in situ* n-doped MoS₂ based photodetector exhibited the photoresponsivity of 99.9 A W⁻¹ and detectivity of 9.4×10^{12} Jones under $V_{\text{ds}} = 0.1$ V and $V_{\text{g}} = 0$ V, much higher compared with pristine MoS₂ photodetectors. The enhanced photoresponse originated from n-type chlorine doping of CVD-grown MoS₂ film which decreased the trapping of photoexcited electrons. Both photoresponsivity and detectivity of doped and pristine MoS₂ photodetectors were found to decrease with increasing incident laser power intensity. The chlorine n-type doping of MoS₂ photodetector increased the photoresponse due to improved photogating effect. The n-doped MoS₂ photodetector exhibited EQE over 100% which was found to increase as a function of decreasing wavelength, confirming photoconversion efficiency under shorter light illuminating wavelengths because of the higher excitation energy. Furthermore, these n-doped MoS₂ photodetectors also showed long-term stability as demonstrated by retaining 94% of the initial photocurrent after up to nine months.

Perovskites, van der Waals and plasmonic materials have been used with MoS₂ to improve the performance of photodetectors. Wang *et al.*^{341a} reported perovskite/black phosphorus/MoS₂ photogate photodiode, which utilizes black phosphorus/MoS₂ photodiodes with perovskite in order to induce high photoresponsivity and a fast photoresponse. This device architecture is constructed based on the fast photovoltaic operation together with the high-gain photogating effect. Under reverse bias condition, the 2D photogate photodiode showed photoresponsivity of 11 A W⁻¹, detectivity of 1.3×10^{12} Jones, fast response times of 150/240 μs, and a low dark current of 3×10^{-11} A in reverse bias. The photogate photodiode device yielded detectivity of 3×10^{11} Jones, external quantum efficiency (EQE) of 80% and $I_{\text{on}}/I_{\text{off}}$ current ratio of 3×10^7 in a zero bias (self-powered mode) indicating their potential in fabricating photodetectors and photovoltaic devices. The black arsenic phosphorus (b-AsP) which is a p-type semiconductor was used with MoS₂ n-type semiconductor to develop vdW p-n heterojunction-based MIR photodetectors.^{341b} The

photoresponsivity of AsP/MoS₂ photodetectors varied from 216.1 mA W⁻¹ to 115.4 mA W⁻¹ as the laser wavelength was increased from 2.36 μm to 4.29 μm and the value of EQE decreased from 11.36% to 3.33%, respectively. The NEP of vdW p-n junction was less than 0.24 pW Hz^{-1/2}, while that of FET device below 4.35 pW Hz^{-1/2} for 8.05 μm MIR wavelength at room temperature. The detectivity of the AsP/MoS₂ vdW p-n junction remained higher than 4.9×10^9 Jones over the 3 μm to 5 μm spectral range and larger than the detectivity of 1.06×10^8 Jones for AsP FET device at 8.05 μm wavelength. These studies demonstrated fabrication of MoS₂ based MIR photodetectors. Wu *et al.*³⁴² used a Au-supported gap-mode surface plasmon increasing configuration to improve the optoelectronic properties, with which a monolayer MoS₂ photodetector exhibited a photoresponsivity of 287.5 A W⁻¹ and a 880% increment in the photocurrent.

The self-powered photovoltaic photodetector developed from individual monolayer MoS₂-WS₂ heterostructures showed photoresponsivity of 4.36 mA W⁻¹, EQE of 1.02%, and detectivity of 4.36×10^{13} Jones at 532 nm under laser power intensity of 28.64 mW cm⁻² under 0 V bias.³⁴³ Photoresponsivity of 4.34 and 10.44 mA W⁻¹ were measured for positive bias (3 V) and reversed bias (-3 V), respectively. Ye *et al.*³⁴⁴ used individual bilayers of MoS₂-WS₂ heterostructures to develop photodetectors which showed photoresponsivity of 6.72×10^3 A W⁻¹ and detectivity of 3.09×10^{13} Jones at 457 nm laser illumination. The performance of bilayered MoS₂-WS₂ heterostructure photodetectors were several orders of magnitude higher compared with MoS₂ and WS₂ monocrystals. All-2D photodetectors were developed using the type-II band alignment of vertically stacked WS₂/MoS₂ heterobilayers and graphene electrodes.³⁵⁹ WS₂/MoS₂ heterobilayer-based photodetectors showed over an order of magnitude increase in the photoresponsivity than that of homobilayer photodetector and two orders of magnitude increase compared to WS₂ and MoS₂ monolayer-based photodetectors. The photoresponsivity of 10³ A W⁻¹ was measured under a power density of 1.7×10^2 mW cm⁻². The significant increase in photoresponsivity resulted from the strong Coulomb interactions occurring between WS₂ and MoS₂ layers.

Color-selective and flexible MoS₂ phototransistors on poly-arylate substrates using integrated Fabry-Perot cavity filters were developed by Yoo *et al.*³⁴⁵ The multilayer MoS₂ phototransistors showed a mobility of >64.4 cm² V⁻¹ s⁻¹ and an $I_{\text{on}}/I_{\text{off}}$ current ratio $>10^6$. The Fabry-Perot filters facilitated the coverage of the visible spectral range from 495 to 590 nm. The MoS₂ phototransistors showed no significant degradation after integrating the Fabry-Perot cavity and the SU-8 interlayer. Huo and Konstantatos^{346a} developed all-2D MoS₂ phototransistors using an out-of-plane vertical MoS₂ p-n junction. The vertical built-in field in the MoS₂ p-n junction reduced the recombination of the photoexcited carriers and yielded a photoresponsivity of 7×10^4 A W⁻¹, detectivity of 3.5×10^{14} Jones, a photoconductive gain of $>10^5$ and a fast photoresponse. Liu *et al.*^{346b} prepared a vertically oriented MoS₂ nanosheets array using TiO₂ buffer thin film on a SiO₂ substrate which also yielded a high photoresponse. The floating monolayer MoS₂



quantum well structure was also used to develop graphene/WS₂/MoS₂ heterostructure based photodetectors, which resulted in higher photoresponsivity of $4.4 \times 10^6 \text{ A W}^{-1}$ at 851 nm laser illumination under 30 fW laser power due to the strong photogating effect.³⁴⁷

5.2 Graphene electrodes

High photoresponsivities ranging from 10^3 A W^{-1} to 10^{10} A W^{-1} have been observed for the graphene/MoS₂ hybrid structure-based photodetectors.^{126,226–228} Han *et al.*³⁰⁰ developed photodetectors using CVD-grown MoS₂ monolayers with both Au and graphene electrodes. MoS₂ photodetectors with graphene electrodes showed the current $I_{\text{on}}/I_{\text{off}}$ ratio of 1×10^5 and mobility of $0.48 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ compared to 2×10^3 and $0.1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ with gold electrodes. The shot-noise-limited detectivity of MoS₂ photodetectors was 8.7×10^{14} Jones for the graphene electrode and 2.7×10^{14} Jones for the Au electrode for 1 Hz bandwidth at the gate voltage (V_g) = -30 V . The detectivity of CVD-grown MoS₂ photodetectors were a few orders of magnitude higher compared to photodetectors made using exfoliated MoS₂. Graphene/MoS₂/graphene photodetectors were also fabricated where the MoS₂ layer was encapsulated with a PMMA layer which showed mobility of $0.59 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, better than non-encapsulated devices. Photoresponsivity of the PMMA encapsulated graphene/MoS₂/graphene photodetectors were measured as $1.4 \times 10^5 \text{ A W}^{-1}$ and $1.1 \times 10^5 \text{ A W}^{-1}$ at 633 nm under applied gate voltage of 10 V and -30 V , respectively. The graphene electrodes were found to be instrumental in improving the photoresponse of the MoS₂ photodetectors. Chee *et al.*^{301a} used graphene/Ag contacts for MoS₂ photodetectors where CVD-grown graphene film was inserted as an interfacial layer between the MoS₂ film and Ag electrode. The MoS₂ FETs having graphene/Ag contacts exhibited the higher mobility of $35 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, photoresponsivity of 2160 A W^{-1} , and current $I_{\text{on}}/I_{\text{off}}$ ratio of 4×10^8 , compared to those of MoS₂ photodetectors with Ti/Au contacts due to the low work function of Ag and the Fermi level tunability in graphene. The n-doping by Ag electrode reduced the Fermi level of graphene, which consequently decreased the Schottky barrier height as well as contact resistance between the MoS₂ layer and electrodes. This study demonstrated that low-resistance contacts with MoS₂ and graphene played a significant role in enhancing the photoresponse of MoS₂ photodetectors. Bipolar phototransistors were developed by Li *et al.*^{301b} using vertical Au/graphene/MoS₂ vdWHS heterojunction where Au functions as the emitter and electrodes, MoS₂ as the collector, and graphene as the base of the bipolar phototransistor. Au/graphene/MoS₂ vdWHS heterojunction-based photodetectors showed the photoresponsivity of $16\,458 \text{ A W}^{-1}$ and detectivity of 1.75×10^{14} Jones at 405 nm laser illumination and incident power intensity of 0.45 mW cm^{-2} . Lee *et al.*^{301c} showed that the performance of MoS₂ photodetectors can be improved using graphene/MoS₂/graphene heterojunction-based phototransistors having short MoS₂ channel length greater than that of carrier diffusion length (220 nm) of MoS₂ and controllable Schottky barrier height (SBH). Fig. 23 shows the schematic illustration of MoS₂

phototransistor with a graphene gate electrode, change of photoresponsivity with MoS₂ channel length, the incident laser power dependent photoresponsivity and detectivity of graphene/MoS₂/graphene heterojunction-based phototransistors at different gate bias voltage and time-dependent normalized photocurrent. The distance between the graphene source and the drain electrode was measured as 30 nm by atomic force microscopy (AFM) technique. The short channel effects were not observed in MoS₂ transistors in the dark state in spite of their sub-30 nm channel length. The carrier recombination as well as the carrier transit time were found to be reduced by using sub-30 nm channel length which is much shorter compared to MoS₂ diffusion length. Longer MoS₂ channel length restricts the efficient flow of carriers from the source to the drain electrode. The Schottky barrier of phototransistors was manipulated by light and gate bias which supported decreased dark current and increased photocurrent. The SBH of the graphene/MoS₂ heterojunction was found to be decreased as the gate bias voltage was increased which supported the formation of ohmic contact. The SBH of the graphene/MoS₂ heterojunction was analyzed under different laser power intensity which was found to be 0.14 eV at $V_{\text{gs}} = -3 \text{ V}$ under the dark state. When 432 nm laser light was illuminated on the heterojunction, the SBH decreased with increasing laser power and, finally, disappeared at 0.14 μW laser power due to the photoinduced doping of graphene that eventually improved the performance of the graphene/MoS₂ heterojunction phototransistors. Both photoresponsivity and detectivity significantly changed as a function of incident laser powers. The graphene/MoS₂/graphene heterojunction-based phototransistor showed photoresponsivity of 1996 A W^{-1} and detectivity of 3.57×10^{10} Jones under laser power intensity of 0.34 nW at 432 nm laser wavelength and the maximum photocurrent of 2.15 μA with $I_{\text{photo}}/I_{\text{dark}}$ current ratio of 4×10^4 at laser power of 0.14 μW . These heterojunction-based phototransistors showed the maximum photoresponsivity, detectivity and response time of $2.2 \times 10^5 \text{ A W}^{-1}$, 3.5×10^{13} Jones and 2.8 ms at 432 nm, respectively, originating from the reduced MoS₂ channel length and controlled Schottky barrier in the graphene/MoS₂ heterojunction. These studies demonstrate that graphene electrodes played an important role in enhancing the performance of MoS₂ photodetectors.

5.3 Semiconducting (2H) and metallic (1T) phases of MoS₂

2D MoS₂ crystals exhibit several types of polymorphs, which have different electronic properties associated with semiconducting to metallic structural phase transformation.^{325a,b} The stable polymorphs of MoS₂ have been used in developing ultra-broadband photodetectors. Wang *et al.*^{325c} fabricated hybrid photodetectors using semiconducting trigonal prismatic 2H-phase and metallic octahedral 1T-phase MoS₂ nanosheets with organolead trihalide perovskites (MAPbX₃ where MA = methylammonium CH₃NH₃ and X = I). 1T-MoS₂/CH₃NH₃PbI₃ hybrid thin films showed 90% PL quenching due to the efficient charge transfer from CH₃NH₃PbI₃ to the 1T-MoS₂ while 2H-MoS₂/CH₃NH₃PbI₃ films showed 60% PL quenching due to the



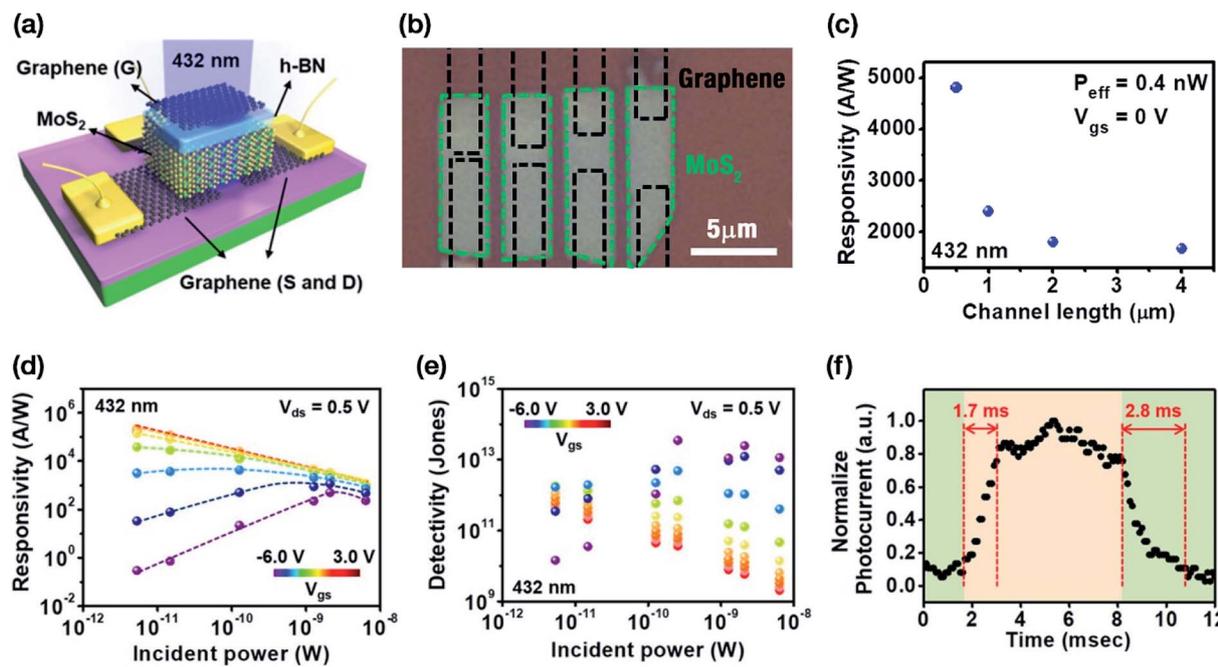


Fig. 23 (a) Schematic illustration of MoS₂ phototransistor with a graphene gate electrode. (b) Optical image showing different MoS₂ channel length. (c) Change of photoresponsivity as a function of MoS₂ channel length at 432 nm laser wavelength, (d) photoresponsivity (e) detectivity of graphene/MoS₂/graphene heterojunction-based phototransistors as a function of incident laser power intensity at different applied gate bias voltage. (f) Normalized photocurrent as a function of time measured under low intensity light at 80 Hz frequency. Reprinted with permission from ref. 301c, copyright © American Chemical Society.

injection of electrons and holes from CH₃NH₃PbI₃ to 2H-MoS₂ as proposed by Kang *et al.*¹⁹² 1T-MoS₂/CH₃NH₃PbI₃ hybrid-based photodetector showed photoresponsivity of 3096 A W⁻¹ and EQE value of 7.7 × 10⁵% at 500 nm under laser power density of 37.9 μW cm⁻² whereas 2H-MoS₂/CH₃NH₃PbI₃ hybrid-based photodetector exhibited comparatively lower photoresponsivity of 142 A W⁻¹ and EQE of 3.5 × 10⁴% at 500 nm under laser power density of 31.3 μW cm⁻². Multilayered 1T-MoS₂/CH₃NH₃PbI₃ hybrid photodetectors exhibited even higher photoresponsivity and EQE, due to the increased conductivity from the thicker multilayers of MoS₂. Multilayered 1T-MoS₂ and 2H-MoS₂ based photodetectors exhibited detectivity of 7 × 10¹¹ Jones and 2.6 × 10¹¹ Jones at 0.14 μW cm⁻² power density, respectively. This study showed that high photoresponsivity and EQE values can be obtained by blending CH₃NH₃PbI₃ perovskite with metallic 1T-phase MoS₂ nanosheets greater than that of semiconducting 2H-phase MoS₂ nanosheets. In another interesting study, Wang *et al.*^{325d} pointed out that photodetectors based on semiconducting 2H-phase MoS₂ show weak photoresponse in spite of their high optical absorption, whereas the metallic 1T-phase MoS₂ exhibit fast carrier transport but suffers from the low photoresponse to visible light. By integrating the positive traits of 2H and 1T phases of MoS₂, hybrid phototransistors were developed using a channel consisting of monolayer 2H-phase MoS₂ on top of 1T@2H-MoS₂ layer. The 1T@2H-MoS₂ has metal-like properties due to the lattice matching. The photodetectors developed from vertically stacked 2H-MoS₂/1T@2H-MoS₂ hybrid structure showed

photoresponsivity of 1917 A W⁻¹, detectivity of 7.55 × 10¹¹ Jones, and EQE of value of 448 384% at 532 nm wavelength under an illuminating power of 2.35 mW cm⁻² with applied bias of 20 V due to the existing metallic 1T-phase MoS₂ contents in the metal-like mixture of 1T@2H-MoS₂. The very high value of EQE in the 2H-MoS₂/1T@2H-MoS₂ photodetectors indicated the Schottky barrier modulated operation mechanism. The photoresponsivity, detectivity and EQE values depended on the illuminating power intensity and were found to be varied by few orders of magnitude with different photodetector device structures. The photodetector solely based on monolayer 2H-MoS₂ (without 1T@2H-MoS₂ layer) showed comparatively low photoresponse with a photoresponsivity of 56 A W⁻¹ and detectivity of ~1.3 × 10¹¹ under the same experimental conditions with source-drain bias voltage of 10 V. Furthermore, the 1T@2H-MoS₂ photodetectors showed extremely low photoresponsivity of 10⁻⁴ A W⁻¹, even much lower than that of 2H-MoS₂ photodetectors (56 A W⁻¹) under similar experimental conditions. The performance of photodetectors followed the sequence: 2H-MoS₂/1T@2H-MoS₂ > 2H-MoS₂ > 1T@2H-MoS₂. The dramatically high performance of 2H-MoS₂/1T@2H-MoS₂ photodetectors originated from the higher carrier mobility of the 1T@2H-MoS₂ layer and the synergistic interfacing between 2H-phase MoS₂ and mixed 1T@2H-MoS₂ layer.

5.4 Configuration of MoS₂ heterojunctions

The role of configuration of device heterojunctions has been emphasized for developing high performance MoS₂



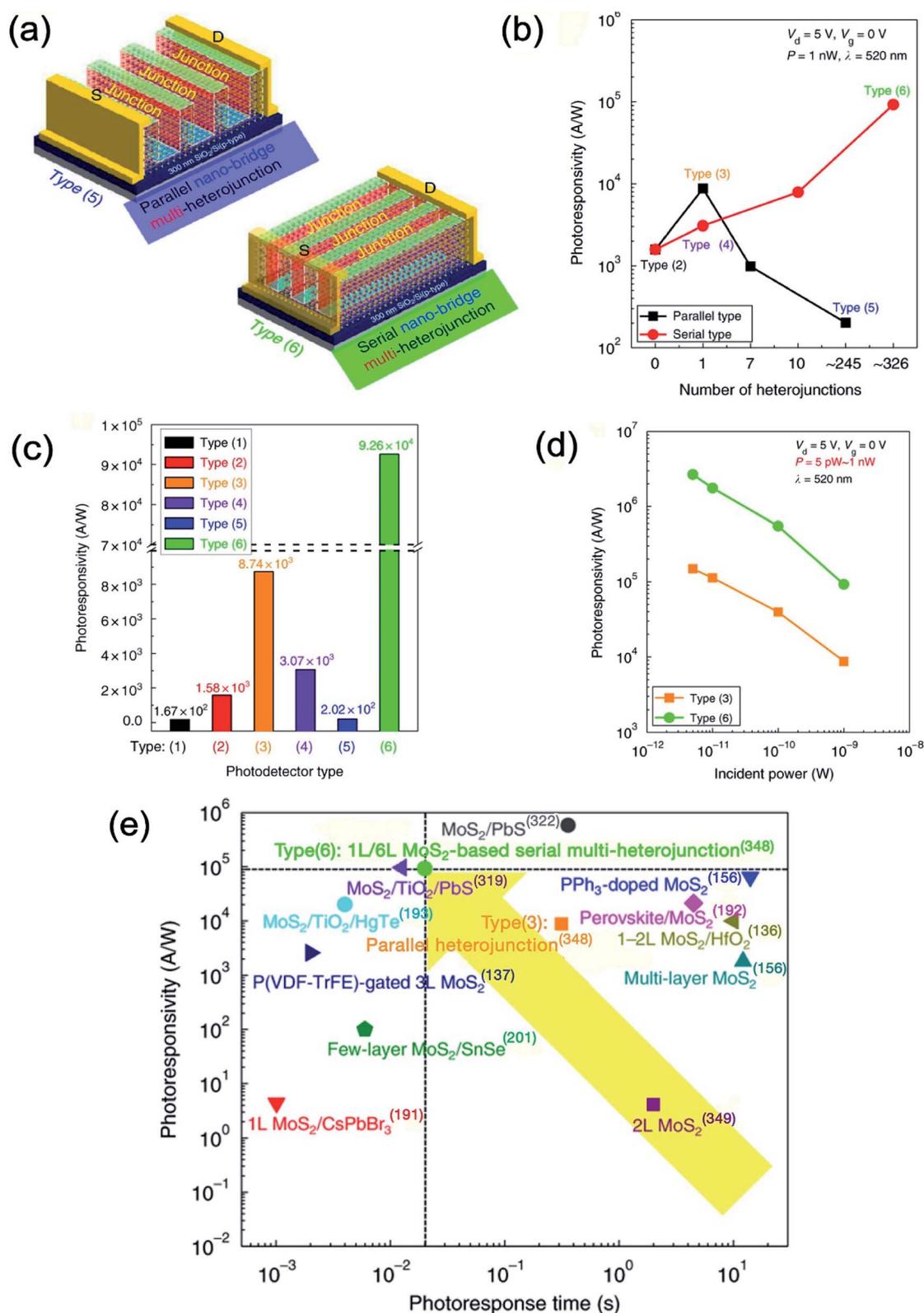


Fig. 24 (a) The schematic diagram of MoS₂ FETs for parallel nano-bridge multi-heterojunction type (5) and serial nano-bridge multi-heterojunction type (6) photodetectors. (b) Photoresponsivity as a function of the number and direction (parallel and serial) of heterojunctions in the MoS₂ channel. (c) The photoresponsivity of type (1) to (6) MoS₂ multi-heterojunctions. (d) Photoresponsivity of type (3) and (6) MoS₂ photodetectors as a function of the incident laser power at 500 nm. (e) A comparison of photoresponsivity and photoresponse time of parallel multi-heterojunction type (3) and serial multi-heterojunction type (6) based MoS₂ photodetectors with previously reported data on MoS₂ photodetectors (see ref. 136, 137, 156, 191–193, 201, 319, 322 and 349). Reprinted with permission from ref. 348, copyright © Springer Nature.



photodetectors. Kim *et al.*³⁴⁸ reported MoS₂ photodetectors using serial nano-bridge multi-heterojunctions. The MoS₂ photodetectors based on laser irradiation were assigned as type (1) monolayer, type (2) multilayer, type (3) parallel heterojunction, type (4) serial heterojunction, type (5) parallel nano-bridge multi-heterojunction, and type (6) serial nano-bridge multi-heterojunction. Fig. 24 shows a schematic diagram of MoS₂ FETs for parallel nano-bridge multi-heterojunction type (5) and serial nano-bridge multi-heterojunction type (6) photodetectors, photoresponsivity as a function of the number of heterojunctions in the MoS₂ channel and the incident laser power as well as their comparison with previously reported data on MoS₂ photodetectors.^{136,137,156,191–193,201,319,322,349} Photoresponsivity of type (2) MoS₂ photodetectors ($1.58 \times 10^3 \text{ A W}^{-1}$) was found to be ~ 9.46 folds higher compared with type (1) photodetectors ($1.67 \times 10^2 \text{ A W}^{-1}$) because more photocarriers were generated by a greater number of layers and lower exciton binding energy associated with 6L-MoS₂ (0.2 eV). The photoresponsivities of type (3) and type (4) MoS₂ photodetectors were $8.74 \times 10^3 \text{ A W}^{-1}$ and $3.07 \times 10^3 \text{ A W}^{-1}$ which were ~ 5.53 and ~ 1.94 times higher compared with type (2) photodetectors ($1.58 \times 10^3 \text{ A W}^{-1}$), respectively, arising from the easier electron-hole pair generation. The highest photoresponsivity of $2.67 \times 10^6 \text{ A W}^{-1}$ at $\lambda = 520 \text{ nm}$ and then $1.65 \times 10^4 \text{ A W}^{-1}$ at $\lambda = 1064 \text{ nm}$ were observed for the MoS₂ serial-type heterojunctions with ~ 326 , and monolayer/multilayer (6L) heterojunctions.

The mechanisms of photoconductivity in atomically thin layered MoS₂ has been described by Furchi *et al.*³⁴⁹ Sun *et al.*³⁵⁰ developed photodetector using multilayer/monolayer MoS₂ heterojunction where the source electrodes were connected to the 0.65 nm thick MoS₂ monolayer and the drain electrodes were connected with the 6.9 nm thick MoS₂ multilayer and the back gate was heavily p-doped Si. The multilayer/monolayer MoS₂ heterojunction showed photoresponsivity of 10^3 A W^{-1} , detectivity of 7×10^{10} Jones and photosensitivity of 1.7×10^5 at 470 nm. Abnormal photoresponse was noticed under positive gate voltage due to the heterojunction formation. Yan *et al.*³⁵¹ fabricated photodetectors using CVD-grown MoS₂ bilayer flakes which showed photoresponsivity of 7160 A W^{-1} , detectivity of 6.62×10^{10} Jones and response/recovery times of 97 ms/291 ms, compared to photoresponsivity of 2900 A W^{-1} and detectivity of 2.44×10^{11} Jones for the monolayer MoS₂ flakes.

5.5 MoS₂/inorganic semiconductor heterostructures

Gallium nitride (GaN), a wide-bandgap semiconductor, has been used with MoS₂ for developing hybrid photodetectors. The self-powered few-layer MoS₂/GaN heterojunction-based deep-ultraviolet (DUV) photodetector was developed by Zhuo *et al.*³¹⁶ In order to develop a MoS₂/GaN p-n heterojunction photodetector, the MoS₂ thin films were transferred onto a GaN substrate where e-beam evaporation method was used to deposit Au electrode on the MoS₂ film and Ni/Au electrode on the GaN layer. The MoS₂/GaN photodetector exhibited the current $I_{\text{on}}/I_{\text{off}}$ ratio of 10^5 at 2.4 mW cm^{-2} and over 10^3 at a laser light intensity of $2 \mu\text{W cm}^{-2}$, indicating the detection of a weak UV light signal under 265 nm laser illumination at zero bias

voltage (0 V) without any external power (self-powered mode). The linear dynamic range (LDR) of 97.3 dB was calculated at zero bias voltage, much better than those of other photodetectors. The rise/fall times were found to be 0.302 ms and 3.605 ms at 100 Hz which further improved at 5 kHz. The MoS₂/GaN photodetectors exhibited photoresponsivity of 187 mA W^{-1} , detectivity of 2.34×10^{13} Jones, fast response speeds of $46.4 \mu\text{s}/114.1 \mu\text{s}$ at 5 kHz under a DUV light illumination of 265 nm at 0 V bias. Liu *et al.*^{317a} fabricated ML MoS₂-based phototransistor on a transparent GaN wafer using CVD technique. High quality MoS₂/GaN hybrid films were obtained because of the small thermal expansion coefficient (TEC) mismatch and near lattice match between MoS₂ and GaN. The photocurrent MoS₂/GaN-based FETs was found to be 3–4 times higher at 405 nm compared with 638 nm arising from high absorption and photocurrent gain process. The value of EQE also increased significantly, from 115% at 638 nm to 5289% at 405 nm. The photoresponsivity of 0.59, 2.78, and 17.2 A W^{-1} was measured at 638, 532, and 405 nm wavelengths under 2 mW laser power, respectively. The response time (t_{rise}) decreased from 4.2 s at 638 nm to 0.1 s at 405 nm. MoS₂/GaN hybrid-based photodetector showed the maximum photoresponsivity of 17.2 A W^{-1} , EQE of 5289%, and photocurrent gain of 53.6 at 405 nm under incident laser-power of 2 mW using an applied voltage of 9 V. The specific detectivity and the low noise equivalent power (NEP) varied from 6.3×10^{10} to 1.8×10^{12} Jones and 2.8×10^{-12} to $9.6 \times 10^{-14} \text{ W Hz}^{-1/2}$ in 405 nm to 638 nm wavelength range, respectively. Li *et al.*^{317b} demonstrated the improvement of photoresponse of ML MoS₂/GaN photodetector by depositing 3 nm thick Al₂O₃ layer on the surface with ALD method. The photocurrent of Al₂O₃/MoS₂/GaN heterostructure-based photodetector increased more than twice compared to MoS₂/GaN device at 365 nm wavelength under applied voltage of 20 V due to the tensile strain enhancement. The Al₂O₃/MoS₂/GaN photodetector showed over two-fold increase in the photoresponsivity, EQE, and photogain than that of MoS₂/GaN photodetector where EQE and photogain values increased from 3848% to 8381% and 239 to 520, respectively.

Indium selenide (InSe) is a well-known photodetector.^{352a-d} The MoS₂ QDs decorated indium selenide (InSe) nanosheets were applied to develop hybrid phototransistors which yielded the photoresponsivity of $9.304 \times 10^3 \text{ A W}^{-1}$, three orders (10^3) of magnitude higher than that of InSe photodetector (12.3 A W^{-1}).³³⁶ The higher photoresponsivity of hybrid photodetector originated from the MoS₂ QDs injected photoexcited carriers to the InSe phototransistor. Ulaganathan *et al.*³⁵³ decorated MoS₂ quantum dots (QDs) with InSe nanosheets to develop hybrid phototransistors. The MoS₂/InSe-FET showed photoresponsivity of 9304 A W^{-1} under illumination at 488 nm at source-drain voltage (V_{ds}) = 1 V and gate voltage (V_{g}) = 0 V, which was 10^5 times higher compared to InSe based photodetector (0.101 A W^{-1} at 532 nm) and nearly 10 times higher compared to 2D-graphene/2D-InSe hybrid photodetector (940 A W^{-1} at 532 nm).^{352d} Here again, the higher photoresponsivity of MoS₂/InSe hybrid photodetectors originated due to the transfer of photogenerated charge carriers from MoS₂ QDs to the InSe nanosheets. $\beta\text{-In}_2\text{Se}_3$ is a group IIIA–VIA atomic layered



semiconducting material having a small bandgap, which has been used for developing MoS₂ hybrid photodetectors. The few layer β-In₂Se₃ thin films were epitaxially grown on the top of CVD-MoS₂ monolayers by Mahapatra *et al.*³⁵⁴ The β-In₂Se₃/MoS₂ vdW heterostructure-based photodetectors showed significantly higher photoresponse than that of pure β-In₂Se₃ photodetectors. The photocurrent of In₂Se₃/MoS₂ hybrid-based devices was found to be 1.3×10^3 times higher compared with the dark current at 532 nm under laser power of 8.47 μW and the EQE value reached $5.49 \times 10^3\%$ under laser power of 4 mW cm⁻² with applied bias.

Silicon³⁵⁵ has been widely used for developing broadband hybrid photodetectors for communication technology. Guo *et al.*³⁵⁶ used MoS₂/Si heterostructure to fabricate a photodetector showing photoresponsivity of 746 mA W⁻¹ and detectivity of 6.03×10^{11} Jones and response/recovery time of 178 μs/198 μs in the 405 to 980 nm range. In another study, multilayer MoS₂ films were deposited on Si quantum dots (Si QDs) to develop heterojunction photodetectors which showed much faster response/recovery time of 60 ns/756 ns and detectivity of 6.1×10^{13} Jones.³⁵⁷ Au nanostructure-based plasmonic-enhanced multilayer MoS₂ photodetector deposited on p-type Si substrate exhibited photoresponsivity of ~ 37 A W⁻¹ and detectivity of $\sim 10^{12}$ Jones in 405–780 nm wavelength range.³⁵⁸ The photoresponsivity of the Au plasmonic-enhanced MoS₂/Si photodetector was enhanced by 8.0, 5.3 and 11 times at 405 nm, 650 nm and 780 nm under 5 V bias, respectively, compared to the pristine p-Si photodetector. The Au plasmonic-enhanced MoS₂/Si photodetector also displayed a fast response time of 1 μs and a recovery time of 18 μs.

The 2D MoS₂/1D ZnO heterostructure-based photodetectors have been developed to increase the light absorption range and photoresponsivity.³⁶⁰ The dark current of p-MoS₂/n-ZnO heterostructure was measured as 0.88 nA which increased significantly to 103.1 nA and 2.52 nA at 365 nm (light intensity of 112 mW cm⁻²) and 532 nm (64 mW cm⁻²) under +5 V of applied voltage, respectively, demonstrating high sensitivity to UV and visible illuminations. The current I_{on}/I_{off} ratio was found to be 109 at 365 nm and 3.3 at 532 nm. The n-MoS₂/n-ZnO photodetectors showed the photocurrents of 33.6 nA to 88.2 nA and 0.3 nA to 1.86 nA at 365 nm (5.7 to 47.2 mW cm⁻²) and at 532 nm (0.525 to 56.7 mW cm⁻²), respectively. The p-MoS₂/n-ZnO photodetectors showed photoresponsivity of 24.36 A W⁻¹ and EQE of $8.28 \times 10^3\%$ under 365 nm light illumination.

Semimetal cadmium arsenide (Cd₃As₂) has been used for fabricating broadband photodetectors due to its higher mobility as well as high absorption of light in a broad spectral region.³⁶¹ Cd₃As₂ nanoplates have been integrated with multilayer MoS₂ to develop a heterojunction photodetector which yielded high photoresponsivity of 2.7×10^3 A W⁻¹ at room temperature.³⁶² Likely, CdSe nanoplates were vertically assembled on MoS₂ monolayer to develop vertical CdSe/MoS₂ heterostructures.³⁶³ CdSe/MoS₂ heterostructure-based photodetector exhibited photoresponsivity of 1.63 A W⁻¹ and fast response speed of 370 μs. In another study, 2D MoS₂ nanosheet and 0D CdSe nanocrystal-based hybrid phototransistor were developed which showed substantially enhanced photoresponsivity of 2.5

$\times 10^5$ A W⁻¹ and the rise/fall times of 60 ms.³²³ The photocurrent of MoS₂/CdSe hybrid photodetector increased with increasing laser power intensities at 405 nm. Ghosh *et al.*³⁶⁴ developed photodetectors using a nanocomposite of single flake MoS₂ and lanthanide doped upconversion nanoparticles (UCNPs). The MoS₂/UCNPs based photodetector showed highest photoresponsivity of 1254 A W⁻¹, detectivity of 1.05×10^{15} Jones and a gain of 7.12×10^{-4} cm² V⁻¹ at 980 nm for 1.0 V bias. The highest detectivity of 9.0×10^{13} Jones at 405 nm and the lowest detectivity of 3.4×10^{13} Jones at 808 nm was observed for MoS₂/UCNPs devices. Likely, MoS₂/UCNPs photodetectors showed the highest normalized gain (I_n) of 1.48×10^{-4} cm² V⁻¹ at 405 nm and the lowest I_n of 2.8×10^{-5} cm² V⁻¹ at 808 nm laser wavelength, which is an order of magnitude higher compared with those reported for MoS₂ monolayer ($I_n = 4.8 \times 10^{-6}$ cm² V⁻¹).¹⁷¹ Multilayer MoS₂ films on monocrystalline SiC substrate using CVD were deposited by Xiao *et al.*³⁶⁵ MoS₂/SiC hybrid photodetector showed photoresponsivity of 5.7 A W⁻¹ at 365 nm under 4.35 μW incident light power. Instead of Si, Wu *et al.*³⁶⁶ used Si nanowire arrays (SiNWA) with few-layer MoS₂ films to enhanced light absorption region of photodetectors. The dark current of the MoS₂/SiNWA photodetector was found to be much lower compared with MoS₂/bulk Si photodetector; contrary to this, the photocurrent of the MoS₂/SiNWA photodetectors was much increased than that of the MoS₂/bulk Si photodetector due to the increased light absorption and contact area between MoS₂ and Si. The I_{on}/I_{off} current ratio of MoS₂/SiNWA photodetectors varied from 3.2×10^2 to 1.1×10^5 as the light intensity changed from 0.031 mW cm⁻² to 32 mW cm⁻² under zero bias at 650 nm. The MoS₂/SiNWA heterojunction-based photodetector showed photoresponsivity of 53.5 A W⁻¹, and detectivity of 2.8×10^{13} Jones under laser power intensity of 6.3 μW cm⁻², much higher than that of MoS₂/bulk Si photodetector.^{311–313} The rise/fall times of photodetectors was found to decrease with increasing light intensity where the rise times varied from 54.2, 27.7, 23.5 to 22.8 μs, and fall times from 69.9, 68.6, 67.4 to 61.5 μs under laser power intensity of 2.5, 16.8, 24.5 and 38.6 mW cm⁻², respectively.

Organic/inorganic hybrid halide perovskite (C₆H₅C₂H₄-NH₃)₂PbI₄ = (PEA)₂PbI₄ with ML MoS₂ has been used to develop hybrid photodetectors by Wang *et al.*³⁶⁷ (PEA)₂PbI₄ functions as an electron reservoir to decrease free charge carriers as well as a to passivate defects. The dark current was reduced by six orders of magnitude by depositing (PEA)₂PbI₄ thin film over MoS₂ photodetector due to the charge transfer from ML MoS₂ to (PEA)₂PbI₄ thin film. The use of (PEA)₂PbI₄ over ML MoS₂ reduced the charge carrier density which yielded a broadband photodetector (200 to 900 nm) exhibiting photoresponsivity of 16.8 A W⁻¹, detectivity of 1.06×10^{13} Jones, EQE of 3.3×10^3 and on/off ratio of 10⁵. The photoresponse speed of a hybrid photodetector was enhanced over 100-times that of a pristine MoS₂ photodetector due to (PEA)₂PbI₄ passivation. (PEA)₂PbI₄/MoS₂ hybrid photodetectors also work at 0 V bias, which is a self-powered mode where photocurrent showed an increase with increasing laser power intensity from 2.8 nW to 109.8 nW and the rise/decay times of 38 ms/24 ms under 637 nm laser



wavelength. Shen *et al.*³⁶⁸ synthesized a series of helicene 5,14-diaryldiindeno[2,1-*f*:1',2'-*j*]picene (DDP) derivatives namely 5,14-dimesityldiindeno[2,1-*f*:1',2'-*j*]picene (1ab), 5,14-bis(2,4,6-trimethoxyphenyl)diindeno[2,1-*f*:1',2'-*j*]picene (1ac), and 9,10-dimethyl-5,14-dimesityldiindeno[2,1-*f*:1',2'-*j*]picene (1bb) and integrated with monolayer MoS₂ to develop hybrid photodetectors. Time-resolved measurements showed the interfacial charge-transfer from the DDP derivative to the monolayer MoS₂ confirmed by the stability of exciton property of the (1ac)/MoS₂ organics/inorganic hybrid heterostructure. The 1ac/MoS₂ hybrid based photodetector showed high photoresponsivity of 4.99×10^7 A W⁻¹ and response time of 45 ms at 633 nm using laser light intensity of 5.75 nW under $V_d = 5$ V and $V_g = 0$ V because of the efficient separation of photoexcited carriers and the alignment of type-II energy band. The photoresponsivity of the organic/inorganic hybrid photodetectors followed the sequence of (1ac)/MoS₂ > (1ab)/MoS₂ > (1bb)/MoS₂ as a function of light intensity and after storing for one month. Furthermore, 1ac/MoS₂ hybrid photodetector did not exhibit any degradation after one-month storage. MoS₂ hybrid heterostructure-based photodetectors have been mainly studied having a type II band alignment.

The MoS₂ atomic layers were also prepared by different methods. Kumar *et al.*³⁶⁹ developed large-area MoS₂ layers using pulsed laser deposition (PLD) method which showed photoresponsivity of 3×10^4 A W⁻¹ and detectivity of 1.81×10^{14} Jones at 365 nm under $24 \mu\text{W cm}^{-2}$ incident light power at applied bias of 2.0 V. The photoresponsivity of the PLD-grown few-layer MoS₂ photodetectors was found to be 3×10^4 , 1.08×10^4 , 7.0×10^3 , and 6.3×10^3 A W⁻¹ at 365, 436, 546, and 655 nm wavelengths under applied bias of 2 V, respectively. FL-MoS₂ photodetectors also showed low dark current of 10^{-10} A and photoresponse of 1.37×10^5 . Schneider *et al.*³⁷⁰ used metalorganic vapor-phase epitaxy (MOVPE) method to develop FL MoS₂ based flexible photodetectors. The photoresponsivity and specific detectivity of MoS₂ photodetectors can be changed between 150 A W⁻¹ to 920 A W⁻¹ and 10^{12} Jones to 10^{10} Jones by using electrostatic gating, respectively.

Singh *et al.*³⁷¹ reported Pd/Al₂O₃/MoS₂/ITO photodetector showing photoresponsivity of 488 A W⁻¹, detectivity of 8.22×10^{12} Jones and EQE of $1.9 \times 10^5\%$ at 308 nm wavelength under laser light intensity of $13.6 \mu\text{W/cm}^2$ with 1 V applied bias voltage. Ye *et al.*³⁷² demonstrated vertical growth of Sb₂O₃ flakes on monolayer MoS₂ crystals by using CVD technique. The Sb₂O₃ flakes of different thickness and size covered monolayer MoS₂ crystals in 10, 21, 44, 54 and 100% ratio and their based back-gated FETs fabricated on SiO₂/Si wafer affected the carrier mobility and subthreshold swing (SS) of the Sb₂O₃/MoS₂ hybrid photodetectors. The photocurrent, photoresponsivity, detectivity, sensitivity and EQE of hybrid FETs were measured as a function of laser power intensity under 360, 457, 532, 671, 914, and 1064 nm light illuminations. The hybrid FET-21% exhibited photocurrents of 263 and 2.5 μA at laser power intensities of 177 and 11 mW cm^{-2} under 360 and 1064 nm laser illuminations, respectively. The photoresponsivity of 5.3×10^4 A W⁻¹ and detectivity of 2.0×10^{15} Jones were measured at 457 nm wavelength whereas in the NIR region, Sb₂O₃/MoS₂ hybrid

photodetectors showed photoresponsivity of 7.8 A W⁻¹, detectivity of 3.4×10^{11} Jones, and response speed of <60 ms with 1064 nm laser wavelength at room temperature. Sb₂O₃/MoS₂ hybrid photodetectors (FET-21%) showed photoresponsivity of 1.1×10^4 A W⁻¹ and detectivity of 4.5×10^{14} Jones under 360 nm laser wavelength. The hybrid FET-21% also showed sensitivity values of 6×10^2 , 2.5×10^3 , 3×10^3 , 2.4×10^3 , 0.2, and $0.5 \text{ cm}^2 \text{ W}^{-1}$ and EQE values of 4.0×10^6 , 1.4×10^4 , 1.0×10^4 , and 5.5×10^3 , 0.4, and 0.9% under laser illuminations of 360, 457, 532, 671, 914, and 1064 nm, respectively. Higher photoresponse has been observed in 2D metal NiTe₂ and semiconductor MoS₂ heterostructure-based back-gated FETs and photodetectors compared to a pristine MoS₂ monolayer.³⁷³ As discussed earlier, that the MoS₂ vdWH-based photodetectors show great promise for practical applications, therefore, new vdW heterostructures have been targeted. The NiTe₂/MoS₂ vdW heterostructure-based photodetectors showed several times faster rise/decay times than that of a MoS₂ photodetector due to the epitaxial grown metallic vdWHs. Photodetectors were also developed from the WSe₂ and MoS₂ vdWHs where MoS₂ was used as a channel in the phototransistor.³⁷⁴ The MoS₂ vdWH-based phototransistor showed high photoresponsivity of 2700 A W⁻¹, detectivity of 5×10^{11} Jones, and response speed of 17 ms. In these phototransistors, the vertical built-in electric field in the WSe₂-MoS₂ p-n junction separated the photoexcited charge carriers, which yielded a photoconductive gain of 10^6 . Ying *et al.*³⁷⁵ fabricated BaTiO₃/MoS₂ heterostructure-based photodetectors with type I band alignment which showed photoresponsivity of 120 A W⁻¹ and EQE of $4.78 \times 10^4\%$ compared with 1.7 A W⁻¹ and $4.5 \times 10^2\%$ for the bare MoS₂ at 365 nm under low laser power intensity of 0.095 mW cm^{-2} . Both micro-Raman spectroscopy and photoluminescence indicated the occurrence of carrier extraction and carrier injection processes in the MoS₂ layer, leading to the boosting of the number of carriers in the MoS₂ channel. The high performance of type-I hybrid heterostructure-based photodetectors originated from the carrier extraction process between BaTiO₃ NPs and MoS₂ layer.

5.6 MoS₂ nanoscrolls

MoS₂ nanoscrolls have been prepared by argon plasma treatment of monolayer MoS₂ nanosheet where the MoS₂ nanoscroll formation also results in a partial removal of sulfur atoms from the top sulfur layer in MoS₂.³⁷⁶ The luminescent MoS₂ nanoscrolls have also been prepared by the supercritical fluid processing method. The exfoliated MoS₂ nanoscrolls showed photoluminescence from 420 to 600 nm due to the size reduction and polydispersity.³⁷⁷ The 1D MoS₂ nanoscrolls have been prepared from CVD-grown triangular MoS₂ nanosheets where the formation of a MoS₂ nanoscroll takes place from the edge of a triangular nanosheet along the armchair orientation of MoS₂.³⁷⁸ The carrier mobility as well as physical contact properties of 1D MoS₂ nanoscroll-based FETs were found to be quite different than that of 2D MoS₂ nanosheets. The 2D semiconductor MoS₂ nanosheets can be transformed into 1D metallic MoS₂ nanoscrolls. This demonstrated that 1D TMD-



based nanoscrolls with unique chemical, physical and electrical properties can be fabricated.

The photodetectors fabricated using MoS₂ or WS₂ nanoscroll were compared with monolayer MoS₂ or WS₂ photodetectors.³⁷⁹ MoS₂ or WS₂ nanoscroll-based devices showed photosensitivity enhancement by two orders of magnitude, demonstrating the potential of 2D TMD-based 1D nanoscrolls in optoelectronic applications. 1D MoS₂ and WS₂ nanoscroll-based photodetectors exhibited very high photocurrent-to-dark-current ratio compared with 2D nanosheet-based photodetectors. Deng *et al.*³⁸⁰ prepared MoS₂ nanoscroll-based photodetectors using the avalanche multiplication effect where a low triggering electrical field is required than that of MoS₂ atomic layers. Fig. 25(a–c) shows schematic illustration of MoS₂ nanoscroll-based avalanche photodetectors having MoS₂ nanoscroll channel (area of 1.8 μm²) connected with source and drain metal electrodes, photoresponsivity and detectivity as a function of applied bias voltage (V_{ds}) under 532 nm laser illumination. Self-assembled MoS₂ nanoscrolls were prepared using ethanol droplet and by rolling the monolayer MoS₂ flake. The doped silicon film was used as the substrate as well as a gate electrode. A MoS₂ nanoscroll is formed under strain compared with a monolayer MoS₂ flake, therefore, an analysis of the MoS₂ bandgap was carried out under applied tensile strain. As the uniaxial tensile strain was increased from 0% to 3% for the

monolayer MoS₂ flake, the bandgap of MoS₂ decreased from 1.77 to 1.5 eV, associated to a decrease in avalanche critical electric field (E_{ava}) in MoS₂ nanoscrolls. The avalanche multiplication effect was noticed in MoS₂ nanoscrolls which also showed high multiplication factor and impact ionization coefficient. The excess noise factor was measured as a function of the multiplication factor. The multiplication factor of 40 was recorded at V_{ds} of 45 V which depended on the applied electric field. The avalanche gain of 24 was also measured at $V_{ds} = 45$ V for the MoS₂ nanoscroll-based devices. The electrical field (E_{ava}) to trigger avalanche multiplication in MoS₂ nanoscrolls was found to be dramatically decreased compared with monolayer MoS₂ flakes. The decrease of E_{ava} in MoS₂ nanoscrolls was associated with several factors including reduced bandgap, substrate contacts and phonons suppressed scattering of the free carriers. The decrease in bandgap of MoS₂ nanoscroll was also evidenced by a red-shift compared with the monolayer MoS₂ flake. MoS₂ nanoscroll-based photodetectors exhibiting photoresponsivity over 10⁴ A W⁻¹, detectivity of 2 × 10¹² Jones and 30 times higher avalanche gain compared with monolayer MoS₂ flake. This study showed low-power consumption MoS₂ nanoscroll-based photodetections where avalanche multiplication effect enhanced the photoresponsivity in MoS₂ nanoscrolls compared with atomically thin monolayer MoS₂. Deng *et al.*³⁸¹ reported photodiodes having p-type WSe₂ and single MoS₂

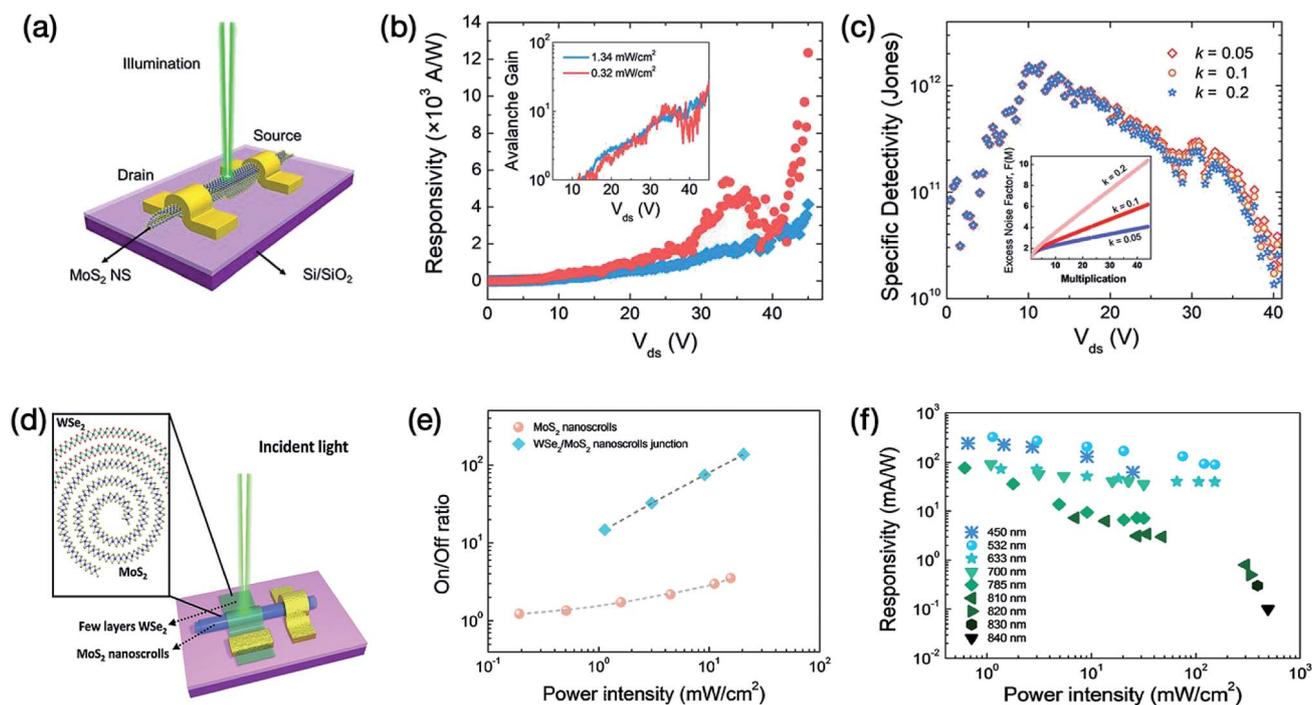


Fig. 25 (a) Schematic illustration of MoS₂ nanoscroll-based avalanche photodetector under illumination. (b) Photoresponsivity of the avalanche photodetector as a function of applied bias voltage (V_{ds}) under 532 nm laser illumination at different incident light power intensities. Inset shows the V_{ds} dependent avalanche gain. (c) Specific detectivity of the avalanche photodetector as a function of V_{ds} . The inset is a plot of excess noise factor as a function of the multiplication factor. Reprinted with permission from ref. 380, copyright © American Chemical Society. (d) Schematic illustration of the WSe₂/MoS₂ nanoscroll-based photodiode along with the cross-section of the heterojunction containing MoS₂ nanoscrolls and few layers WSe₂ where electrodes are source and drain, respectively. (e) The current I_{on}/I_{off} ratio of single MoS₂ nanoscroll and WSe₂/MoS₂ nanoscroll-based heterojunction as a function of incident power intensity at 532 nm wavelength. (f) Photoresponsivity of the WSe₂/MoS₂ nanoscroll-based hybrid photodetectors as a function of incident laser power intensity at different wavelengths, showing broadband photo-detection by the hybrid heterostructure photodiode. Reprinted with permission from ref. 381, copyright © Wiley.



nanoscrolls which showed suppressed dark current and two orders of magnitude increase in the ratio of photocurrent to dark current compared with a single MoS₂ nanoscroll-based photodiode. Fig. 25(d–f) shows schematic illustration of the WSe₂/MoS₂ nanoscroll-based photodiode, incident power intensity dependent current $I_{\text{on}}/I_{\text{off}}$ ratio of single MoS₂ nanoscroll and WSe₂/MoS₂ nanoscroll-based heterojunction and photoresponsivity of the WSe₂/MoS₂ nanoscroll-based hybrid photodetector as a function of incident power intensity. The response speed of WSe₂/MoS₂ nanoscroll-based device increased by three orders of magnitude than that of a single MoS₂ nanoscroll device. The WSe₂/MoS₂ nanoscroll-based photodetectors exhibited the photoresponsivity of 0.3 A W⁻¹, EQE values of 25% at zero bias voltage and 75% at a reverse bias of 1 V at 532 nm under laser power intensity of 1.14 mW cm⁻² and response speed of 5 ms with laser power intensity of 60 mW cm⁻². The atomic layered WSe₂/MoS₂ nanoscrolls were found to be superior to those of MoS₂ nanoscrolls. Wang *et al.*³⁸² developed WS₂/MoS₂ vdWH-based nanoscrolls by dropping aqueous alkaline droplets on CVD-grown BL WS₂/MoS₂ vdW hybrid heterostructure, which were prepared by growing 1L WS₂ islands on top of 1LMoS₂ nanosheets. The optical microscopy, AFM, ultralow frequency (ULF) Raman spectroscopy and TEM techniques were used to characterize the WS₂/MoS₂ vdWH-based nanoscrolls and ULF breathing and shear mode peaks were observed which originated from the strong interlayer interaction. The photosensitivity of WS₂/MoS₂ nanoscroll-based photodetectors were found to be ten times higher compared with WS₂/MoS₂ vdWH-based photodetectors under blue, green and red laser illuminating lights; the ultrafast charge transfer process occurring in nanoscroll structure at alternative WS₂/MoS₂ and MoS₂/WS₂ multi-interfaces could be associated with such a high performance.

The research data collected on the figure-of-merit of MoS₂ based photodetectors have been extensively documented. Tremendous interest is growing in the field of MoS₂ based photodetectors as new research ideas are emerging on a rapid pace.^{383–387} Ahmed *et al.*³⁸⁸ fabricated graphene/h-BN/MoS₂ tri-layer vdW heterostructure-based phototransistors to examine

the photogating mechanism where h-BN layer was used as an interfacial barrier to monitor the charge transfer process. Mechanically exfoliated monolayer graphene, MoS₂ and multi-layer h-BN were used to fabricate the graphene/h-BN/MoS₂ vdW heterostructures on the SiO₂/p⁺⁺-Si substrate. All optoelectronic measurements were conducted at 180 K temperature under the high vacuum conditions. The incident power density dependent photoresponsivity, NEP and detectivity of vdW heterostructure-based photodetectors were measured from 640 nm to 1720 nm wavelength region. Fig. 26 shows the figure of merit of the graphene/h-BN/MoS₂ vdW heterostructure-based photodetectors. The photoresponsivity values of 5 × 10⁹ A W⁻¹ in the visible region, 2 × 10⁹ A W⁻¹ at 940 nm, 10⁸ A W⁻¹ at 1550 nm and 2 × 10⁷ A W⁻¹ at 1720 nm were observed using source drain bias of 20 mV at low power density. The low NEP value of 2 × 10⁻²⁰ W Hz^{-1/2} at 940 nm was found to be further increased to 2 × 10⁻¹⁸ W Hz^{-1/2} for 1720 nm. Being correlated to NEP, the high detectivity values of 10¹⁶ Jones at 640 nm, 5 × 10¹⁵ Jones at 940 nm and 5 × 10¹³ Jones at 1720 nm were deduced for the vdW hybrid photodetectors, which are two orders of magnitude higher compared to graphene-based IR photodetectors. The values of photoresponsivity, NEP and detectivity for vdWHs differ by several orders of magnitude as a function of laser power density at different measurement wavelengths. This study demonstrated that the defect states can strongly affect to the photoresponse of the graphene/h-BN/MoS₂ hybrid photodetectors. The strong photoresponse has been observed in MoS₂ atomic layers when integrated with other vdW nanomaterials. As summarized in Table 1 and documented throughout this article, MoS₂-based photodetectors show photoresponsivity ranging from mA W⁻¹ to 10¹⁰ A W⁻¹, detectivity from 10⁷ to 10¹⁵ Jones and ultra-fast response speed up to nanoseconds (10⁻⁹ s), which varies by several orders of magnitude over an ultra-broadband spectral range from 265 nm to 10 μm under different incident power densities. The performance of MoS₂ photodetectors is derived from the design of MoS₂ based phototransistors, photoconductors and photodiodes. Several different mechanisms including photoconductive, photovoltaic, photobolometric and surface plasma-wave

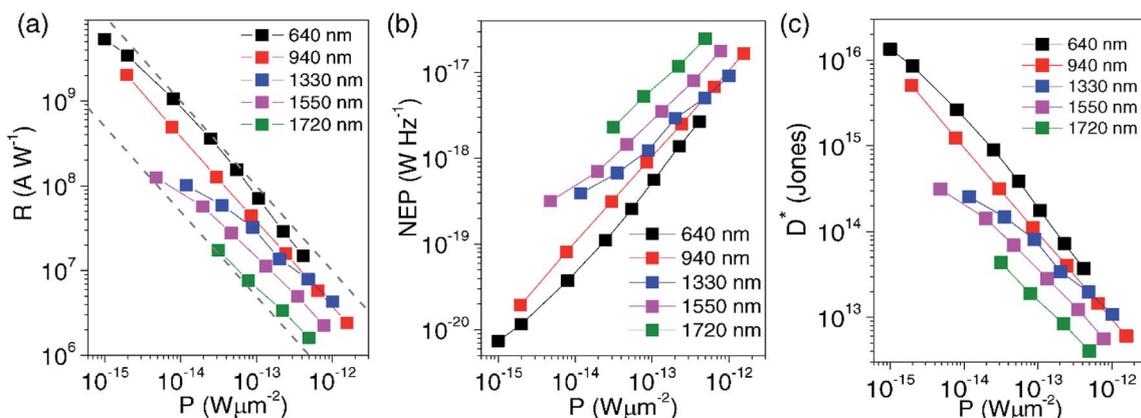


Fig. 26 Figure of merit of the graphene/h-BN/MoS₂ vdW heterostructure-based photodetectors. Plots of (a) photoresponsivity (R), (b) noise equivalent power (NEP) and (c) specific detectivity (D^*) as a function of incident power density (P) in the 640 nm to 1720 nm wavelength region. Reprinted with permission from ref. 388, copyright © Institute of Physics Publishing.



enhanced effects contribute to the performance of MoS₂ photodetectors. Abraham and Majumdar³⁸⁹ analyzed the benchmarking and characterization protocol for photodetectors developed using various types of nanomaterials and pointed out that NEP is one of the most important sensitivity parameters for the figure of merit compared to the photoresponsivity and detectivity. The measurements of NEP as a function of modulation frequency of a photodetector could be used for a better comparison of the figure of merit of heterogeneous photodetectors. The photoresponsivity (R) data were plotted as a function of modulation frequency (f) and NEP for a wide variety of nanomaterial-based photodetectors. Photodetectors with vertical current transport were found to perform much better than photodetectors having lateral current transport as a function of modulation frequency with an exception of graphene-based photodetectors, which is associated with high carrier mobility. However, the measurements of photoresponsivity of these nanomaterials significantly vary depending upon the applied bias voltage and different intensity of laser power; therefore, an accurate comparison is rather convoluted. In the photoresponsivity *versus* NEP plot, the MoS₂-based photodetectors such as monolayer MoS₂ phototransistors,¹⁷¹ vertical MoS₂ p-n junction,³⁴⁶ MoS₂/PbS,³²² graphene/WS₂/MoS₂,³⁴⁷ *etc.*, are in par with PbS-based photodetectors.³⁹⁰ The plot of photoresponsivity *versus* NEP does not show an important correlation between these two parameters and the higher values of photoresponsivity does not inevitably lead to high sensitivity of photodetectors. High sensitivity of a photodetector can be achieved with a relatively low photoresponsivity by controlling the noise of photodetectors.

6. Strain-induced and self-powered MoS₂ photodetectors

Energy harvesting from piezoelectric materials such as ceramics, single crystals, organic polymers and their based hybrid composites has been investigated over many decades. The commonly known piezoelectric materials include lead zirconate titanate (PZT), barium titanate (BaTiO₃), lithium niobate (LiNbO₃), zinc oxide (ZnO), quartz, polyvinylidene difluoride (PVDF), poly(vinylidene fluoride-trifluoroethylene) (VPF-TrFE), nylons, liquid crystal polymers, biomaterials, *etc.*^{391–398} The piezoelectric, pyroelectric and ferroelectric materials offer capabilities of generating usable energy under applied mechanical strain, temperature, electric field, water, light, *etc.* therefore, have been integrated in energy harvesting devices for their applications in self-powering footwears, speakers and headphones, motors, batteries, sensors, actuators, catalysis, photovoltaics, optical memory devices, spintronics, robotics, triboluminescence, *etc.*^{399–402} In addition to silicon, Ge, SiC, ZnO, TiO₂, cement, polymers, diamond, and carbon nanotubes, the piezoresistive effects have been studied in 2D nanomaterials^{403–410} for applications in piezoresistive strain/stress sensors. Black phosphorus (BP) has been used with MoS₂ to develop BP/MoS₂ vdW heterostructure-based photodetectors with a type-II band alignment.⁴⁰⁴ The strain induced

piezoelectric effect was observed in BP and MoS₂ vdW heterostructures.

Piezoelectric and pyroelectric generators have been developed using different types of inorganic and organic materials for their applications in optoelectronic and photonic devices. Kim *et al.*⁴¹¹ reported piezoelectric effect in CVD-grown monolayer MoS₂ films and fabricated direction-dependent flexible piezoelectric nanogenerators where the atomic orientation in monolayer MoS₂ dictated the magnitude of the piezoelectric effect. The output power of nanogenerators (NG) was found to be two times higher for the armchair direction of MoS₂ compared with zigzag direction of MoS₂ under the same applied strain and the strain velocity. The armchair MoS₂ showed piezoelectric coefficient d_{11} of 3.78 pm V⁻¹ whereas the zigzag MoS₂ exhibited much lower d_{11} value of 1.38 pm V⁻¹. The armchair MoS₂ nanogenerator exhibited higher output voltage of 20 mV as well as the higher output current of 30 pA than that of 10 mV and 20 pA for the zigzag MoS₂ nanogenerator, confirming the role of atomic orientation in enhancing piezoelectric effect in 1L MoS₂ nanogenerators. Tsai *et al.*⁴¹² reported piezoresistive effect in flexible MoS₂ FETs which was associated with the strain-induced bandgap change as confirmed using optical reflection spectroscopy. The bandgap of the 3L MoS₂ was found to decrease by 0.06 eV (from 1.58 eV to 1.52 eV) under 0.2% applied strain. The effects of both piezoelectricity and piezoresistivity on photoresponse of atomic layered MoS₂ have been studied.

Strain-induced photodetection has been observed in MoS₂ atomic layers and MoS₂ based hybrid heterostructures. The monolayer MoS₂ based photodetector showed photoresponsivity of 2.3×10^4 A W⁻¹ under a compressive strain of -0.38% at low laser power intensity of 3.4 μW cm⁻² with 442 nm wavelength.²⁶⁴ Gas sensitivity has been reported in 1L MoS₂ *via* photogating and piezo-phototronic effects.⁴¹³ Monolayer MoS₂ based flexible homogenous photodiode showed 619% and 319% enhancement in photoresponsivity and detectivity after applying 0.51% tensile strain, where the photoresponsivity and detectivity reached maximum values of 1162 A W⁻¹ and 1.72×10^{12} Jones, respectively, compared with a strain-free photodiode.⁴¹⁴ The in-plane anisotropic photoresponse has been reported in monolayer MoS₂.⁴¹⁵ The crystal symmetry of monolayer MoS₂ was found to be reduced from D_{3h} to C_{2v} under applied uniaxial tensile strain. The absorption along the armchair direction of monolayer MoS₂ was found to be two times higher compared to zigzag direction under 4.5% strain, which also increased with increasing strain, whereas the absorption along zigzag direction of monolayer MoS₂ showed a slight change. The photoresponse of monolayer MoS₂ changed from isotropic to anisotropic at 520 nm light illumination. The photoresponse was not observed in monolayer MoS₂ devices until uniaxial tensile strain reached to a value of 3.5% and the anisotropic ratio higher than 2 was recorded under 4.5% uniaxial tensile strain at 520 nm and 830 nm wavelengths.

In another study, the photoresponsivity of flexible MoS₂/WSe₂ heterojunction-based photodiodes increased by 86% under a static strain of -0.62% when applied along the



armchair direction of MoS₂ due to the piezoelectric effect.⁴¹⁶ Similar strain-induced photoresponse has been measured in MoS₂ and ZnO p–n heterojunction photodiodes where photocurrent increased over four times under an applied pressure of 23 MPa.⁴¹⁷ The photoresponsivity of ML MoS₂/GaN heterojunction photodiodes was also enhanced by 3.5 times under 258 MPa applied pressure originating from strain-induced piezoelectric phenomenon.⁴¹⁸

The effect of piezoresistivity on performance of MoS₂ photodetectors has also been studied. Li *et al.*⁴¹⁹ reported piezoelectric and piezoresistive effects in a flexible CVD-grown monolayer MoS₂/polyethylene naphthalate (PEN) substrate-based photodetector. An enhancement of photoresponse such as light–dark current ratio from 29 to 1200.3 (41 times) at 1 V bias voltage and self-powered current from 25.2 to 213.6 pA (8.3 times) was observed in armchair direction monolayer MoS₂ after applying 0.8% strain due to the piezoelectric effect. However, the photoresponsivity decreased from 163.9 to 95.7 A W⁻¹ under 0.8% applied strain at 5 V bias voltage. The photocurrent increased from 0.37 to 2.35 μA and photoresponsivity from 114.3 to 590 A W⁻¹ in zigzag direction monolayer MoS₂ under 1.4% applied strain at 5 V due to the piezoresistive effect. The zigzag direction monolayer MoS₂ showed no piezoelectric effect. Kelvin probe force microscopy was used to analyze both piezoelectric and piezoresistive effects in monolayer MoS₂. Gant *et al.*⁴²⁰ reported strain-induced increase in both photocurrent and photoresponsivity of flexible monolayer MoS₂ photodetectors. The monolayer MoS₂ based photodetector showed significant increase in photocurrent at 740 nm light illumination under incident power density of 5 mW cm⁻² with 5 V applied bias voltage when tensile biaxial strain was increased from 0.16% to 0.48%. In biaxially strained polycarbonate substrate-based flexible MoS₂ photodetectors, the photoresponsivity increased 100 and 1000 times by increasing applied strains from -0.8% to 0.48% and from -1.44% to 0.48%, respectively.

2D material-based self-powered heterojunction photodetectors are also gaining much attention because no external power is required for the photodetection.⁴²¹ The self-powered p-MoTe₂/n-MoS₂ vdWH-based photodetector having ITO electrodes showed photoresponsivity of 146 mA W⁻¹ and fast photoresponse of ~172 μs from 450 nm to 980 nm.⁴²² The photodetector also exhibited bidirectional photocurrent response associated with type-II band aligned heterostructure and ITO electrodes. Yang *et al.*⁴²³ used narrow bandgap layered semiconductor germanium selenide (GeSe) for developing hybrid photodetectors where photoconductive GeSe/graphene photodetectors were compared with photovoltaic GeSe/MoS₂ photodetectors. The photodetector based GeSe/graphene vdWHs showed higher photoresponsivity up to 10⁴ A W⁻¹ under laser light intensity of 0.17 mW cm⁻² at 532 nm whereas the GeSe/MoS₂ heterostructure based photodetector exhibited low photoresponsivity of 0.1 A W⁻¹ but a faster recovery time of 5 ms compared with 2 s for GeSe/graphene heterostructure. GeSe/MoS₂ photodetector retained constant photoresponse under applied reverse bias because of the reduced carrier conduction within depletion region. Xin *et al.*⁴²⁴ fabricated

polarization-sensitive self-powered photodetectors using type-II band aligned GeSe/MoS₂ vdWHs to increase photoresponse spectrum and support efficient separation as well as transportation of the photoexcited charge carriers. GeSe/MoS₂ p–n heterojunctions were developed using mechanically exfoliated GeSe and MoS₂ nanoflakes on p-doped Si substrate having a SiO₂ film of 300 nm thickness. Fig. 27(a–c) shows a type-II band aligned GeSe/MoS₂ p–n heterojunction photodetector with Ti/Au electrodes, and time-dependent photoresponse at different illumination wavelengths (UV–NIR region), and photoresponsivity and detectivity of the self-powered GeSe/MoS₂ heterojunction photodetectors as a function of wavelength from 380 to 1064 nm at V_{ds} of 0 V (zero bias). The self-powered GeSe/MoS₂ photodetector achieved I_{on}/I_{off} current ratio of 10⁴ at zero bias, photoresponsivity of 105 mA W⁻¹ and EQE of 24.2%. GeSe/MoS₂ vdWH-based photodetectors showed photoresponse from 380 to 1064 nm and showed maximum photoresponsivity of 590 mA W⁻¹ at 532 nm wavelength. The photoresponsivity of the self-powered GeSe/MoS₂ p–n heterojunction photodetector varied from 6.1 mA W⁻¹ to 150 mA W⁻¹ while the detectivity remained at 10¹¹ Jones in the 380 nm to 1064 nm (UV–NIR) range. Photoresponse was also measured as a function of polarization angle from 0° to 360° where the current on/off ratios were found to be 7.23 × 10³, 3.65 × 10⁴, and 2.99 × 10³ at 30°, 90° and 120° polarization angles under 532 nm light illumination with V_{ds} of 0 V, respectively. The highest photocurrent of -3.08 × 10⁻¹⁰ A was recorded at 90° and 270° polarization angles corresponding to the armchair direction of GeSe which was also a parallel direction of the polarization, whereas the lowest photocurrent of -1.04 × 10⁻¹⁰ A was measured at 0° and 180° polarization angles which corresponded to the zigzag direction of GeSe. The polarization-sensitive photodetection in the self-powered GeSe/MoS₂ photodetector was observed due to the absorption and anisotropic electronic transport associated with GeSe nanoflakes.

The perovskites have been used with MoS₂ to develop organic/inorganic hybrid-based photodiode–solar cell bifunctional devices. Shin *et al.*⁴²⁵ used MoS₂ as a hole transport layer (HTL) and graphene (GR) co-doped with AuNPs and (trifluoromethanesulfonyl)-amide (co-GR) as transparent conductive electrodes (TCE) to develop flexible p–i–n-type perovskite-based photodiode/solar cell bifunctional devices (PPSBs). Self-powered p–i–n-type PPSB devices were fabricated on both rigid glass and flexible PET substrates. Fig. 27(d–f) shows a diagram of p–i–n-type PPSB device consisting of Al/BCP/PCBM/MAPbI₃/MoS₂/co-GR; energy levels diagram, wavelength-dependent photoresponsivity and detectivity at 0 V bias (self-powered mode), current I_{on}/I_{off} switching curve at 500 nm and as a function of bending cycles. In the solar cell (photovoltaic) mode under 1 sun illumination (100 mW cm⁻²), the PPSB device on a glass substrate showed open-circuit voltage (V_{oc}) of 0.88 V, the short circuit current density (J_{sc}) of 19.97 mA cm⁻², a fill factor (FF) of 74.46%, and power conversion efficiency (PCE) of 13.09%. PPSB device also operated without any external power (0 V bias), which is a self-powered mode. The current of the PPSB photodiode was found to increase 10⁶ times under illumination in the self-powered mode



and the photocurrent rise/fall times ranged 0.7–0.9 s and 0.4–0.6 s, respectively. The PPSB photodiode (0 V bias) on a glass substrate showed current $I_{\text{on}}/I_{\text{off}}$ ratio of 1310, 9580, 136 110, and 8; and photoresponsivities of 50, 309, 410, and 12 mA W^{-1} at 300, 500, 700, and 800 nm, respectively. The noise equivalent power (NEP), detectivity, and linear dynamic range (LDR) were calculated as $3.95 \times 10^{-12} \text{ W Hz}^{-1/2}$, 7.99×10^{10} Jones, and 117 dB, respectively. The flexible PPSB-based solar cell showed PCE of 11.91%. The flexible PPSBs also exhibited the similar photoresponse at 500 nm as glass substrate ones. The photoresponsivity and detectivity of the flexible PPSB device were measured between 350 nm to 750 nm where optimum values

reached at 0.39 A W^{-1} and 3.41×10^{10} Jones, respectively. The same research group⁴²⁶ also developed organic/inorganic perovskites/ MoS_2 hybrid-based photodiode–solar cell nano-systems (PPSNs) using BL MoS_2 as the electron-transport layer (ETL) and triethylenetetramine-doped graphene (TETA-GR) as the transparent conductive electrode (TCE). The TETA-GR/ MoS_2 /MAPbI₃/PTAA/Au glass substrate-based rigid and PET substrate-based flexible PPSNs showed photoresponsivity of 0.42 and 0.40 A W^{-1} , NEP of 37.2 and $80.1 \text{ pW Hz}^{-1/2}$, detectivity of 1.1×10^{10} and 5.0×10^9 Jones in the photodiode mode under zero bias (self-powered), respectively. The PPSNs exhibited the PCE value of 14.27% for rigid device and 12.12% for flexible

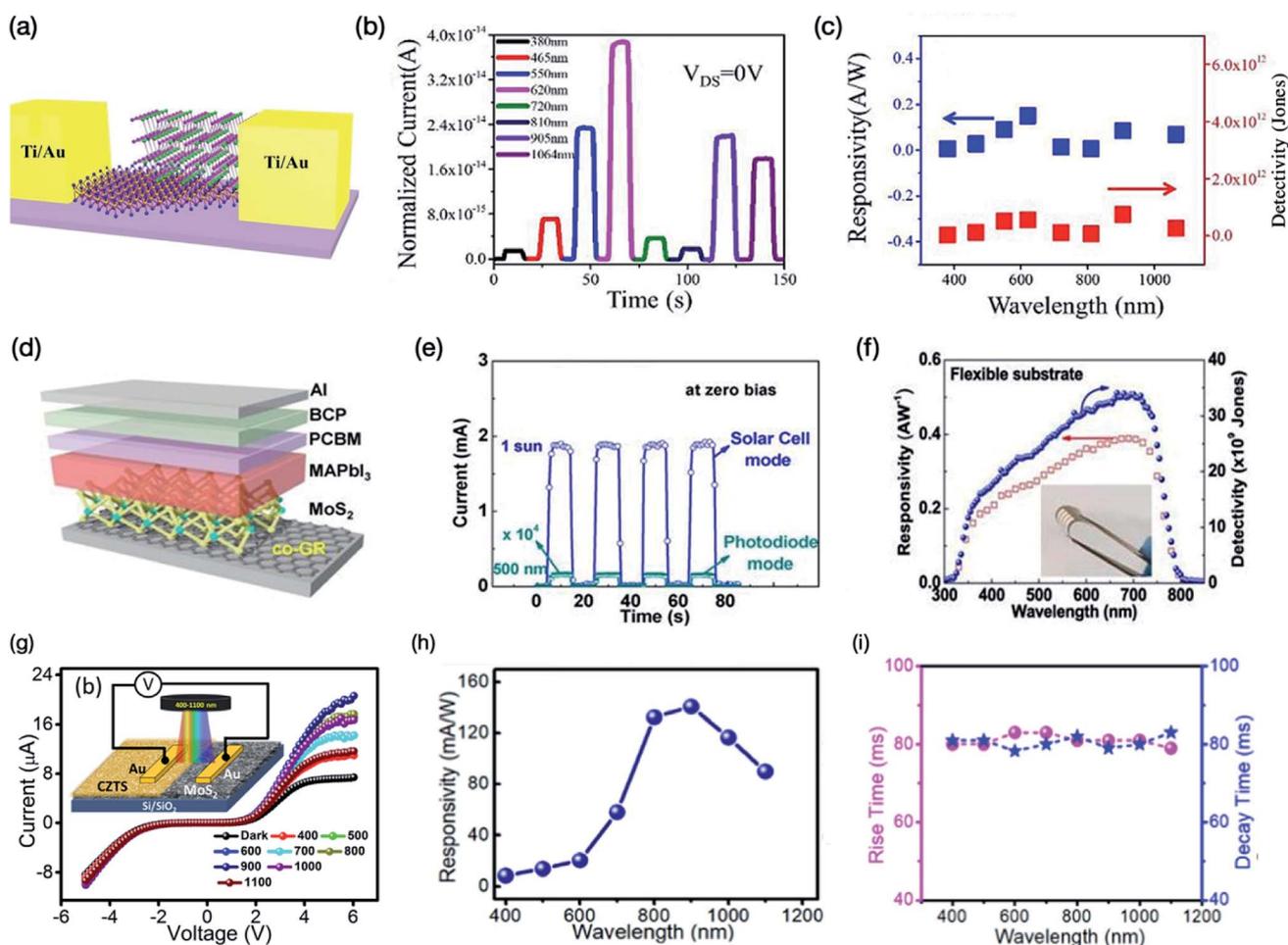


Fig. 27 (a) Schematic illustration of a type-II band aligned GeSe/ MoS_2 p–n heterojunction photodetector having Ti/Au electrodes, separation of the photoexcited electrons–holes carriers and energy band diagram under 532 nm light illumination at V_{ds} of 0 V (zero bias). (b) Time-dependent photoresponse of the heterojunction photodetector under different illumination wavelengths at $V_{\text{ds}} = 0 \text{ V}$. (c) Photoresponsivity and detectivity of the GeSe/ MoS_2 heterojunction photodetectors as a function of wavelength. Reprinted with permission from ref. 424, copyright © American Chemical Society. (d) Schematic illustration of a self-powered MoS_2 photodetector based on p–i–n-type perovskite photodiode/solar cell bifunctional (PPSB) device consisting of Al/BCP/PCBM/MAPbI₃/ MoS_2 /co-GR; where BCP = bathocuproine, PCBM = phenyl-C₆₁-butyric acid methyl ester, MAPbI₃ = methylammonium lead tri-iodide perovskite, co-GR = graphene electrode co-doped with AuNPs and (tri-fluoromethanesulfonyl)-amide. MoS_2 based flexible PPSB photodetector was fabricated using a PET substrate. (e) Current $I_{\text{on}}/I_{\text{off}}$ switching behavior of flexible PPSB photodetectors at 500 nm at 0 V bias. (f) Wavelength-dependent photoresponsivity and detectivity of flexible photodetector recorded at 0 V bias, therefore in a self-powered mode. Inset is an optical image of a flexible self-powered PPSB photodetector. Reprinted with permission from ref. 425, copyright © Elsevier; (g) schematic illustration of CZTS/ MoS_2 p–n heterojunction-based photodetector and current–voltage (I – V) curves in the dark and at different laser wavelengths (400 nm to 1100 nm) under illumination. (h) Curve of photoresponsivity as a function of wavelength and (i) calculated rise/decay times of the self-powered CZTS/ MoS_2 photodetector from 400 nm to 1100 nm wavelengths. Reprinted with permission from ref. 428, copyright © Elsevier.



PPSN device in the photovoltaic mode. PTAA refers to poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] which is used as a hole transport material (HTM) in solar cells.

He *et al.*⁴²⁷ developed broadband self-powered photodetectors using vertically-stacked multilayer GaSe/MoS₂ heterostructures where the thickness of GaSe and MoS₂ films was 6.3 nm (≈ 7 L) and 6 nm (≈ 8 L), respectively. Wavelength-dependent photoresponsivity, EQE, and detectivity of self-powered GaSe/MoS₂ hybrid heterostructure-based photodetectors were measured as a function of wavelength without bias voltage ($V = 0$). The performance of photodetectors having ohmic-contact ITO electrodes were compared with Schottky-contact Ni/Au electrodes. The GaSe/MoS₂ heterostructure-based self-powered photodetectors covered visible to NIR spectral range. Self-powered photodetectors with ITO/GaSe/MoS₂/ITO configuration showed a high rectification ratio of 1.5×10^4 at $V_{ds} = \pm 1$ V, EQE value of 160%, photoresponsivity of 0.67 A W^{-1} , and detectivity of 2.3×10^{11} Jones at 520 nm and four times faster response time than that of (Ni/Au)/GaSe/MoS₂/(Ni/Au) photodetectors indicating the role of electrode-contact in influencing the performance of GaSe/MoS₂ hybrid photodetectors. The photoresponsivity of ITO/GaSe/MoS₂/ITO photodetector was found to be very weak at 808 nm wavelength compared with visible light wavelengths.

Copper zinc tin sulfide (Cu₂ZnSnS₄) thin films have been used as absorbing layers in photovoltaic devices. Agrawal *et al.*⁴²⁸ reported type-II band-alignment by forming a hybrid heterostructure between p-type 3D Cu₂ZnSnS₄ (CZTS) and n-type 2D MoS₂ which showed significant PL quenching at the heterojunction interface. Fig. 27(g–i) shows a schematic illustration of CZTS/MoS₂ p–n heterojunction photodetector and the current–voltage (I – V) curves in the dark and at different laser wavelengths (400 nm to 1100 nm) under illumination, photoresponsivity and calculated rise/decay times of the self-powered CZTS/MoS₂ photodetector from 400 nm to 1100 nm wavelengths. The CZTS/MoS₂ p–n heterojunction-based photodetectors exhibited photovoltaic properties, with a photoresponsivity of 141 mA W^{-1} , the current I_{on}/I_{off} ratio of 112, and fast rise/decay times of 81/79 ms at 900 nm. The CZTS/MoS₂ p–n heterojunction photodetector also showed an increase in photoresponsivity to 79 A W^{-1} under an applied bias voltage of 6 V and high stability up to 1500 h (62.5 days) of operation. Agrawal *et al.*⁴²⁹ also developed self-powered photodetector using p-type Cu₂ZnSnS₄ (CZTS) active electrodes with n-type vertical MoS₂ flakes. The CZTS/MoS₂ p–n heterojunction photodetector showed photoresponsivity of 49.31 A W^{-1} and EQE value of $7.6 \times 10^3\%$ with enhanced photoresponse in visible and NIR region compared with metal electrodes. The heterojunction photodetector showed detectivity of 3.4×10^{13} Jones in NIR and 2.4×10^{13} Jones in visible wavelength region. The photoresponsivity of photodetector with CZTS electrodes was increased by 11 times compared with gold electrodes at 1100 nm (NIR region). Furthermore, the photodetector with CZTS electrodes also exhibited long term stability up to 4000 h (166 days).

Self-powered photodetectors were developed using MoS₂ thin film and bulk GaAs.⁴³⁰ The MoS₂/GaAs heterojunction-based photodetector operated from DUV to NIR spectral region and exhibited photoresponsivity of 35.2 mA W^{-1} ,

detectivity of 1.96×10^{13} Jones and rise and fall times of 3.4 μs /15.6 μs at zero bias. These MoS₂/GaAs heterojunction photodetectors also demonstrated polarization sensitivity to an optical signal with 4.8 ratio between the peak-to-valley. The 0D/3D MoS₂ QDs/GaAs heterojunction photodetectors were fabricated using n-type GaAs substrates with varied doping concentrations which resulted in n–n heterojunctions between n-type MoS₂ QDs and bulk n-type GaAs.⁴³¹ The MoS₂/GaAs heterojunction photodetectors operated between 400–950 nm and showed highest photoresponsivity of 400 mA W^{-1} and detectivity of 4×10^{12} Jones at 500 nm at zero bias voltage, which is a self-powered photodetection. Shi *et al.*⁴³² integrated 0D InP@ZnS core–shell QDs with 2D bilayer MoS₂ to develop self-powered hybrid phototransistors by using interdigitated Pt electrodes which acted as light collectors as well as plasmonic resonators. 0D/2D InP@ZnS/MoS₂ hybrid photodetectors showed photoresponsivity of 1374 A W^{-1} and self-powered properties with a fast photospeed of 21.5 μs . The photoresponsivity of MoS₂ was measured as 222.5 A W^{-1} at 532 nm under illuminating power intensity of 34 mW cm^{-2} at gate voltage of 40 V, which decreased to 81 A W^{-1} as the laser power intensity was increased to 170 mW cm^{-2} due to the temperature induced increment in recombination and scattering of carriers. The photocurrent of InP@ZnS/MoS₂ hybrid phototransistors was enhanced compared to MoS₂ phototransistors as a function wavelength at 447, 532 and 671 nm. The stability of hybrid photodetectors was also examined at 8%, 17%, 25%, 31%, and 42% of humidity at 532 nm illuminating laser light applying 200 Hz frequency without any applied bias voltage. InP@ZnS/MoS₂ hybrid phototransistor devices exhibited reproducibility over 4000 repeated cycles under 2000 Hz frequency and improved environmental stability over MoS₂ phototransistors over a period of three months, indicating the protecting role of InP@ZnS QDs for the MoS₂ layer in order to prevent the degradation of photodetectors under ambient environmental conditions.

The self-powered photodetectors were developed using MoS₂/AlN/Si hybrid heterostructure where MoS₂ thin films were deposited on AlN/Si(111).⁴³³ The hybrid photodetector exhibited photoresponse from 300 nm to 1100 nm at 0 V, having the photoresponsivity of 9.93 A W^{-1} at 900 nm and rise/decay times of 12.5 μs /14.9 μs . At zero bias, the photoresponsivity of MoS₂/AlN/Si hybrid (9.93 A W^{-1}) was found to be over five times higher than that of MoS₂/Si photodetector (1.88 A W^{-1}). The oxygen defects in AlN supported the transportation of photo-generated charge carriers in the hybrid photodetector. Self-powered photodetectors using a vertical MoO₃/MoS₂ heterojunction⁴³⁴ have been fabricated which showed photoresponsivity of 670 mA W^{-1} and detectivity of 4.77×10^{10} Jones under 0 V bias and current I_{on}/I_{off} current ratio of 10^3 . MoS_{2(1-x)Se_{2x}} nanostructured alloys have been used for developing photodetectors by atomic-level tailoring. Hou *et al.*³⁸⁵ used hydrothermal method to prepare MoS_{2(1-x)Se_{2x}} ($x = 0, 0.5, 1$) nanostructures. The MoS₂Se alloy-based photodetectors exhibited photocurrent of 12.6 μA , photoresponsivity of 1.753 mA W^{-1} and rise/fall times of 4.7 s at 660 nm under laser illumination of 1.75 mW mm^{-2} . The photocurrents of MoS₂Se alloy



based photodetectors were considerably increased compared with pure MoS₂ and MoSe₂ based photodetectors. The photoresponsivity of CVD-grown Au/MoS_{1.15}Se_{0.85}/Au based photodetectors was found to be much higher, 2.06×10^3 mA W⁻¹ at 532 nm with rise/fall times of 18 s/35 s.³⁸⁶ In another study by Xu *et al.*,⁴³⁵ the bandgap of MoS_{2(1-x)}Se_{2x} was tailored between 1.83 eV to 1.73 eV by adjusting the Se composition and thickness within a single domain. The spatially bandgap-graded MoS_{2(1-x)}Se_{2x} based phototransistors showed photoresponsivity of 191.5 A W⁻¹, detectivity of 10¹² Jones, photoconductive gain of 10⁶ to 10⁷ and response speed of 51 ms under -0.5 V bias voltage. The phototransistor showed photoresponsivity values of 23.2, 191.5, and 26.2 A W⁻¹ at 405, 650, and 808 nm laser illuminations under -0.5 V bias voltage, respectively. The homojunction phototransistors yielded photoresponsivity of 311 mA W⁻¹, detectivity of 10¹¹ Jones, and the current I_{on}/I_{off} ratio of 10⁴ at 405 nm wavelength under laser power intensity of 1.73 mW mm⁻² for the at zero bias (self-powered operation) MoS_{2(1-x)}Se_{2x} phototransistors. Han *et al.*⁴³⁶ developed a self-driven photodetection device using an MoS₂ nanosheet-based photodetector, a vertical contact-separate mode triboelectric nanogenerator (CS-TENG) as the power source, and LEDs as the alarm system. The mechanism is

based on the impedance matching effect between TENG specific output and the performance of MoS₂ photodetector which is sensitive to visible light. The voltage of TENG is regulated with a Zener diode, the enabling self-powered MoS₂ photodetector to function properly. The MoS₂ photodetector exhibited high current I_{on}/I_{off} ratio of 1.02×10^4 and the rise/fall times of 0.32/0.36 s at 405 nm wavelength. Zhao *et al.*⁴³⁷ developed a surface ionic gate (SIG) transistor using MoS₂ monolayer where gas ions are adsorbed by MoS₂, which functions as a gate to control the carrier concentration and transport process. The modulation can be adjusted by the TENG operation cycles and the current I_{on}/I_{off} ratio of 10⁴ was obtained. In this SIG-based MoS₂ photodetector, the photocurrent recovery time is 74 ms, much faster than devices without SIG modulation. The photocurrent of the SIG-based MoS₂ photodetector was found to increase linearly as a function of time for a period of 120 s, useful for developing a photodetector with luminous flux.

Table 2 summarizes the data collected on the photoresponsivity, detectivity and response/recovery time (τ_{rise}/τ_{decay}) of self-powered MoS₂ based photodetectors along with their measurement wavelengths and incident laser powers. The strain-induced 2D nanomaterial-based self-powered photodetectors have been reported where piezoelectric effect also plays

Table 2 The performance of self-powered MoS₂ photodetectors in terms of their photoresponsivity, detectivity and response/recovery time (τ_{rise}/τ_{decay}) along with their measurement wavelength and incident laser power (Jones = cm Hz^{1/2} W⁻¹)^a

MoS ₂ Photodetector	Conditions (wavelength, incident power)	Photoresponsivity (A W ⁻¹)	Detectivity (Jones)	Response time (τ_{rise}/τ_{decay})	Ref.
RGO-MoS ₂ /pyramid Si	(λ = 808 nm, 100 nW)	21.8	3.8×10^{15}	2.8 μ s/46.6 μ s	178
p-WSe ₂ /n-MoS ₂	(λ = 514 nm, 5 μ W)	0.04	—	100 μ s	123
p-WSe ₂ /n-MoS ₂	(λ = 532 nm, 7 μ W)	0.01	—	1 μ s	165
p-GaTe/n-MoS ₂	(λ = 633 nm, 100 mW cm ⁻²)	1.365	—	10 ms	167
1L MoS ₂ /P-Si	(λ = 532 nm, 0.95 mW)	117	1×10^9	74 ms/115 ms	314
MoS ₂ /Si (PVD)	(λ = 808 nm)	0.21	10 ¹³	3 μ s/40 μ s	162
ITO/GaSe/MoS ₂ /ITO	(λ = 520 nm)	0.67	2.3×10^{11}	155 μ s/215 μ s	427
(Ni/Au)/GaSe/MoS ₂ /(Ni/Au)	(λ = 520 nm)	0.037	1.9×10^{10}	620 μ s/740 μ s	427
p-MoTe ₂ /n-MoS ₂	(λ = 800 nm)	0.038	—	—	205e
p-MoS ₂ /n-WS ₂	(λ = 532 nm, 28.64 mW cm ⁻²)	0.043	4.36×10^{13}	4 ms	343
p-GaN nanowires/n-MoS ₂ (strain)	(λ = 550 nm)	734.5	—	5 ms	258c
p-MoS ₂ /n-GaN	(λ = 265 nm)	0.187	2.34×10^{13}	46.4 μ s/114.1 μ s	316
p-GeSe/n-MoS ₂	(λ = 532 nm)	0.105	1.03×10^{10}	110 ms/750 ms	424
MoS ₂ /CH ₃ NH ₃ PbI ₃		0.06	—	2149 ms/899 ms	327
p-Cu ₂ ZnSnS ₄ /n-MoS ₂	(λ = 900 nm)	0.141	—	81 ms/79 ms	—
MoS ₂ /GaAs	(λ = 780 nm, 73 nW cm ⁻²)	0.352	1.96×10^{13}	3.4 μ s/15.6 μ s	430
n-MoS ₂ /n-GaAs	(λ = 650 nm, 20 mW cm ⁻²)	0.43×10^{-3}	2.28×10^{11}	1.87 μ s/3.53 μ s	438
MoS ₂ / β -Ga ₂ O ₃	(λ = 245 nm, 20.1 μ W cm ⁻²)	2.05×10^{-3}	1.21×10^{11}	—	439
Pd-MoS ₂ /Si	(λ = 950 nm, 1.0 mW cm ⁻²)	0.654	1.0×10^{14}	2.1 μ s/173.8 μ s	440
Pd/Al ₂ O ₃ /MoS ₂	(λ = 532 nm, 0.95 mW)	0.308	—	972 ms/878 ms	441
Pd/HfO ₂ /MoS ₂	(λ = 532 nm, 0.95 mW)	0.538	—	969 ms/875 ms	441
Graphene/InSe/MoS ₂	(λ = 532 nm, 1.26 mW cm ⁻²)	0.11	1.08×10^{10}	1 ms	442
InSe/Se	(λ = 460 nm, 1 mW cm ⁻²)	0.032	1.7×10^{11}	30 ms/37 ms	352c
p-Cu ₉ S ₅ /n-MoS ₂	(λ = 532 nm, 9.36 mW cm ⁻²)	76	1.6×10^{12}	8 ms/2 ms	443
p-Cu ₂ ZnSnS ₄ /n-MoS ₂	(λ = 900 nm, 1.02 μ W cm ⁻²)	0.141	—	81 ms/79 ms	428
Au/p-Cu ₂ ZnSnS ₄ /n-MoS ₂ /Au	(λ = 800 nm, 0.43 μ W cm ⁻²)	49.31	2.4×10^{13}	0.5 s/0.8 s	429
TETA-GR/MoS ₂ /MAPbI ₃ /PTAA	(λ = 500 nm)	0.42	1.1×10^{10}	1.63 μ s/0.98 μ s	426
InP@ZnS/MoS ₂	(λ = 532 nm, 34 mW cm ⁻²)	1.3×10^3	—	21.5 μ s/133.3 μ s	432
MoS _{2(1-x)} Se _{2x}	(405 nm, 1.73 mW mm ⁻²)	0.311	10 ¹¹	—	435

^a (TETA-GR): triethylenetetramine-doped graphene, PTAA: poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine].



an important role. In addition to various MoS₂ hybrid heterostructures, MoS₂ vdWHs with other 2D atomic layered nanomaterials have also been studied for developing self-powered photodetectors. For example, MoS₂,¹⁷³ MoS₂-PDPP3T,¹⁸⁹ MoS₂/MoTe₂,²⁰⁵ MoS₂/P-Si,³¹⁴ MoS₂/GaN,³¹⁶ MoS₂/perovskite,³²⁷ and MoS₂/WS₂,³⁴³ based self-powered photodetectors show high photoresponsivity, detectivity, fast photoresponse time and high current $I_{\text{on}}/I_{\text{off}}$ ratio. The performance of self-powered MoS₂ photodetectors can be enhanced due to the piezoelectric effect.^{221,258,272} MoS₂ based self-powered broadband photodetectors have been reported for n-MoS₂/n-GaAs,⁴³⁸ MoS₂/β-Ga₂O₃,⁴³⁹ Pd-MoS₂/Si,⁴⁴⁰ Pd/Al₂O₃/MoS₂,⁴⁴¹ Pd/HfO₂/MoS₂,⁴⁴¹ graphene/InSe/MoS₂,⁴⁴² p-Cu₉S₅/n-MoS₂,⁴⁴³ and MoS₂(1-x)-Se_{2x}.^{385,386,435} MoS₂ based self-powered photodetectors exhibit photoresponsivity from mA W⁻¹ to A W⁻¹, detectivity from 10⁹ to 10¹⁵ Jones and response time from microseconds (μs) to seconds (s), depending on the measurement wavelengths and incident power densities which could be used for healthcare applications.

7. Stability of MoS₂ photodetectors

Photodetectors are generally used under harsh working environmental conditions for surveillance including imaging and optical communications; therefore, long-term stability of photodetectors are of immense importance for commercial applications. The environmental stability for extended period of time has been observed in some MoS₂ hybrid heterostructure-based photodetectors.^{162,187,192} A few examples are discussed here. The detectivity of 10¹³ Jones and a fast response speed of 3 μs have been reported for the self-driven visible-NIR MoS₂/Si heterojunction-based photodetector.¹⁶² These MoS₂/Si heterojunction photodetectors also showed negligible degradation of photovoltage in air for a period of one month under light illumination at 808 nm. The RGO-MoS₂/pyramid Si heterojunction-based photodetectors¹⁷⁸ showed environmental stability over three months of storing in the air under 808 nm and 1550 nm light illuminations due to the synergistic effect of RGO, MoS₂ and the graphene electrodes. Kang *et al.*¹⁸⁰ measured the stability of APTES-doped MoS₂ photodetectors, which were (i) as-doped, (ii) exposed to the air for 36 h and (iii) for 120 h. The on-current of APTES-doped MoS₂ photodetectors was found to decrease by 15% from 273 μA μm⁻¹ to 231 μA μm⁻¹ after air exposing for 36 h and 37% decrease from 273 μA μm⁻¹ to 171 μA μm⁻¹ after air-exposing for 120 h. The environmental stability of various MoS₂ based photodetectors are shown in Fig. 28(a-e). MoS₂/PDPP3T heterojunction-based self-powered phototransistor showed photodetection from UV (365 nm) to NIR (859 nm) region with photoresponsivity of 276, 445, and 269 mA W⁻¹ and detectivity of 2.59 × 10⁸, 3.14 × 10⁸, and 2.53 × 10⁸ Jones at 365, 660, and 850 nm wavelengths, respectively.¹⁸⁹ Flexible MoS₂/PDPP3T photodetectors fabricated on the PET substrate were found to be mechanically stable up to 10 000 bending cycles with a little decrease in photocurrent under repeated bending test conducted at a bending radius of 9 mm with 660 nm. Furthermore, MoS₂/PDPP3T heterojunction photodetectors also exhibited stability in air atmosphere up to 35 days in

dark and light illumination without any encapsulation of devices. Gold chloride hydrate *in situ* doped MoS₂ based photodetectors retained 94% of the initial photocurrent value after a period of nine months, evidencing good environmental stability.³³⁷ The stability in air for one month has been measured for the self-driven an MoS₂/GaAs heterojunction photodetectors when encapsulated with poly(methyl methacrylate) to avoid oxidation.³¹⁸ High photoresponsivity and detectivity have been measured in MoS₂/P(VDF-TrFE) hybrid photodetector where [P(VDF-TrFE)] ferroelectric copolymer was used as a gate.¹³⁷ This MoS₂/P(VDF-TrFE) hybrid photodetector also displayed mechanical stability over 90 000 repeated cycles at incident light power of 100 nW at 635 nm light illumination. The mechanical stability of various flexible MoS₂-based hybrid photodetectors including MoS₂/Kapton and MoS₂/PET,²¹⁷ MoS₂/PI,²²⁰ MoS₂/graphene,^{233,234} MoS₂-MoS₂/CNT,²⁵⁶ MoS₂/WS₂,¹⁶⁴ MoS₂/ZnO,²⁷⁷ stacked PEDOT:PSS/PVP/PEDOT:PSS/MoS₂/PEN,²¹⁹ MoS₂/perovskite,⁷⁶ MoS₂/Ag nanocubes,¹⁸⁶ and surface-functionalized monolayer MoS₂²⁹³ have been discussed in details earlier. MoS₂-based photodetectors display both environmental and mechanical stability for a long period of time against oxidative atmosphere to mechanical testing. Cu₂ZnSnS₄/MoS₂ p-n heterojunction-based self-powered photodetectors showed stability up to 62.5 days of operation.⁴²⁸ When Cu₂ZnSnS₄ was used on an active electrode, the self-powered Cu₂ZnSnS₄/MoS₂ photodetector exhibited even longer stability up to 166 days.⁴²⁹ The photoresponse of a rigid p-i-n-type perovskite-based photodiode/solar cell bifunctional device (PPSB) device decreased by 38% after storing for 30 days under 30% humidity at 25 °C compared to 60% degradation of PEDOT:PSS HTL device after 10 days. The flexible PPSB devices showed 43% degradation of their initial photocurrent values after 3000 bending cycles at 4 mm a curvature radius. This demonstrated that use of MoS₂ thin film as a HTL in perovskites-based self-powered PPBS devices and for improving their stability.⁴²⁵ The organic/inorganic perovskites/MoS₂ hybrid-based photodiode-solar cell nanosystem (PPSN) rigid device retained over 78% of the initial photoresponsivity after 30 days of storing period in ambient conditions and flexible device retained 80% of the initial photoresponsivity at 600 nm wavelength up to 1000 bending cycles demonstrating strong mechanical stability due to the synergistic effect between TETA-GR electrodes and BL-MoS₂ ETL.⁴²⁶

8. Applications of MoS₂ photodetectors

The photoresponse of MoS₂ photodetectors was also found to increase remarkably by the piezophototronic effect.^{226,263,265} The photoresponse of MoS₂ photodetectors significantly increases after different types of doping.^{183,188,194,325} The photoresponsivity, detectivity and photoresponse of atomic layered MoS₂-based hybrid heterostructured photoconductors range over several orders of magnitude, from mA W⁻¹ to 10¹⁰ A W⁻¹,^{122,138,187,199,280} from 10⁷ to 10¹⁵ Jones^{178,301-303} and from nano/microseconds to seconds,^{163,170,171,304,305} over an ultra-broadband spectral range



varying from DUV wavelength³¹⁶ to LWIR,^{169,340} respectively. As evident from these summarized data that MoS₂-based photoconductors show outstanding performance with highest photoresponsivity of 10¹⁰ A W⁻¹, the capability of detecting a weak signal up to 10¹⁵ Jones and a very fast photoresponse speed of nanosecond over a broad spectral bandwidth. Marin *et al.*⁴⁴⁴ reported the integration of an MoS₂ photodetector with a silicon nitride (Si₃N₄) photonic circuit where near-unity lateral absorption was measured that gave rise to a higher photoresponsivity. The speed of the MoS₂ photodetector was found to increase when MoS₂ channel was combined with h-BN substrate. Furthermore, low power operation in MoS₂

photodetector was also achieved by using graphene gates. Kim *et al.*⁴⁴⁵ proposed the application of the Ge-gated MoS₂ photoconductor for developing a multilevel optic-neural synaptic device. Fig. 29 shows various applications of MoS₂ based ultra-broadband photodetectors. The MoS₂ based photodetectors can be used in a wide range of industrial products operating from DUV to LWIR spectral range that include optical communications, environmental monitoring, chemical and medical sensors, biomedical imaging, remote sensing, lasers, satellites, military, security systems, night vision, smartphones, laptops, computers, Internet-of-Things (IoT), LEDs, data storage, optical microscopy and other next-generation optoelectronic-based

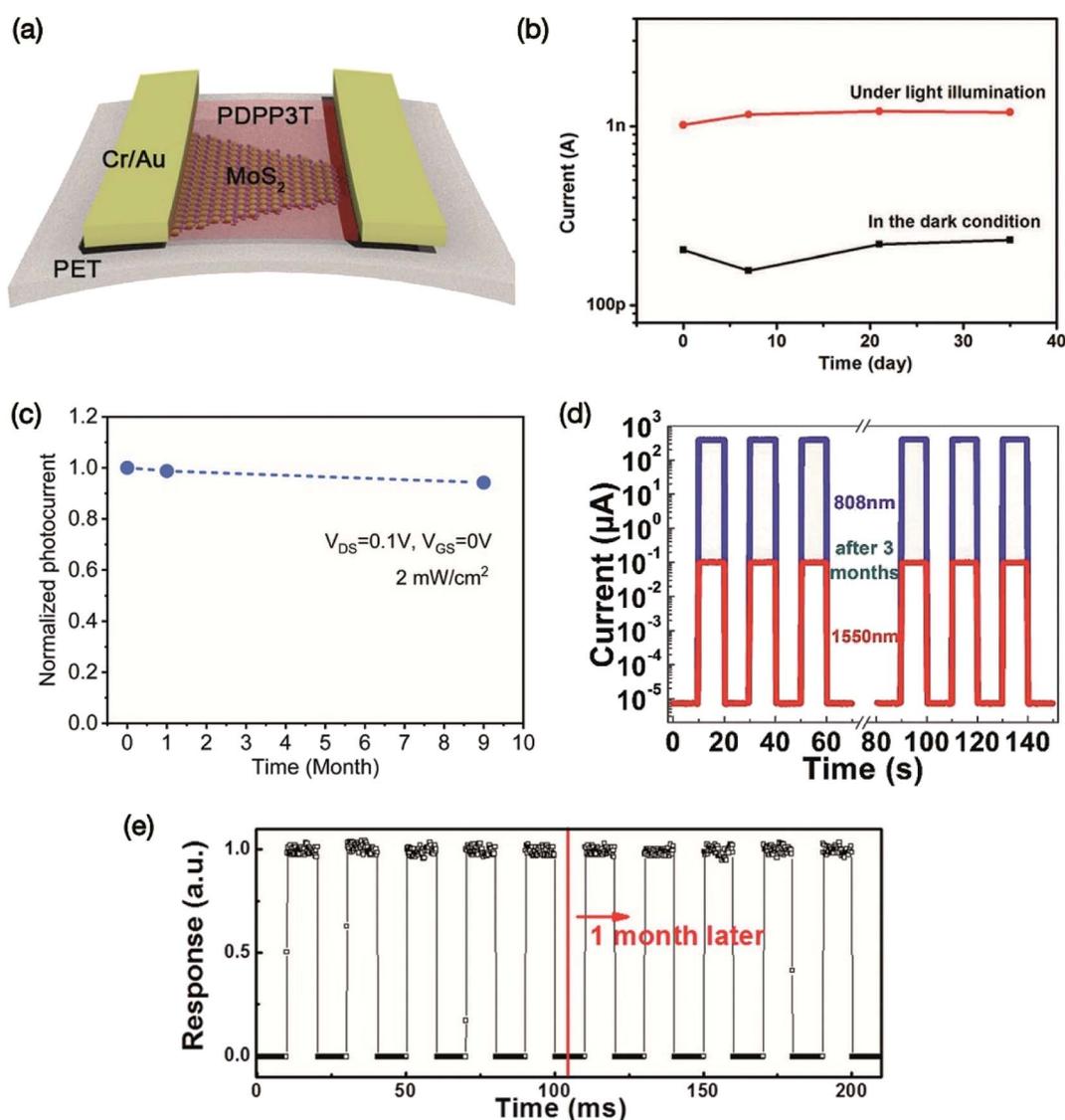


Fig. 28 (a) The schematic of the flexible MoS₂/PDPP3T photodetector on the PET substrate. (b) Current versus time curves in the dark and under 660 nm light illumination when photodetectors were stored in air up to 35 days. Reprinted with permission from ref. 189, copyright © Wiley. (c) The stability of photocurrent of the gold chloride hydrate *in situ* doped MoS₂ photodetector measured over several months where 94% of the initial photocurrent value was retained after nine months. Reprinted with permission from ref. 337, copyright © American Chemical Society. (d) Photoresponse of RGO–MoS₂/pyramid Si heterojunction photodetector measured at 808 and 1550 nm light illumination before and after storing devices in the air for three months. Reprinted with permission from ref. 178, copyright © Wiley. (e) Normalized response of the MoS₂/GaAs heterojunction-based self-driven photodetector measured after storage in air atmosphere for one month. Reprinted with permission from ref. 318, copyright © Elsevier.



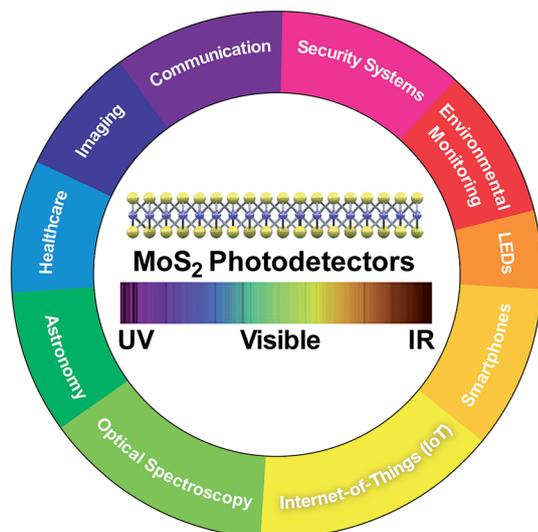


Fig. 29 Applications of atomic layered MoS₂ based ultra-broadband photodetectors.

devices. Self-powered flexible MoS₂ photodetectors have a great potential in the field of healthcare for their applications in point-of-care wearable biosensors for monitoring vital signs and metabolic parameters, bioimaging, diagnostics, and cardiac pacemakers.

9. Conclusions and perspective

The role of 2D TMDs, especially atomic layered MoS₂ and its diverse vdWHs, in developing photodetectors for optoelectronic applications has been discussed, where the photoresponse can be tailored over a broad spectral range from UV to IR regions. The review has particularly summarized the progress made in the design and fabrication of broadband photodetectors using atomic layered MoS₂ and MoS₂-based hybrid heterostructures for wearable optoelectronics. The performance of MoS₂ photodetectors can be dramatically improved and optimized by forming MoS₂ hybrid heterostructures with 0D QDs,^{193,194} 2D materials,^{169,196,226,281,309,329,334–336} ferroelectric polymers,^{137,324} halide perovskites,^{179,191,192,327} piezo-phototronic effect,^{221,258,272} and *via* chemical doping,^{187,326} as has been studied for the phototransistor, photodiode and photoconductor components. The photoresponsivity, detectivity and photoresponse of MoS₂ hybrid heterostructure-based photodetectors vary by several orders of magnitude depending on the nature of the vdWHs, doping agents, optical wavelengths and intensity of applied incident power.^{122,171,178,179,199,226,321,325,326,326} Temperature-dependent ultrahigh photoresponsivities of 5×10^8 A W⁻¹ to 1×10^{10} A W⁻¹ have been observed for the monolayer graphene/ML MoS₂ hybrid vdWH-based photodetectors.¹²² Similarly, an ultrahigh detectivity of 3.8×10^{15} Jones was recorded for the rGO–MoS₂/pyramid Si hybrid heterostructure-based photodetector, which also operated in the ultra-broadband spectral range of 350 nm to 4.3 μm (UV to MIR)

due to the bandgap narrowing instigated by the sulfur (S) vacancy defects in MoS₂ crystals.¹⁷⁸ The MoS₂ hybrid heterostructure-based photodetectors also exhibit improved environmental stability for extended periods of time.^{162,187,192} The flexible photodetectors developed from MoS₂ layers and MoS₂ hybrids with TMDs, graphene, CNTs, and ZnO and their applications in wearable optoelectronics have been discussed. The figures-of-merit of a very wide range of MoS₂-based photodetectors have been compared in terms of their important parameters including photoresponsivity, detectivity, and response speed measured at different wavelengths and incident laser power densities.

The integration of MoS₂ photodetectors with other flexible electronic, photonic and optoelectronic functional devices remains challenging. Research activities on MoS₂ based photodetectors is emerging on a fast pace.^{446–451} The large family of 2D materials can be explored for developing photodetection devices. For example, 2D TMDs such as MoSe₂, MoTe₂, WS₂, WSe₂, PtS₂, PtSe₂, PtTe₂, PdS₂, PdSe₂, PdTe₂, GeS₂, GeSe₂, HfS₂, TiS₂, FeS₂, CoS₂, NiS₂, SnS₂, MoSe₂, NbSe₂, TaSe₂, NiSe₂, FeSe₂, and CoSe₂ can be explored for developing new vdWHs for high-performance photodetectors. Tremendous opportunities exist for TMD-based vdWHs with other nanomaterials, including BP, graphene, h-BN, Janus TMDs, mono-chalcogenide layered materials (GaS, GaSe, GaTe, InS, InSe, InTe, SnS, SnSe, SnTe, PbS, PbTe, *etc.*), 2D transition-metal carbides, nitrides and carbonitrides (MXenes), transition-metal trichalcogenides (MX₃; M = Ti, Zr, Hf, Nb, Ta, and X = S, Se, Te), perovskites, surface-functionalized 2D TMDs and transition metal oxides (TMOs), inorganic semiconductors and organic materials, which represent the future direction of TMD-based photodetectors for their application in fields from biomedical imaging to optical communication. TMDs are also advancing as multifunctional materials for future wearable electronic and optoelectronic devices as an inexpensive alternative to the costly manufactured semiconducting materials currently employed in the electronics industry. 2D atomic layered material-based hybrid vdWHs hold great potential for self-powered flexible photodetectors in the healthcare industry especially for biomedical imaging, point-of-care wearable biosensors for monitoring vital signs and metabolic parameters, cardiac pacemakers, and for patients. There are many significant challenges to producing low-cost, large-scale high-performance photodetector devices for commercial applications, therefore factors such as optimization of the photoresponse, reproducibility, long-term environmental stability of photodetector devices, mechanical flexibility, packaging, and the toxicity of nanomaterials^{452–460} used in photodetectors need much more attention. TMDs are very promising nanomaterials for developing high-performance flexible photodetectors and for integration of such devices into the next generation of wearable technologies.

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Conflicts of interest

There are no conflicts to declare.

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