Recent advances in perovskite/2D materials based hybrid photodetectors

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Abstract
Since 2009, metal halide perovskites have attracted a great deal of attention in different optoelectronic applications, such as solar cells, photodetectors (PDs), light-emitting diodes, lasers etc, owing to their excellent electrical and optoelectrical properties. However, since the discovery of graphene, atomically thin 2D materials have been the central focus of materials research due to its exciting properties. Thus, integrating 2D materials with perovskite material can be highly promising for various optoelectronic applications, in particular for ultrasensitive photodetection. In these PDs, 2D materials serve various roles, such as charge transport layer, Schottky contacts, photo absorbers, etc, while perovskite is the light-harvesting active layer. In this review, we focus on the recent findings and progress on metal halide perovskite/2D material phototransistors and hybrid PDs. We comprehensively summarize recent efforts and developments of perovskite/graphene, perovskite/transition-metal dichalcogenides, perovskite/black phosphorus, and perovskite/MXene based phototransistor and heterojunction PDs from the perspective of materials science and device physics. The perovskite/2D material phototransistor can exhibit very high photoresponsivity and gain due to the amplification function of transistors and the pronounced photogating effect in 2D material, while perovskite/2D material heterojunction PD can operate without external bias due to built-in potential across the heterojunction. This review also provides state-of-the-art progress on flexible, transparent, self-powered and PD systems and arrays based on perovskite/2D materials. After summarizing the ongoing research and challenges, the future outlook is presented for developing metal halide perovskite/2D material hybrid PDs for practical applications.

1. Introduction

Photodetectors (PDs) are representative optoelectronic devices that can convert optical signals into electrical ones based on the photoelectric effect, which are one of the key components in contemporary multifunctional technologies [1–7]. The efficient and ultrasensitive photodetection is very important in a wide range of fields, such as optical communication, spectroscopy, biomedical imaging, environmental monitoring, biological/chemical sensing, security, fire detection, night vision, motion detection, video imaging, etc [1–6, 8]. In the high-performance PD, typically semiconducting materials are used which efficiently absorb incident photons and produce electron–hole pairs upon photoexcitation, and built-in electric field or external bias is used to separate photogenerated carriers effectively to generate an electrical output [1–6, 9, 10]. Currently, the PDs in the entire UV–NIR spectrum are dominated by detectors fabricated from inorganic semiconductors, such as GaN, Si, and InGaAs [8, 11]. Typically, GaN operates in the UV region, while Si and InGaAs respond in Vis–NIR and NIR–far NIR region, respectively. However, there are certain limitations and shortcomings in these PDs due to their complicated manufacturing technologies, high-quality crystals with low defects, high power consumption, high cost and bulkiness of the
devices. Therefore, low-cost PDs with broadband light detection, high sensitivity and fast response are highly desirable. Low-cost and solution-processable optoelectronic materials, such as organic semiconductors, different semiconductor nanostructures have been explored for efficient photodetection. However, the performance of these PDs is often restricted due to their poor charge–carrier mobility. Interestingly, halide perovskite-based semiconductors have shown great promises in low-cost PDs.

We provide below a brief account of the important properties of perovskite and 2D materials that are most relevant for PD applications.

1.1. Fundamentals of photodetectors

A PD generally converts the incident optical signal into charge–carrier flux. A PD can work based on two mechanisms: (a) an external field-assisted transport of photoexcited carriers in which the applied external field can stimulate a large number of carrier transport by detrapping of trapped photogenerated carriers; (b) the photogenerated charge carriers produce an electric field, which decreases the injection barrier and consequently amplifies the charge injection. The diffusion and drift of photogenerated charge carriers, the recombination dynamics, and the role of contacts need to be fully understood for the design and optimization of the device for a good PD [12]. Typically, there are two types of architectures for PDs: (a) lateral structure device and (b) vertical structure device [13, 14]. In a lateral structure device, active materials are directly connected to lateral electrodes, while the vertical structure device consists of different stacking layers. Lateral structure-based PDs include photoconductors and phototransistors, while vertical structure PDs include photodiodes, heterojunction PDs, solar cell structure (p-i-n or n-i-p) devices. Except for phototransistors, all the other PDs are two-terminal devices; one electrode is the cathode, and another is the anode. In contrast, phototransistors are PDs with three-terminal: gate, source, and drain electrodes [15, 16]. Figure 1(a) depicts a simple schematic of vertical structure PD while figure 1(b) shows a schematic illustration of PD with a lateral structure.

The photoconductor is a simple lateral system with two electrodes and light-absorbing direct bandgap semiconductors in between the electrodes. In this type of PDs, an external bias is applied to observe the light-induced change in conductivity. These PDs can exhibit high gain through multiple carrier recirculation. The metal-semiconductor Schottky junction also plays an important role in photodetection. The Schottky photodiode can operate in different modes: (a) photoexcitation and carrier generation in semiconductors, (b) generation of the carrier from metal to semiconductor over the Schottky barrier. The Schottky PDs can be very useful for broadband photodetection.

Photo field-effect transistors (photo FETs) comprise of a photoconductive channel with drain and source electrodes on two sides, and the carrier transport of the channel can be controlled by the applied gate bias. However, incident light produces photocarriers and serves as an additional gate. Thus, the electrical gate can be either open-circuited or externally biased to control the photoresponse characteristics of the device. In photo FETs, the channel materials should simultaneously exhibit both high carrier mobility and superior light-harvesting capability for efficient charge generation [17]. Note that 2D material shows superior mobility for carrier transport, whereas perovskites have excellent photodetector conversion efficiency with high absorption coefficient. Thus, the perovskite/2D material hybrid system is a very promising channel material for photo FET. Under illumination, electron and hole pairs are generated in the perovskite layer, which further transports to the source and drain electrodes via the 2D material layer. The applied gate voltage can effectively increase the separation of photocarriers. In a photo FET, often one type of carrier is intentionally immobilized in the device, while another type is free to move and contribute to photocurrent by suitable choice of the materials and designs in the device [18]. This phenomenon is known as photogating effect. Due to the inherent amplification in FET, phototransistor can have high external quantum efficiency.
(EQE) exceeding 100%, high gain, and high responsivity, which can be controllable by gate bias. Though photo FET exhibits high responsivity, the response speed is significantly lower due to the trapped carriers.

Photodiodes are vertical structure devices in which the active layers are sandwiched between electrodes. Photodiodes may be p–n junction type or semiconductor heterojunction (like type-II) type. Photodiodes possess a very low dark current and very high detectivity of light due to interface layers. Due to the built-in potential, the photodiodes can also detect light without applied bias. These devices are known as self-powered PDs, which are very attractive from the application perspective. In the reverse-bias condition, photodiodes show superior performance due to an increase in charge collection efficiency. Besides, the response speed of photodiodes is generally higher compared to the other kinds of PDs.

Perovskite PDs with ‘solar cell structure’ (p-i-n or n-i-p) show excellent photodetection without any applied bias due to the photovoltaic effect. In this type of vertical structure device, perovskite serves as light-absorbing intrinsic (i) active layer and one-hole transporting layer (HTL) (p) and another electron transporting layer (ETL) (n) are used to separate and transport photogenerated electrons and holes to the electrodes. However, as this involves the deposition of additional layers, it is not very commonly used.

To quantitatively measure the performance of a PD, there is a series of figures of merit, such as photoresponsivity ($R$), gain, specific detectivity ($D^*$), EQE, linear dynamic range (LDR) and response speed, etc. The photoresponsivity of a PD indicates how efficiently the detector responds to an incident optical signal. The responsivity of a PD is a very important figure of merit that describes incident photon to the electrical signal conversion efficiency of the given PD. Responsivity is defined as the ratio of photocurrent to the power of incident light on the active area of PD by the following equation,

$$R = \frac{I_{ph}}{P_{in}}, \quad (1)$$

where $I_{ph}$ is the photocurrent defined as the difference between the current of the device in the presence of illumination and dark condition ($I_{ph} = I_{light} - I_{dark}$). $P_{in}$ is the incident optical power on the active area of the PDs. Responsivity is generally proportional to the EQE for a PD. It is very significant for a good PD to have a high conversion rate from photons to electron–hole pairs.

The photoconductive gain ($G$) is defined as the number of charge carriers circulating through an external circuit per incident photon. It can be expressed as

$$G = R \times E_{hv}, \quad (2)$$

where $E_{hv}$ represents the energy of the incident photons. By increasing the photoconductive gain of a PD, the sensitivity of the PD is also increased, which further makes it suitable for different applications, such as receivers in optical fiber communications, single-photon counting, laser microscopy, high-resolution imaging, etc [19]. The gain can also be obtained by measuring transit time and carrier-recombination lifetime as expressed by the given equation:

$$G = \frac{\tau_t}{\tau_{tr}} = \frac{\tau_t}{d^2/\mu V}, \quad (3)$$

where $d$ is the device thickness, $\mu$ is the carrier mobility and $V$ is the applied bias.

The specific detectivity ($D^*$) of a PD signifies the capability of detecting low-level light signals, taking into account the influences of the photocurrent and dark current. $D^*$ can be calculated by the equation:

$$D^* = \frac{R}{(2qJ_d)^{1/2}}, \quad (4)$$

where $R$ is the spectral responsivity, $q$ is the elementary electron charge and $J_d$ ($J_d = I_{dark}$/area of the device) is the dark current density of the device. Note that this equation is valid only under the approximation that the dark current of the PD is dominated by the shot noise. The high detectivity of a PD represents that the PD can detect very low-intensity light signals. The specific detectivity of a PD can also be expressed by noise-equivalent power and the electrical bandwidth of device noise:

$$D^* = \frac{(AB)^{1/2}}{\text{NEP}}, \quad (5)$$

where NEP represents the noise equivalent power, $A$ is the working area of the PD and $B$ is the bandwidth. The NEP of a PD is defined as the signal power that gives a signal-to-noise ratio of one. For a sensitive PD, NEP should be small.
A good PD should detect incident light with an identical responsivity over a wide range of light intensity. This characteristic of the PD is defined as the LDR. LDR can be expressed as

$$LDR = 20 \log \left( \frac{I_{ph}}{I_d} \right),$$  (6)

where $I_{ph}$ and $I_d$ are the photocurrent and dark current densities of the device, respectively. In the LDR limit, a PD can detect light precisely.

One of the critical parameters of a PD is response speed. The response speed of a PD is calculated from the rise and fall times of a transient photocurrent. The photocurrent growth and decay times can be defined as the times taken for a photocurrent to reach 63.2% and to drop to 36.8% of the steady-state values, respectively. For pulsed illumination cases, the rise and fall times are the time intervals between photocurrent level reaching $\sim$10% and $\sim$90% of its saturation level upon on/off illumination cycles. The rise and fall time constants can also be calculated from the curve fitting of the temporal photoresponse by the exponential function [20].

### 1.2. Properties of perovskites for photodetection

Over the past one decade, halide perovskite materials with structural formula $ABX_3$ (where A and B are cation and X is halide anion) have emerged as a promising class of semiconductors, which are being studied extensively for their applications in different optoelectronic devices, such as, solar cells, light-emitting diode (LED), PD, laser etc [2, 11, 21–37]. A tremendous research attention has been drawn for the investigation of high-performance hybrid perovskite-based optoelectronic devices due to its unique optical and electronic properties, such as high optical absorption coefficient, high carrier mobility, tunable optical bandgap, long electron–hole diffusion lengths, and low-cost and low-temperature processing [2, 13, 21, 38–40]. Recently, the power conversion efficiency (PCE) of perovskite-based solar cells has achieved $\geq$23% [26, 41–43]. Thus, this new family of semiconducting optoelectronic materials with good solution-processability and low-cost fabrication can be a good alternative of well-established crystalline Si and group III–V semiconductors. Solution-processed low-cost metal halide perovskites have direct-bandgap with large absorption coefficients in the UV–visible range as compared to that of its low value in Si. For example, at 600 nm, the absorption coefficient of CH$_3$NH$_3$PbI$_3$ perovskite is $\sim$10$^5$ cm$^{-1}$, while that of Si is $\sim$10$^4$ cm$^{-1}$ [44]. Due to the high absorption coefficient, a very thin layer of perovskite is adequate for almost complete light absorption in optoelectronic devices. Recently, low-cost perovskite PDs have been revealed to high responsivity and fast photoresponse, which makes them promising candidates for different light-detection devices [13, 45–58]. The responsivity ($R$) and detectivity ($D^*$) of perovskite PDs have achieved over $10^9$ A W$^{-1}$ and $10^{16}$ Jones, respectively, while the response speed is $<1$ ns, which are significant compared to other PDs [13, 48]. However, the application of perovskites in optoelectronic devices is limited due to the poor air-stability and toxicity of lead-based perovskite [42, 59]. The most of the reported high-performance perovskite PDs with high responsivity and ultrafast response time are planar or mesoporous ‘perovskite solar cell structure’. Perovskite solar cells are photovoltaic devices with vertical architecture, which consists of different stacked layers. It generally comprises of (a) transparent conducting oxide electrode, (b) electron transporting material (ETM), (c) light-harvesting perovskite active layer, (d) hole transporting material (HTM), (e) counter electrode. In these types of p-i-n or n-i-p PDs, typically one electron transporting material and another hole transporting material are used to separate and transport photogenerated electrons and holes, while the perovskite layer acts as a light-absorbing active material. Dou et al have reported a vertical architecture of perovskite PD, similar to a solar cell configuration of ITO/PEDOT:PSS/CH$_3$NH$_3$PbI$_3$–xCl$_x$/PCBM/PFN/Al [60]. This PD showed a broad photoresponse in the wavelength region from 300 to 800 nm, while the reported detectivity was observed to be $10^{14}$ Jones, which is nearly one order of magnitude higher than the detectivity of a commercial Si-based PD. Bao et al presented a highly flexible self-powered perovskite PDs with the configuration of PEN/Au nanowire/CH$_3$NH$_3$PbI$_3$/PCBM/Al with responsivity 321 mA W$^{-1}$ and response speed of 4 $\mu$s [61]. Sutherland et al demonstrated a self-powered perovskite PD with the structure of FTO/TiO$_2$/Al$_2$O$_3$/PCBM/CH$_3$NH$_3$PbI$_3$/spiro OMeTAD/Au by exploiting interface engineering of the device with the response speed of 1 $\mu$s and responsivity of 0.4 A W$^{-1}$ [62]. Zhang et al reported perovskite PDs with a controllable photoconductive gain by designing a trapped-electron-induced hole injection structure of ITO/PEDOT:PSS/CH$_3$NH$_3$PbI$_3$/PCBM:F4–TCNQ/BCP/Au [63]. Interestingly, these PDs with solar cell architecture can operate without any external bias. The high performance of the perovskite PDs was attributed to the outstanding intrinsic optical and electronic properties of the solution-processed hybrid perovskites, as well as judicious device configuration and interface engineering.
1.3. Properties of 2D materials for photodetection

Since the discovery of graphene, 2D materials have attracted extensive attention due to their unique structures and their mechanical and physical properties [64–68]. The large family of 2D materials is mainly composed of the 2D atomic layer with the same elements, such as graphene and black phosphorus (BP), transition-metal dichalcogenides (TMDs), chalcogenides, oxides, nitrides, thiophosphates, halides etc [65, 69]. Due to the unique structure and novel properties, these 2D layered materials are very promising in various applications, such as solar cells, PDs, LEDs, biosensors, and so on [3, 66, 70–72]. Monolayer and few-layer 2D materials have been extensively applied in PDs, which shows very fast photoresponse speed along with high photoresponsivity due to the high carrier mobility, unique optical properties, and superior mechanical flexibility [70, 73–75].

However, the performance of these 2D material-based PD is usually limited by the low light-harvesting capacity of the monolayer and few-layer material. On the other hand, halide perovskite has an excellent capability of light-harvesting with a large absorption coefficient. Thus, the integration of 2D TMD materials with high light-harvesting perovskite materials can lead to substantial enhancement in the light absorption resulting in high photoresponsivity and fast response due to high carrier mobility in atomically thin 2D TMDs.

In this review, we present state-of-the-art progress in the PDs based on halide perovskite/2D material hybrids. First, we discuss the fundamental operational principles, device architectures, and different figures of merit of PDs. Next, we discuss on advances in perovskite/graphene based heterostructure PDs followed by a state-of-the-art account of the progress in TMDs/perovskite based heterostructure PDs. In particular, we discuss the advances in the perovskite/MoS₂, perovskite/WS₂, and perovskite/other TMDs (such as MoSe₂, WSe₂, PtSe₂, PdSe₂) based PDs. Next, we discuss about perovskite/BP-based PDs and perovskite/MXene based PDs, which are more recent additions to the family of 2D materials and are under development.

Finally, we summarize the challenges and opportunities in the field followed by the future outlook for developing metal halide perovskite/2D material hybrid PDs for practical applications.

2. Perovskite/2D material photodetector

2.1. Perovskite/graphene photodetector

Graphene is a very interesting 2D material for optoelectronic applications [76, 77]. The concentrations of electrons and holes in the graphene layer exceed 10¹³ cm⁻², while the carrier mobility is higher than 15 000 cm²V⁻¹s⁻¹ [78]. Because of its zero bandgap, graphene exhibits an extremely broadband absorption from UV to THz region. Most of the graphene-based PDs are constructed with a metal–graphene Schottky junction, a homogeneous p–n junction, or a tunneling barrier junction. The metal–graphene–metal Schottky junction PDs can operate by taking advantage of the work-function difference between a contact metal and graphene, which shows a very ultrafast photoresponse [79]. Xia et al demonstrated ultrafast phototransistor with monolayer and few-layer graphene with a 40 GHz bandwidth and a 0.5 mA W⁻¹ photoresponsivity [80]. Mueller et al reported metal–graphene–metal PD with maximum external photoresponsivity of 6.1 mA W⁻¹ by using two different metal electrodes [81]. The asymmetric metallization scheme results in a break of the mirror symmetry of the internal electric-field profile in conventional graphene FET channels, enabling an efficient photodetection. In the back gated graphene FET, the photocurrent is generated due to the illumination of metal/graphene interfaces, and the field arises from the charge transfer from the metal electrode to graphene. For the controllable p- or n-type doping in graphene, mostly surface chemical modification methods have been adopted. Liu et al reported a large area and flexible p–n junction graphene PD with good photoresponsivity in the infrared (IR) region [82]. The p–n junction in the graphene layer was fabricated by selected-area chemical doping in the chemical vapor deposition (CVD) process [82, 83]. Liu et al reported an ultra-broadband phototransistor consisting of a pair of stacked graphene monolayers separated by a thin tunnel barrier [84]. In the presence of light, hot carriers are generated in the top layer of graphene and further tunnel into the bottom layer, which leads to a charge build-up on the gate and a strong photogating effect occurs on the channel conductance. This PD could operate in the visible to the mid-infrared range, with mid-infrared responsivity higher than 1 A W⁻¹.

The fundamental shortcomings of pure graphene-based PDs are its zero bandgap and poor absorbance of the incident light. Single-layer graphene can absorb only 2% of incident light over a broad wavelength region [84]. Due to the high conductivity, the dark current is also very high in pure graphene-based PDs. Thus, the integration of additional high light-harvesting materials, such as halide perovskite, can lead to substantial enhancement in the light absorption resulting in high photoresponsivity along with fast response due to high carrier mobility in graphene.

In 2015, Lee et al first reported a novel hybrid photo FET consisting of graphene and CH₃NH₃PbI₃ perovskite layers [85]. CH₃NH₃PbI₃ perovskite/graphene hybrid PD was fabricated on a SiO₂/Si substrate...
A heavily n-doped Si wafer served as a gate electrode, while 300 nm thick SiO$_2$ layer was employed as the dielectric layer for the photo FET. Au electrodes deposited by thermal evaporation performed as source and drain electrodes. Further, CVD-grown single-layer graphene was transformed and pattern by photolithography. Finally, the CH$_3$NH$_3$PbI$_3$ perovskite layer was deposited by spin coating of perovskite precursor onto the graphene surface. The UV–visible–IR absorption spectra of the perovskite–graphene hybrid system and pristine perovskite layer are shown in figure 2(b). The perovskite layer shows broad absorbance over the UV–visible–NIR region, which is essential for the broadband photodetection, and the absorption of the hybrid system was nearly unaltered in the presence of single-layer graphene. Interestingly, the photoluminescence (PL) intensity of the perovskite–graphene hybrid film was quenched by $\sim 65\%$ compared to the pristine perovskite film (figure 2(c)), which was attributed to the effective transfer of photogenerated carriers through $\pi-\pi$ interaction between perovskite and sp$^2$ hybridized graphene layer. The incident wavelength-dependent photoresponsivity and EQE of the hybrid device match well with the absorbance of perovskite, i.e. the responsivity increases in the wavelength region <780 nm (figure 2(d)). The detection limit of the PD was 400–850 nm, which matches well with the absorbance of the composite film. Thus, the photocurrent was generated mainly due to the photoexcitation of the active perovskite layer. The variation of photoresponsivity and EQE as a function of the illumination power for the perovskite/graphene hybrid system is shown in figure 2(e). The inset of figure 2(e) shows the variation of detectivity of the hybrid PD. The hybrid PD showed a photoresponsivity of 180 A W$^{-1}$ and an EQE of $\sim 5 \times 10^4\%$ for an optical power in the microwatt range, while it decreases with the increase in intensity of the incident light. Note that the decrease in responsivity with increasing incident power is attributed to a higher probability of scattering and recombination under stronger illumination. The device exhibited good photoswitching behavior under the illumination of 520 nm with 0.5 mW incident power with gate ($V_g$) and drain voltages ($V_{ds}$) of 0 and 0.1 V, respectively (see figure 2(f)). The upper portion of figure 2(f) shows the pulsed laser illumination response, while the bottom one is the temporal response of the photocurrent. The photocurrent growth and decay time constants were observed to be 87 and 540 ms, respectively. Thus, this work demonstrated a hybrid PD consisting of graphene and CH$_3$NH$_3$PbI$_3$ perovskite layers with a broad spectral response and high EQE due to increased optical absorption and photogating mechanisms of the hybrid system. However, photoresponsivity of the reported device was relatively low compared to the other reported photo FETs based on perovskite/graphene which may be due to the defects and poorer morphology of perovskite active layer.

Wang et al reported a hybrid PD with an exceptionally high photoconductive gain of $\sim 10^9$ electrons per photon and responsivity of $\sim 6.0 \times 10^5$ A W$^{-1}$ that consists of monolayer graphene covered with a thin layer
of dispersive CH$_3$NH$_3$PbBr$_2$I halide perovskite islands [86]. The perovskite–graphene hybrid material was the photoactive channel material in phototransistor (figure 3(a)). CVD-grown graphene was transferred on Si/SiO$_2$ substrate by wet transfer method, while perovskite precursor solution was deposited on top of the graphene layer by antisolvent assisted spin coating method to form perovskite islands. The transfer characteristics of the bare graphene transistor in dark condition show a charge-neutral point at ~40 V, which is attributed to the p-doping effect caused by the SiO$_2$ substrate and water trapped beneath the graphene film (figure 3(b)). In the case of graphene/CH$_3$NH$_3$PbBr$_2$I perovskite island hybrid phototransistor, the charge neutral point was shifted to lower gate voltage, which was attributed to the decrease in p-doping effect in graphene due to electron transfer from perovskite. Due to the work function mismatch between these two materials, electrons were transferred from perovskite to graphene and a built-in electric field was developed, which led to band bending in perovskite to enable effective detrapping of photocarriers, as illustrated in the inset of figure 3(b). The temporal response of the photocurrent of the hybrid PD under the illumination of 405 nm light with power 921.5 nW shows good photo-switching behavior (figure 3(c)). The detection range of the PD was in the range of 250–700 nm, which matches with the absorbance of the perovskite active layer. The growth and decay times for photocurrent of perovskite–graphene island phototransistor were measured to be less than 120 and 750 ms, respectively (figure 3(d)). The variation of responsivity with drain–source ($V_{ds}$) voltage at zero gate voltage is shown in figure 3(e). The responsivity was observed to be 6.0 × 10$^5$ A W$^{-1}$ under the illumination of 405 nm with a power of 1.052 nW and $V_{ds}$ of 3 V. The photocurrent was increased linearly with the increase of incident light power at relatively low power and eventually gets saturated at high power due to the increased recombination rate of photoexcited carriers (figure 3(f)). The photoreponsivity decreases with the increase in light power (figure 3(f)). Thus, the high performance of CH$_3$NH$_3$PbBr$_2$I perovskite islands/graphene phototransistor is attributed to the effective charge transfer and photogating effect. Compared to the bulk perovskite thin-film case, the perovskite
islands have a low bulk recombination rate of photogenerated carriers, which results in the enhanced performance of the PD [85, 86]. Further, Spina et al reported CH$_3$NH$_3$PbI$_3$ nanowire/graphene phototransistor, which shows photoresponsivity of $2.6 \times 10^6$ A W$^{-1}$ [87]. CH$_3$NH$_3$PbI$_3$ nanowire was fabricated by the slip-coating method on top of single-layer graphene FET (figure 3(g)). The variation of responsivity of the PD with incident light intensity is shown in figure 3(h). The rise and fall times of the hybrid CH$_3$NH$_3$PbI$_3$ nanowire/graphene PD were observed to be 55 and 75 s, respectively (figure 3(i)), which are comparatively higher than the perovskite-film based PD. The photocurrent response times can be divided into two components (figure 3(i)). The slow component is attributed to the multiplicity of charge traps in the nanowire film that stems from different surface states. The high responsivity of CH$_3$NH$_3$PbI$_3$ nanowire/graphene is attributed to the enhanced light-harvesting capacity of perovskite with nanowire morphology.

Plasmonic metal nanoparticles (NPs) play a vital role in boosting the performance of optoelectronic devices [88–90]. Under photon illumination, plasmonic NPs can give rise to localized surface plasmon resonance (LSPR), which prompts strong photon absorption and scattering. Thus, due to the plasmonic scattering and near-field enhancement induced excitation effects, the photon absorption of the surrounding material can be improved [91–93]. Plasmonic metal NPs also helps in better charge separation and transport. Sun et al demonstrated a CH$_3$NH$_3$PbI$_3$/graphene/Au NP hybrid photo FET with enhanced performance [94]. The perovskite active layer was physically separated by the ultrathin graphene layer from the Au NPs film. Au-NPs with a diameter of $\sim$40 nm were synthesized by a well-known citrate reduction method and transferred on the Si/SiO$_2$ substrates through a self-assembled monolayer (SAM) of 3-aminopropyltriethoxysilane (APTES). The SEM image of the Au-NP-coated Si/SiO$_2$ substrate is shown in figure 4(a), which confirms dense and uniform coverage of Au NPs on the substrate. The absorption of Au NPs is shown in the inset of figure 4(a), which depicts a plasmonic absorption peak at $\sim$530 nm. The schematic illustration of the CH$_3$NH$_3$PbI$_3$/graphene/Au NP hybrid phototransistor is shown in figure 4(b). On top of the Au-NP film-coated Si/SiO$_2$ substrate, graphene was transferred, and then CH$_3$NH$_3$PbI$_3$
perovskite film was deposited on top of it by spin coating of the perovskite precursor solution. As illustrated in figure 4(b), in the CH$_3$NH$_3$PbI$_3$/graphene/Au NP hybrid device, due to the near-field enhancement of Au-NPs, most of the photogenerated carriers are produced near the interface between graphene and perovskite layer. The enhanced photogenerated carriers due to the LSPR effect of Au NPs results in the enhanced photocurrent of the hybrid device. The transfer characteristics of pristine (without Au NPs) and the hybrid PD (with Au NPs) at $V_{DS} = 0.1 \, \text{V}$ under the illumination of 532 nm for different intensities are shown in figures 4(d) and (e), respectively. The transfer characteristics confirm the p doping in graphene due to the transfer of photogenerated charge carriers from perovskite to graphene, and a slightly enhanced photocurrent was observed in the hybrid device. The light intensity–dependent responsivity and EQE of the pristine and hybrid PDs are shown in figure 4(f). The responsivity of the hybrid device was increased by $\sim 100\%$ compared to the device without Au-NPs, as shown in figure 4(f). Interestingly, as the photogenerated carriers are mostly produced near perovskite–graphene interface (figure 4(c)) due to the LSPR effect of Au NPs, the recombination rate can be greatly reduced, which leads to a slower drop of responsivity with increasing light intensity in the hybrid device compared to the pristine one (figure 4(c)). However, the highest responsivity was observed to be only $\sim 2 \times 10^4 \, \text{A W}^{-1}$. The photoresponse speed of the pristine and hybrid PD is shown in figure 4(g). The photocurrent growth time increased in the hybrid device compared to the pristine device. Thus, the possible mechanisms for the enhanced performance of the hybrid device may be attributed to: (a) the increase in light-harvesting in CH$_3$NH$_3$PbI$_3$ perovskite due to the near-field enhancement on the surface of Au-NP, (b) superior separation of excitons and the extraction of carriers due to Au-NPs, (c) trap of minority carriers or suppression of the carrier recombination in CH$_3$NH$_3$PbI$_3$ by Au NPs, (d) generation of hot carriers in Au-NPs and transfer into graphene and thus increase the photocurrent.

Another approach to improve the performance of perovskite/2D material hybrid PD is the incorporation of charge–transporting layers, which enhances the efficiency of photocarrier separation. Note that generally in perovskite solar cells, one ETL and another HTL are used, which separate and transport the photo-induced carriers. Xie and co-workers reported CH$_3$NH$_3$PbI$_{1-x}$Cl$_x$ perovskite/poly(3-hexylthiophene) (P3HT)/graphene multi-junction ultra-sensitive phototransistor [95]. The device configuration of CH$_3$NH$_3$PbI$_{1-x}$Cl$_x$/P3HT/graphene multi-junction PD is shown in figure 4(h). In this multi-junction device, CH$_3$NH$_3$PbI$_{1-x}$Cl$_x$ mixed halide perovskite layer serves as a light-absorbing active material that absorbs light and generates electron and hole pairs, while P3HT acts as an HTL to efficiently separate electrons and holes reducing the carrier recombination. The graphene layer provides a fast charge transfer channel. The light intensity–dependent responsivity of CH$_3$NH$_3$PbI$_{1-x}$Cl$_x$ perovskite/P3HT/graphene phototransistor as a function of back–gate voltage, $V_G$, is shown in figure 4(i). The device exhibits a responsivity as high as $\sim 4.3 \times 10^9 \, \text{A W}^{-1}$ along with a photoconductive gain of $\sim 8.9 \times 10^9$ at the excitation wavelength of 598 nm (figure 4(i)). The maximum responsivity of the device with P3HT HTL was increased by one order of magnitude compared to the device without the HTL layer due to the enhanced carrier separation by the HTL. The photoresponse speed of the phototransistors with a perovskite layer thickness of $\sim 300$ nm (top panel) and $\sim 40$ nm (bottom panel) under the illumination of intensity 14.15 mW cm$^{-2}$ at $V_G = -25 \, \text{V}$ are shown in figure 4(j). Here, the device with a lower thickness of perovskite layer shows faster response due to the superior photogating effect. The PD showed good photoresponsivity in the wavelength range 400–800 nm. Note that the photoresponsivity of the device is significantly higher than the other reported perovskite/graphene PDs, which is attributed to the enhanced carrier separation by the HTL.

Qin and co-workers used [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) electron transport layer in CH$_3$NH$_3$PbI$_3$/graphene phototransistor for improved charge separation and transport [96]. They dissolved different amounts of PCBM (0%, 0.1%, and 1%) into the perovskite precursor solution and spin-coated it by a one-step antisolvent method on top of the graphene. After transforming into perovskite film, PCBM was observed near the grain boundaries of perovskite forming a bulk heterojunction (BHJ). Figure 5(a) shows the schematic architecture of BHJ–graphene PD. Under photon excitation, electrons, and holes were generated in the perovskite. The electrons were captured by PCBM while holes were transferred effectively to the graphene layer resulting in the superior carrier separation (figure 5(a)). Thus, a net amount of negative charges build up in the BHJ layer, which induces hole doping in graphene and lowers the Fermi level ($E_F$). The charge transfer process and the electronic structure of the heterojunction are shown in figure 5(b). The photocurrent growth time was observed to be 2.5, 49, and 92 s for a device with 0%, 0.1%, and 1% of PCBM, respectively. Thus, the longer photocurrent rise time found in the BHJ devices indicates that a longer time is required to attain the equilibrium carrier concentration within the active layer. This behavior is attributed to the de-trapping of electrons from PCBM, which is an extremely slow process that can last up to a few seconds. Similarly, the photocurrent decay times were found to be 65, 226, and 6203 s for the devices with 0%, 0.1%, and 1% of PCBM, respectively (figure 5(c)). The photo responsivities for the three devices as a function of incident illumination power are shown in figure 5(d). The responsivity of the pristine device was
observed to be $3 \times 10^4$ A W$^{-1}$ for an incident power of 1.2 nW, while it increases to $1 \times 10^5$ A W$^{-1}$ for the 0.1% PCBM device and $8 \times 10^5$ A W$^{-1}$ for the 1% PCBM device under the same incident power. Thus, the responsivity of the device with 1% of PCBM was increased by a factor of $\sim 30$ as compared to the device without the PCBM. Thus, a small amount of electron acceptor in the perovskite active material is responsible for the 30 fold enhancement of photo responsivity due to the superior carrier separation. Though the photo FET exhibited high responsivity, it showed very slow photoresponse speed compared to other perovskite/graphene photo FETs, which may be due to the trap states present at the interface of the device.

Despite excellent optoelectronic performances, conventional 3D organometal halide perovskite materials start degrading upon exposure to water and oxygen molecule. These limitations may restrict the practical applications of conventional 3D perovskite/graphene-based PD. On the other hand, 2D organic–inorganic perovskites emerge as an alternative material for their 3D counterparts in optoelectronic applications with improved moisture resistance. Shao et al demonstrated hexagonal boron nitride (hBN)/graphene/2D multiphase perovskite PD (figure 5(e)) [20]. (BA)$_2$(MA)$_{n-1}$Pb$_n$I$_{3n+1}$ (where BA$^+$ \(= \text{CH}_3\text{(CH}_2\text{)}_3\text{NH}_3^+$ and $n$ is the number of lead iodide octahedral layers between two BA$^+$ organic spacers). Depending upon integer $n$, the energy level diagram of the heterojunction is shown in figure 5(f). The photogenerated holes can be trapped effectively in 2D perovskite active layer while the electron can transfer easily to the graphene underlayer. Due to the excellent carrier mobility of graphene on top of the ultra-smooth h-BN flake, the responsivity of the detector was observed to be $\sim 10^6$ A W$^{-1}$ (figure 5(g)) while the photoresponse time was found to be less than 200 ms (figure 5(h)). The photo FET exhibits a detectivity of $3.3 \times 10^{10}$ Jones under incident light power of 42 pW. Interestingly, photo responsivity and the response time of the 2D perovskite/graphene phototransistor is much superior compared to the control CH$_3$NH$_3$PbI$_3$/graphene device (figures 5(g) and (h)). The PD exhibits good responsivity in a broad wavelength range from 400 to
Flexible PDs are highly desirable for different daily life applications. It can be attached to human skin for human–machine interfaces or combined into clothing to detect harmful light while being worn [97, 98]. Dang and coworkers fabricated CH$_3$NH$_3$PbI$_3$/graphene hybrid phototransistor on a flexible substrate [99]. The photo FET consisted of a photoactive CH$_3$NH$_3$PbI$_3$ perovskite and charge transporting graphene hybrid material that was a channel within the phototransistor. The hybrid channel was placed on top of the gate dielectric layer of Al$_2$O$_3$/polyvinylphenol/Al$_2$O$_3$ fabricated on the Ni gate electrode over the flexible polyimide substrates (figure 6(a)). Figure 6(b) shows the variation of responsivity of the flexible PD with incident light intensity. The maximum responsivity of the device was observed to be ∼115 A W$^{-1}$, which is comparable to the first reported perovskite/graphene phototransistors on a rigid substrate [85]. The comparison of responsivity (figure 6(c)) and photoresponse time (figure 6(d)) up to 3000 bending cycle to a radius of 12 mm shows excellent performance of the device. No notable change in responsivity and response time was observed up to 3000 bending cycles suggesting robust bending durability of the flexible detector. Though the photoresponsivity of the device was relatively low, these results were significant for perovskite-based flexible PDs. Cheng and colleagues reported a barristor-type phototransistor based on graphene/CH$_3$NH$_3$PbI$_3$ perovskite/graphene vertical heterostructure with maximum gain and responsivity of 2200 and 950 A W$^{-1}$, respectively [100]. The photocurrent rise time and fall time were obtained to be 22 and 37 ms, respectively.

In the above discussed PDs, solution-processable spin coating technique was used for the deposition of the perovskite active layer. Though these devices show excellent optoelectronic performances, reproducibility and large-scale perovskite deposition are the significant problems in spin coating and other solution-processable deposition techniques. Chang et al demonstrated a very smooth and compact perovskite film compared to the spin coating method by dual-source vapor deposition technique for graphene/perovskite hybrid phototransistor [101]. Graphene was transferred on a SAM-modified SiO$_2$ substrates, which significantly improved the electrical properties of graphene (figure 6(e)). Finally, poly(methyl methacrylate) (PMMA) was spin–coated on the entire surface of the device for encapsulation from ambient moisture and oxygen (figure 6(e)). The maximum responsivity of the device was observed to be ∼1.73 × 10$^7$ A W$^{-1}$, while the photoconductive gain was ∼10$^6$ (figure 6(f)). The highest detectivity of the device was observed to be 2 × 10$^{15}$ Jones. The PD exhibited good photoresponsivity in the wavelength region of 450–700 nm. These results are significant for the development of large area perovskite/graphene PD with high photoresponsivity.

Bessonov et al reported a perovskite/graphene phototransistor based on a CH$_3$NH$_3$PbI$_3$–PbS quantum dot (QD) absorbing layer, which shows a responsivity of ∼2 × 10$^5$ A W$^{-1}$: a specific detectivity of

740 nm. 2D multiphase perovskite/graphene hybrid device also showed excellent ambient air stability compared to the conventional 3D perovskite.

Figure 6. (a) Schematic architecture of CH$_3$NH$_3$PbI$_3$/graphene flexible phototransistor. (b) Variation of the responsivity with an incident power of the flexible PD. (c) Responsivity of the flexible PD before and after cyclic bending up to 3000 cycles at a 12 mm bending radius. (d) Comparison of the response speed of the flexible PD up to 3000 bending cycles. Reprinted from [99], Copyright (2016), with permission from Elsevier. (e) Schematic illustration of the vapor–deposited perovskite–graphene hybrid phototransistor. (f) Variation of responsivity as a function of light intensity for the hybrid phototransistor at different drain voltages. Reproduced from [101]. CC BY 4.0.
Figure 7. (a) Schematic illustration of the structure of NGQDs-CH$_3$NH$_3$PbI$_3$/mrGO phototransistor. (b) Energy level diagrams of CH$_3$NH$_3$PbI$_3$ perovskite, NGQDs, and mrGO heterojunction. (c) Variation of photocurrent and responsivity as a function of incident light intensity at 660 nm. (d) Wavelength-dependent responsivity and gain. (e) Temporal photocurrent response of the PD. (f) Stability of the device with and without PMMA encapsulation stored in ambient air. [103] John Wiley & Sons. © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (g) The schematic of the Schottky junctions of the perovskite PD (P), the graphene/perovskite vertical structure device without channel (GP), and graphene/perovskite/graphene horizontal structure PD with one channel on the graphene (GPG). (h) I–V characteristics of the Schottky junction devices under illumination. (i) Time-dependent photocurrent response of the three devices. [104] John Wiley & Sons. © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (j) Schematic illustration of CH$_3$NH$_3$Br$_3$/N-rGO hybrid PD device with the gap between two ITO plates to be 40 µm. (k) Photocurrent and responsivity at different incident power under 4 V bias. (l) Wavelength-dependent responsivity of the device. Inset shows the absorbance of the active material. Reprinted with permission from [105]. Copyright (2020) American Chemical Society.

5 × 10$^{12}$ Jones, and a gain of >10$^7$ [102]. The photocurrent response time was observed to be ~0.5–1 s. In their device, PbI$_2$ and CH$_3$NH$_3$I co-mediated ligand exchange in PbS QDs enhances surface passivation and facilitates electronic transport, yielding faster charge recovery with larger gain [102].

Qian et al reported a solution-processed phototransistor utilizing perovskite-nitrogen-doped graphene QDs (NGQDs) composite film and a mildly reduced graphene oxide (mrGO) layer [103]. The device configuration of NGQDs-modified perovskite/mrGO phototransistor is illustrated in figure 7(a). GO sheets were spin-coated and bonded to the SiO$_2$/Si substrate via a monolayer of APTES. Next, the GO film was mildly reduced by annealing at 150 ºC for 30 min to increase its electrical conductivity. The NGQDs–CH$_3$NH$_3$PbI$_3$ layer was spin-coated onto the mrGO layer. The device configuration is shown in figure 7(a). In this device, perovskite serves as light-absorbing active material, which produces photo-generated carriers, while mrGO acts as a charge transport layer. NGQDs serve as an electron acceptor in the device, which further optimizes the band alignment to facilitate photo-generated electron transfer from perovskite to mrGO. The band structure of the hybrid system is shown in figure 7(b). With the increase in light illumination intensity, the photocurrent increased gradually, while the responsivity and detectivity decreased (figure 7(c)). The responsivity of the PD was observed to be 1.92 × 10$^4$ A W$^{-1}$ at a gate voltage of 40 V and under light irradiance of 23.6 µW cm$^{-2}$, while the corresponding specific detectivity ($D^*$) was calculated to be 2.71 × 10$^{13}$ Jones (figure 7(c)). The incident light wavelength-dependent responsivity and gain are shown in figure 7(d). It is evident that the hybrid phototransistor can detect photon in the entire UV–visible–NIR region due to the broadband optical absorbance by the CH$_3$NH$_3$PbI$_3$ perovskite active layer. The photoconductive gain of the NGQDs–CH$_3$NH$_3$PbI$_3$/mrGO phototransistor was measured to be as high as 1.0 × 10$^4$ at 660 nm of the incident light (figure 7(d)). The photocurrent growth and decay times of the device were found to be 13 and 10 ms, respectively, at 660 nm (figure 7(e)). Thus, the hybrid photo FET exhibited faster photoresponse speed as compared to other reported perovskite/graphene based PDs. As perovskites possess poor ambient stability, the device was encapsulated with PMMA to improve the ambient stability. The PMMA encapsulated device maintained 85% of its initial photoresponsivity for as long as 20 d of storage in ambient air (figure 7(f)). Thus, this device showed excellent stability compared to other reported graphene/perovskite PDs.

The above discussed all the graphene/perovskite PDs consist of FET type architecture. Chen and co-workers recently reported graphene/CH$_3$NH$_3$PbI$_3$ perovskite/graphene (GPG) Schottky junction
high-performance hybrid PD [104]. Graphene films of area ~3 × 5 mm² were synthesized on the copper foil through the CVD process and then transferred to the SiO₂ (300 nm)/Si or flexible PET substrates. They fabricated three types of PD: (a) perovskite PD (P), (b) graphene/perovskite PD with vertical structure and without channel (GP), and (c) graphene/perovskite/graphene horizontal structure PD with one channel on the graphene (GPG). The schematic of the Schottky junctions of the three devices is shown in figure 7(g). A 20 μm wide channel on graphene converted the Schottky junction from vertical structure to vertical/horizontal hybrid structure in the GPG device (figure 7(g)). The GPG heterojunction device exhibited a very low dark current, which results in a high photoconductive gain. The I–V characteristics of the three devices are shown in figure 7(h), while the time-dependent photocurrents of the respective devices are shown in figure 7(i). The GPG detector showed a responsivity of 22 mA W⁻¹ under the illumination of 452 nm light at 2.1 mW cm⁻² with the corresponding photoconductive gain of 2.64 × 10³. Thus, the performance of the GPG Schottky junction PD was improved due to the low leakage current. They also demonstrated GPG PD on flexible PET substrate that showed robust bending durability.

Tang et al recently reported the spontaneous growth of CH₃NH₃PbBr₃ perovskite/nitrogen-doped reduced graphene oxide (N-rGO) hybrid structures using a facile solution-processable method and their application for PDs [105]. CH₃NH₃PbBr₃/N-rGO hybrid material was drop cast onto two neighboring ITO electrodes and fabricated PD, as illustrated in figure 7(j). The device shows the responsivity of as high as 3.6 × 10⁴ A W⁻¹ and LDR of 94.4 dB at 4 V of applied bias (figure 7(k)). The detection range of the phototransistor was 365 to 940 nm. The variation of responsivity of the device with a wavelength of incident light is shown in figure 7(l), while the inset of the figure shows the absorption for the CH₃NH₃PbBr₃/N-rGO hybrid. The photoresponsivity of the device matches well with the absorbance of active material. The EQE of the device was observed to be >10⁵% and detectivity up to 10¹⁵ Jones were measured in the optimal device with the weight ratio between perovskites and N-rGO to be 16:3. The rise and decay times of the device were reported to be 30 and 100 ms, respectively. The superior performance of the CH₃NH₃PbBr₃/N-rGO PDs is dominated by both intrinsic conductivity of the micro-crystals film and interfacial separation of photo-induced charge carriers. Chen and co-workers demonstrated all-inorganic CsPbBr₃ perovskite/multilayer graphene structure for optical-fiber-compatible PD, which showed a responsivity of 2 × 10⁴ A W⁻¹ [106].

Perovskite nanocrystals (NCs) and QDs show very interesting size-dependent optical and electronic properties that vary significantly from their bulk counterpart. Perovskite NCs and QDs also show excellent optoelectronic properties with the advantages of low-cost solution process synthesis, high PL quantum yield, higher photostability and widely tunable spectral range [107, 108]. Kwak et al reported hybrid CsPbBr₃₋ₓIₓ NCs/graphene photo FET [109]. Figure 8(a) depicts the schematic device configuration of the hybrid CsPbBr₃₋ₓIₓ NCs/graphene PD. The responsivity and detectivity of the hybrid device under 405 nm illumination with different optical power densities are shown in figure 8(b). This hybrid PD exhibits an impressive photodetectors responsivity of ~10⁸ A W⁻¹ and detectivity of ~10¹⁶ Jones under 405 nm illumination at 0.07 mW cm⁻². The wavelength-dependent photocurrent (black dot) of the graphene–CsPbBr₃₋ₓIₓ NCs PD and absorption spectra (blue line) of CsPbBr₃₋ₓIₓ NCs are shown in figure 8(c). The spectral dependence of photocurrent of the device matches well with the absorption spectra of the active material exhibiting the photocurrent peak at 580 nm (figure 8(c)) and good photocurrent up to 600 nm. The photocurrent growth and decay times of the PD were found to be 0.81 and 3.65 s, respectively (figure 8(d)). Note that the reported photodetectors responsivity is the highest among the inorganic perovskite/graphene-based hybrid PDs, though the response speed is relatively slow.

Pan et al demonstrated formamidinium lead bromide perovskite QDs (FAPbBr₃)–graphene hybrid phototransistor [110]. The schematic diagram of hybrid graphene–FAPbBr₃ QDs PD fabricated on SiO₂/Si substrate is presented in figure 8(e). The built-in electric field of the perovskite/graphene interface helps in the separation of the photogenerated carriers (figure 8(f)). The photoexcited electrons in perovskite QDs are transferred to graphene, leaving holes trapped in the QDs due to the built-in electric field. The wavelength-dependent photodetectors responsivity of the PD at V₈₅₂ = 2 V matches well with the absorption spectrum of the perovskite (figure 8(g)). A responsivity of 1.15 × 10⁵ A W⁻¹ and EQE of 3.42 × 10⁸% of the PD were found under an illumination power of 3 μW at 520 nm wavelength (figure 8(h)). The PD exhibited a photodetectors responsivity of 60 ms at 538 nm of the incident light.

Surendran et al reported all-inorganic CsPbBr₃I₃₋ₓ perovskite NCs/graphene hybrid phototransistor where NCs serve as a photo absorber and graphene act as a transport layer [111]. The schematic of the device configuration is shown in figure 8(i). Figure 8(j) illustrates the energy band diagram of the hybrid PD before and after the graphene–perovskite contact and photogenerated hole injection and p doping in the graphene layer. The PD exhibits maximum of responsivity of 1.13 × 10⁴ and 7.12 × 10³ A W⁻¹ at 80 μW cm⁻² for gate bias voltages VGS = −60 V and +60 V, respectively, while the corresponding detectivity was found to be
1.17 × 10^{11} \text{ Jones} and 6.16 × 10^{10} \text{ Jones}, respectively (figure 8(k)). The photocurrent growth and decay times of the phototransistor were found to be 273.6 ms and 2.26 s, respectively (figure 8(l)).

Cottam et al. recently demonstrated a defect-assisted high photoconductive gain in graphene all-inorganic cesium lead halide perovskite NCs phototransistors [112]. The device shows stable performance with photoresponsivity > 10^{6} \text{ A W}^{-1} in the UV–visible range.

Xia and co-workers recently reported a two-step CVD method for the fabrication of high-quality CH_{3}NH_{3}PbI_{3}/graphene van der Waals (vdWs) heterostructure for high-performance flexible PD and image sensor [113]. Inorganic PbI_{2} was deposited on top graphene layer by a CVD process and further CH_{3}NH_{3}I was used for fabrication of CH_{3}NH_{3}PbI_{3}/graphene heterostructure. The schematic of the device configuration of the flexible PET substrate is shown in figure 9(a). The photoresponsivity of the device at 633 nm for different light intensities is shown in figure 9(b). At 0.1 V, the PD exhibited photoresponsivity of \sim 2.7 \times 10^{3} \text{ A W}^{-1} with an illumination power of 100 nW (figure 9(b)). They further constructed an integrated flexible PD array using CH_{3}NH_{3}PbI_{3}/graphene heterostructure with (24 × 24) pixels inspired by the human visual system. The human eye has two basic functions: color and shape recognition. They used different illumination wavelengths using different filters to examine the color recognition capabilities of the as-prepared PD array. Figure 9(c) depicts the schematic illustration of the image sensor, which is imaged by using three filters of wavelengths 380, 633, and 750 nm under incident white light. The photocurrent was highest for 633 nm illumination and the smallest for the 750 nm illumination, showing great potential for application in artificial visions (figure 9(d)).

Though the above-mentioned perovskite/graphene hybrid PDs show very high responsivity with high photoconductive gain, an external power source is essential for their operation. Interestingly, the self-powered PDs that can work without an external power source has great application potential in next-generation devices that can operate wirelessly and independently are being studied extensively [115, 116]. Li et al. reported a self-powered TiO_{2}/graphene/CH_{3}NH_{3}PbI_{3} perovskite PD [114]. The graphene layer forms a Schottky junction with TiO_{2} that can efficiently separate and transport photogenerated excitons at the graphene/perovskite interface with superior device performance. The device configuration of the self-powered PD is shown in figure 9(e). The photoswitching behavior of the PD with and without bis(4-phenyl)(2,4,6-trimethylphenyl)amine (PTAA) HTL under 560 nm light with a power density of
40 mW cm\(^{-2}\) at 0 V (no bias) is shown in figure 9(f). The PD exhibits a photocurrent of 0.2 mA under self-powered mode. The \(I-V\) characteristics of the PD under solar simulator illumination at different light intensities are shown in figure 9(g). The self-powered device shows photocurrent rise time and fall time of \(\sim5\) ms (figure 9(h)). Thus, the self-powered device showed fast photoresponse as compared to the other reported perovskite/graphene-based phototransistor.

The 0D graphene QDs (GQDs) have a tunable bandgap due to the quantum confinement of carrier and edge effects [117, 118]. Interestingly, the structural, optical, and electrical properties of GQDs can be simply tuned by chemical doping for high-performance GQDs based optoelectronics [119]. Algadi et al demonstrated NGQDs/all-inorganic cesium lead bromide perovskite NCs heterostructure PD, which exhibits a responsivity of 0.24 A W\(^{-1}\), specific detectivity of 2.5 \times 10^{12}\ J one, and on/off ratio of 7.2 \times 10^4 [114]. Subramanian et al reported TiO\(_2\)/CH\(_3\)NH\(_3\)PbI\(_3\)/GQDs hybrid PD, which shows a photoresponsivity of 12 A W\(^{-1}\), and a detectivity up to 6.5 \times 10^{11}\ J one [120]. In this device, GQDs serve as HTL, while TiO\(_2\) serves as ETL which effectively increases the carrier separation in the perovskite layer. A summary of the performance parameters of the reported metal halide perovskite/graphene based PDs is presented in table 1. However, as the fabrication method, device configuration and device interfaces are different for different cases, so the comparison is not straightforward. It is apparent that the CH\(_3\)NH\(_3\)PbI\(_3-x\)Cl\(_x\) film/P3HT/graphene photo FET possesses the highest photoresponsivity and a quite high gain in the device.

2.2. Perovskite/TMDs photodetector

One of the major shortcomings of pure graphene in heterojunction PD and other optoelectronics is its zero bandgap and metallic nature. Thus, most of the reported graphene/perovskite PDs are photo–FET or Schottky junction type. On the other hand, TMDs such as MoS\(_2\), WS\(_2\), MoSe\(_2\), and WSe\(_2\) with a 2D semiconducting structure, are considered to be promising nanomaterials for future electronic and optoelectronic applications [3, 119, 121–131].

2.2.1. Perovskite/MoS\(_2\) photodetector

In 2016, Kang et al first reported a perovskite/MoS\(_2\) hybrid phototransistor in which CH\(_3\)NH\(_3\)PbI\(_3\) perovskite acted as a light-harvesting photoactive layer and mechanically exfoliated multilayer MoS\(_2\) was adopted as a charge transporting layer [132]. Additionally, they applied APTES surface treatment to SiO\(_2\)/Si, which resulted in the n-doping of MoS\(_2\) and suppressed the recombination rate of photogenerated carriers in the perovskite layer (figure 10(a)). The responsivity and detectivity of perovskite/MoS\(_2\)/APTES hybrid photo FET were observed to be 2.11 \times 10^{10}\ A W\(^{-1}\) and 1.38 \times 10^{10}\ J one, respectively (figure 10(b)). The responsivity of the photo FET decreases at 785 nm, which matches with the absorbance of the perovskite layer. The comparison of photocurrent growth and decay times of MoS\(_2\), MoS\(_2\)/APTES, and perovskite/MoS\(_2\)/APTES PDs are shown in figure 10(c). The perovskite/MoS\(_2\)/APTES device showed the fastest response of 6.17 and 4.5 s of rise and fall time, respectively. The improved photoresponse of the device with APTES was due to (a) the addition of APTES prevented the higher recombination rate of carriers by the interfacial traps at

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**Figure 9.** (a) Schematic illustration of CH\(_3\)NH\(_3\)PbI\(_3\)/graphene heterostructure-based flexible PD. (b) Photoresponsivity of the device under different applied voltages and illumination power. (c) Schematic diagram of the flexible image sensor using CH\(_3\)NH\(_3\)PbI\(_3\)/graphene heterostructure. (d) The corresponding output image under different light illumination (380, 633 and 750 nm of power 75 mW). Reprinted from [113]. Copyright (2020), with permission from Elsevier. (e) Schematic of the device configuration of perovskite/graphene/TiO\(_2\)/FTO self-powered PD. (f) Temporal photosresponse of the PD with and without PTAA operated with external bias of 1 V and without bias voltage. (g) Temporal response of the PD showing photocurrent rise and fall time of 5 ms. Reprinted with permission from [114]. Copyright (2017) American Chemical Society.
Table 1. Summary of performance parameters of reported metal halide perovskite/graphene PDs.

| Device structure                                      | Responsivity (A/W) | Detectivity (Jones) | Rise time/fall time (s) | Gain | EQE (%) | Reference |
|-------------------------------------------------------|--------------------|---------------------|-------------------------|------|---------|-----------|
| CH$_3$NH$_3$PbI$_3$ film/graphene photo FET            | 180                | $10^9$              | 0.087/0.54              | —    | $5 \times 10^4$ | [85]      |
| CH$_3$NH$_3$PbBr$_2$I islands/graphene photo FET      | $6.0 \times 10^5$  | —                   | 0.12/0.75               | 10$^9$ | —       | [86]      |
| CH$_3$NH$_3$PbI$_3$ nanowire/graphene photo FET       | $2.6 \times 10^6$  | —                   | 55/75                   | —    | —       | [87]      |
| CH$_3$NH$_3$PbI$_3$/graphene/Au NP photo FET          | $2 \times 10^7$    | —                   | 1.5/-                   | —    | $\sim 8 \times 10^3$ | [94]      |
| CH$_3$NH$_3$Pb$_{1-x}$Cl$_x$ film/P3HT/graphene photo FET | $4.3 \times 10^8$  | —                   | —                       | 8.9 $\times 10^9$ | —       | [95]      |
| CH$_3$NH$_3$PbI$_3$-PCBM BHJ film/graphene photo FET  | $8 \times 10^7$    | —                   | 92/6203                 | —    | —       | [96]      |
| 2D multiphase perovskite/graphene/(hBN) photo FET     | $\sim 10^7$        | $3.3 \times 10^{10}$ | <0.2                    | —    | —       | [20]      |
| CH$_3$NH$_3$PbI$_3$/graphene flexible photo FET       | 115                | $3 \times 10^{12}$  | —                       | —    | —       | [99]      |
| Graphene/CH$_3$NH$_3$PbI$_3$/graphene barristor-type photo FET | 950          | —                   | 0.022/0.037             | 2200 | —       | [100]     |
| Vapor deposited CH$_3$NH$_3$PbI$_3$ film/graphene photo FET | $1.73 \times 10^7$ | $2 \times 10^{15}$  | 0.879                   | $10^6$ | —       | [101]     |
| CH$_3$NH$_3$PbI$_3$–PbS QDs/graphene flexible photo FET | $2 \times 10^7$   | $5 \times 10^{12}$  | $\sim 0.5$-1            | $10^7$ | —       | [102]     |
| NGQDs–CH$_3$NH$_3$PbI$_3$/mrGO photo FET              | $1.92 \times 10^7$ | $2.71 \times 10^{13}$ | $0.013/0.010$           | $10^4$ | —       | [103]     |
| Graphene/CH$_3$NH$_3$PbI$_3$ perovskite/graphene Schottky junction PD | 0.022           | $3.55 \times 10^{9}$ | —                       | 2.64 $\times 10^3$ | —       | [104]     |
| CH$_3$NH$_3$PbI$_3$/N-rGO hybrid PD                   | $3.6 \times 10^4$  | $10^{12}$           | 0.030/0.10              | —    | $>10^5$ | [105]     |
| CsPbBr$_{3-x}$I$_x$ NCs/graphene photo FET            | $8.2 \times 10^4$  | $2.4 \times 10^{16}$ | 0.81/3.65               | —    | —       | [109]     |
| FAPbBr$_{3}$/graphene photo FET                        | $1.15 \times 10^3$ | —                   | 0.058/0.60              | 3.42 $\times 10^7$ | —       | [110]     |
| CsPbBr$_{3-x}$I$_x$ NCs/graphene photo FET            | $1.13 \times 10^4$ | $1.17 \times 10^{13}$ | 0.273/2.26              | $9.32 \times 10^9$ | —       | [111]     |
| CH$_3$NH$_3$PbI$_3$/graphene flexible PD              | $2.7 \times 10^3$  | —                   | —                       | —    | —       | [113]     |
| TiO$_2$/graphene/CH$_3$NH$_3$PbI$_3$ self-powered PD  | 0.375              | $\sim 10^{11}$     | <0.005                  | —    | —       | [114]     |
MoS$_2$/SiO$_2$ junction, and (b) the n-doping reduced the carrier recombination rate in the MoS$_2$. However, the reported PDs showed low responsivity and slow photoresponse speed compared to the other reported MoS$_2$/perovskite-based PDs, which may be due to defects and poorer morphology of perovskite film.

Wang and co-workers demonstrated solution-processed CH$_3$NH$_3$PbI$_3$/MoS$_2$ heterostructure PD using the semiconducting and metallic phase of MoS$_2$ [133]. MoS$_2$ can exist in two phases: (a) the trigonal prismatic (2H phase) crystal structure with semiconducting behavior, and (b) the octahedral (1T phase) crystal structures with metallic behavior. The 1T phase of MoS$_2$ is metastable and can be transformed into the thermodynamically stable 2H-MoS$_2$ phase by annealing. MoS$_2$ nanosheets (NSs) were fabricated by the chemical exfoliation method from bulk MoS$_2$. Perovskite/MoS$_2$ NS PD is illustrated in figure 10(d). Energy band diagrams of CH$_3$NH$_3$PbI$_3$/MoS$_2$ and the carrier transfer mechanisms in the metallic 1T and semiconducting 2H phase of MoS$_2$ are shown in figure 10(e). In the case of the 1T phase of MoS$_2$, both photogenerated electrons and holes can be injected from photoactive perovskite to the MoS$_2$ layer, leading to the recombination of charges in the layer (figure 10(e)). However, due to the higher current in the device with metallic 1T phase of MoS$_2$ compared to the PD with 2H phase, higher photoresponsivity with low on/off ratio was observed. In the case of the perovskite/2H-MoS$_2$ PD, photo-induced electrons and holes are injected from the perovskite into the conduction and valence bands of the 2H phase MoS$_2$, respectively (figure 10(e)), and the electrons and holes transfer to the electrodes due to the Schottky barrier at the contacts. The spectral responsivity and EQE of the devices with 1T-MoS$_2$ and 2H-MoS$_2$ are shown in figures 10(f) and (g). As compared to the PD with the 2H phase, PD with the 1T phase shows higher photoresponsivity due to the injection of a higher concentration of charges due to the metallic nature of 1T-phase. The PD with the 1T phase exhibits a responsivity of $3.09 \times 10^3$ A W$^{-1}$ and an EQE of $7.7 \times 10^3$%. The photoresponsivity decreased at $\sim$800 nm due to the decrease in the absorbance of perovskite active layer. Power-dependent responsivity and detectivity of the device with the 1T phase of MoS$_2$ under the illumination of the light of wavelength 500 nm is shown in figure 10(h). The device exhibits photoresponsivity of $3.3 \times 10^5$ A W$^{-1}$ at a power of 0.14 $\mu$W (figure 10(h)). The temporal photoresponse of the PD with 1T and 2H phase of MoS$_2$ are shown in figures 10(i) and (j), respectively. The rise and decay times were measured to be 0.45 and 0.75 s for PD with 1T phase (figure 10(i)), respectively, while the same for the 2H phase were observed to be $<25$ and 50 ms, respectively (figure 10(j)). It was believed that the higher defects in the 1T
phase of MoS$_2$ led to the trapping of photocarriers, resulting in slower response speed. However, the reported photoresponsivity was very high compared to other perovskite/MoS$_2$ based photo FETs. Peng et al demonstrated a solution-processed phototransistor utilizing CH$_3$NH$_3$PbI$_3$ perovskite–MoS$_2$ BHJ as the active material and a reduced graphene oxide (rGO) layer as the transporting layer as illustrated in figure 11(a) [134]. The energy band diagram of the hybrid system is illustrated in figure 11(b), showing the transfer process of photogenerated carriers. The photo-induced electrons in perovskite transferred to MoS$_2$ avoiding the radiative recombination, while holes transferred from perovskite to the rGO channel, which resulted in an increased photocurrent (figure 11(b)). Thus, with the introduction of MoS$_2$, the carrier separation and transport was significantly improved in the photoactive layer. The phototransistor with MoS$_2$ exhibits a responsivity of $1.08 \times 10^4$ A W$^{-1}$ at $V_{GS} = 0$ V and $V_{DS} = 2$ V, at an incident power density of 0.53 $\mu$W cm$^{-2}$, which is about five times higher than that of the device without MoS$_2$ (figure 11(c)). The specific detectivity of the hybrid device was calculated to be $4.28 \times 10^{13}$ Jones (figure 11(c)). Thus, these PDs exhibited higher detectivity than the other perovskite/MoS$_2$ based PDs.

Bai and co-workers fabricated a self-powered PD based on a CH$_3$NH$_3$PbI$_3$ perovskite/monolayer MoS$_2$ vertical type heterojunction [135]. They also investigated the performance of the device with a planar structure configuration. Monolayer MoS$_2$ was synthesized on SiO$_2$/Si substrates by the CVD method. Figure 11(d) presents a schematic illustration of CH$_3$NH$_3$PbI$_3$/MoS$_2$ planar structure PD in which the electrodes are bonded to two different materials. The schematic structure of CH$_3$NH$_3$PbI$_3$/MoS$_2$ vertical type heterojunction PD is shown in figure 11(e) in which one electrode is connected to MoS$_2$, and another electrode is connected to the CH$_3$NH$_3$PbI$_3$ film. The band alignment of planar and vertical PD under light illumination are shown in figures 11(f) and (g), respectively. In the case of planar structure device, under photon irradiation, electron–hole pairs are generated in the photoactive perovskite film, and these electrons are transferred to MoS$_2$ film, which increases the Fermi level of MoS$_2$ and reduces the Schottky barriers between Au electrode and material, causing the high photocurrent of the device (figure 11(f)). For vertical heterojunction devices, under illumination, the photogenerated electrons are transferred to the MoS$_2$ side, while holes are transferred to the perovskite side figure 11(g). Due to the suitable band alignment and built-in potential across the heterojunction, the vertical PD can operate without any external bias. The

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**Figure 11.** (a) Device structure of rGO/CH$_3$NH$_3$PbI$_3$–MoS$_2$ BHJ phototransistor. (b) Energy level diagram showing the carrier transfer process in the of rGO/CH$_3$NH$_3$PbI$_3$–MoS$_2$ BHJ hybrid structure. (c) Variation of responsivity and detectivity of the devices as a function of incident power at 660 nm in presence and absence of MoS$_2$ layer under $V_{GS} = 0$ V, $V_{DS} = 2$ V. [134] John Wiley & Sons. © 2018 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (d) Schematic illustration of planar type CH$_3$NH$_3$PbI$_3$/MoS$_2$ PD. (e) Schematic structure of the vertical type CH$_3$NH$_3$PbI$_3$/MoS$_2$ PD. Energy band diagram under the illumination of (f) planar PD (g) vertical PD. (h) Temporal photoresponse of self-powered vertical PD. [135] John Wiley & Sons. © 2018 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.
excitation intensity-dependent photocurrent and responsivity of the planar and vertical device are shown in figures 11(h) and (i), at a bias of 1 V, respectively. The planar and vertical type PD device exhibit a responsivity of 28 and 68.11 A W$^{-1}$, respectively, at illumination intensity of 0.7 mW cm$^{-2}$ and a voltage bias of 5 V. The photocurrent growth and decay times for the vertical self-powered device were found to be 2149 and 899 ms, respectively (figure 11(j)). However, the photoresponse speed of this device is relatively slow which may be due to the traps and defects.

Chandrasekar et al synthesized solution-processed CH$_3$NH$_3$PbI$_3$-MoS$_2$ nanohybrid for photodetection \[136\]. The interfacial carriers transfer between the 2D MoS$_2$ NS and perovskite cuboid heterojunction are utilized to enhance the device performance. The Au/CH$_3$NH$_3$PbI$_3$-MoS$_2$/Au PD exhibits responsivity of 696 mA W$^{-1}$ with the response speed of 50/16 ms (photocurrent growth/decay), while pristine Au/CH$_3$NH$_3$PbI$_3$/Au PD shows responsivity of 312 mA W$^{-1}$ with the response speed of 73/60 ms (photocurrent growth/decay). The device with MoS$_2$ also exhibits a very low dark current as compared to the pristine device.

He and co-workers fabricated lead-free (CH$_3$NH$_3$)$_3$Bi$_2$Br$_9$ perovskite/MoS$_2$ PD \[137\]. MoS$_2$ NSs were synthesized by scotch tape exfoliation method, while the perovskite layer was deposited by simple solution process technique. (CH$_3$NH$_3$)$_3$Bi$_2$Br$_9$-coated MoS$_2$ PD exhibit photoresponsivity of 112 A W$^{-1}$, detectivity of 3.8 $\times$ 10$^{12}$ Jones, and photoresponse time of 0.3 ms.

Wang et al reported self-powered CH$_3$NH$_3$PbI$_3$/2D BP/MoS$_2$ hybrid 2D photogate photodiode \[138\]. The schematic diagram of a 2D photogate photodiode is shown in figure 12(a). The electronic band structure of the heterojunction PD demonstrates type I and type II heterojunctions formation at the CH$_3$NH$_3$PbI$_3$/BP and BP/MoS$_2$ interfaces, respectively (figure 12(a)). Due to the lower exciton binding energy in perovskite materials, excitons can easily dissociate \[139\]. In the presence of light, the photocarriers are spontaneously generated in the CH$_3$NH$_3$PbI$_3$ perovskite film, which further diffuses into the underlying BP layer followed by carriers separation and collection due to the existence of a built-in electric field at the BP/MoS$_2$ junction.
(figure 12(b)). Thus, the observed photocurrent of the device is determined by the photoactive perovskite layer and photocarrier separation/transport at the BP/MoS$_2$ interface. The $I$–$V$ characteristics of the device show a typical diode-like rectifying behavior with a rectification ratio of 20 (figure 12(b)). In figure 12(c), the solid lines correspond to the dark current, while the dotted lines represent the photocurrent of the device. Due to the increase in electric field under reverse bias, the photocurrent was increased. The temporal photoresponse for CH$_3$NH$_3$PbI$_3$/BP/MoS$_2$ PD at $-2$ V is shown in figure 12(d). The photocurrent growth and decay times were observed to be 150 and 240 $\mu$s, respectively. The responsivity of the PD was calculated to be 11 A W$^{-1}$ under the illumination of 457 nm laser of intensity 0.02 mW cm$^{-2}$ and the highest photocurrent on–off ratio was reported to be $\sim 6 \times 10^4$ at 119 mW cm$^{-2}$ under 2 V reverse bias (figure 12(e)). Due to the built-in potential, the device can also operate without any external bias. The photocurrent switching performance at a different incident wavelength and intensity of 100 mW cm$^{-2}$ shown in figure 12(f) depicts the good photoswitching behavior of the self-powered PD. Thus, this report is significant for self-powered perovskite/MoS$_2$ based PDs.

Fang et al fabricated lead-free 2D Ruddlesden–Popper perovskites/MoS$_2$ vertical heterojunction PD [140]. They used few layers n-type MoS$_2$ and p-type (PEA)$_2$SnI$_4$ (where PEA = C$_6$H$_5$C$_2$H$_4$NH$_3$) 2D perovskite in the heterojunction PD, while graphene serves as electrodes for the device, as illustrated in figure 12(g). Due to the suitable band alignment and built-in potential, the heterojunction PD can operate without any external bias. The variation of responsivity and EQE of the heterojunction PD with incident light intensity at 451 nm under zero bias is shown in figure 12(h). The maximum responsivity and EQE were observed to be 121 mA W$^{-1}$ and 38.2% under the illumination of power 36 pW. They also reported the responsivity of 1100 A W$^{-1}$ at 3 V. The self-powered device shows photocurrent rise time and fall time of 34 and 38 ms, respectively (figure 12(i)). However, responsivity of the device was relatively low compared to the other reported results.

Sun et al reported perovskite PD based on triple cation mixed-halide perovskite layer and CVD grown atomically thin MoS$_2$ layer, as illustrated in figure 13(a) [141]. The MoS$_2$ layer was grown by the CVD process, while triple cation Cs$_{0.05}$(MA$_{0.17}$FA$_{0.83}$)$_{0.95}$Pb(I$_{0.83}$Br$_{0.17}$)$_3$ was deposited by simple solution process technique. The variation of responsivity and photocurrent of the device with incident light power at wavelength 520 nm and 2 V of applied bias is shown in figure 13(b). The PD exhibits a responsivity of 342 A W$^{-1}$ and detectivity of $1.14 \times 10^{12}$ Jones at incident power of 2.2 pW. The photocurrent rise and fall times were observed to be 27 and 21 ms, respectively (figure 13(c)). They also fabricated devices on flexible PET substrate, which showed stability up to 20000 bending cycles. These results are significant for the realization of flexible PDs.
Figure 14. (a) Schematic illustration of CsPbBr$_3$ NSs/MoS$_2$ hybrid PD. (b) Photogenerated charge transfer process in CsPbBr$_3$/MoS$_2$ heterojunction. (c) $I$–$V$ characteristics of the hybrid device in the dark and under 442 nm laser illumination with different intensities. (d) Comparison of photoresponsivity with the light intensity of pristine MoS$_2$, CsPbBr$_3$, and hybrid PD. (e) Wavelength-dependent responsivity of the hybrid device at 2, 5, and 10 V of applied bias. (f) Temporal photoresponse of the hybrid device showing rising and decay times. Reprinted with permission from [145]. Copyright (2018) American Chemical Society.

Wang et al. recently demonstrated (C$_6$H$_5$C$_2$H$_4$NH$_3$)$_2$PbI$_4$ 2D halide perovskite/MoS$_2$ hybrid PD with a very low dark current figure 13(d) [142]. Multilayer MoS$_2$ flakes were synthesized by the mechanical exfoliation method and transferred on the SiO$_2$/Si substrates. 2D hybrid perovskite was deposited on MoS$_2$ by solution process spin coating technique. The dark current of (PEA)$_2$PbI$_4$/MoS$_2$ PD was significantly suppressed compared to the pristine MoS$_2$ PD, which effectively increased the on/off ratio. Figure 13(e) shows the comparison of the on/off ratio of pristine MoS$_2$ PD and (PEA)$_2$PbI$_4$/MoS$_2$ PD for different intensity of light illumination. The on/off ratio of perovskite/MoS$_2$ PD was found to be $\sim 10^9$ (figure 13(e)). The spectral responsivity of the device at 0.06 nW is shown in figure 13(f). The hybrid PDs showed good responsivity in the wavelength range 200–650 nm. The device exhibits a specific detectivity of $1.06 \times 10^{11}$ Jones at 0.1 nW. Note that the ambient stability of 2D halide perovskite-based PDs is much higher than the 3D perovskite-based PDs, which is beneficial for their practical applications.

Compared to the organic–inorganic hybrid perovskite, all-inorganic perovskite NCs and QDs show superior ambient stability [143, 144]. Song et al. reported all-inorganic CsPbBr$_3$ NSs/2D MoS$_2$ heterostructure PD [145]. Monolayer MoS$_2$ was synthesized by a CVD process and then transferred onto SiO$_2$/Si substrates. The CsPbBr$_3$ NSs were then drop-cast on MoS$_2$ on Si/SiO$_2$, followed by annealing at 60 °C. The device configuration of CsPbBr$_3$ NSs/MoS$_2$ PD is shown in figure 14(a). The type-II heterojunction formation between CsPbBr$_3$ NSs and MoS$_2$ PD helps in better carrier separation suppressing the radiative recombination. Under light illumination, electron–hole pairs are generated in the photoactive perovskite NSs, and due to the strong driving force of the built-in electric field, these photogenerated electrons are transferred to the monolayer MoS$_2$, whereas the holes are confined in the perovskite NSs (figure 14(b)). This results in superior photodetection in the hybrid device. The $I$–$V$ characteristics of the hybrid PD as a function of the illumination intensity at 442 nm are shown in figure 14(c). Under the illumination, the current increases dramatically, and the on/off ratio was calculated to be more than $10^4$. The responsivity of the CsPbBr$_3$ NSs/MoS$_2$ PD device was increased nearly 20 or 1000 times compared with that of the CsPbBr$_3$ or bare MoS$_2$ PD at different intensities of incident light (figure 14(d)). The hybrid PD exhibits a photosresponsivity of 4.4 A W$^{-1}$, a detectivity of $2.5 \times 10^{10}$ Jones, and EQE of 302%. The hybrid PD showed good photosresponsivity up to 550 nm due to the absorption of CsPbBr$_3$. The spectral responsivity of the hybrid PD at different applied bias matches well with the absorbance of photoactive CsPbBr$_3$ NSs (figure 14(e)). The photocurrent rise and fall times of the hybrid PD were observed to be 0.72 and 1.01 ms, respectively (figure 14(f)). Overall, the photosresponsivity of the device was relatively low, which may be due to the traps at the perovskite/MoS$_2$ interface.

Noh et al. reported CsPbBr$_3$ QDs/monolayer MoS$_2$ phototransistor [146]. CsPbBr$_3$ QDs were synthesized by the hot injection method, while monolayer MoS$_2$ was grown by the CVD process [147]. They suggested
significant transfer of photogenerated electron form perovskite QDs to MoS$_2$ and n doping of MoS$_2$, which effectively increased the performance of the device. The hybrid photoFET exhibits a responsivity of $\sim 1$ A W$^{-1}$, which is nearly independent of the incident light intensity.

Lin and co-workers fabricated CsPbBr$_3$ QDs/few-layer MoS$_2$ phototransistor, which exhibited a high photoresponsivity of $10^4$ A W$^{-1}$ and a fast photoresponse speed of 8 ms [148].

Liu et al reported MoS$_2$ QDs encapsulated in a CH$_3$NH$_3$PbI$_3$ perovskite film for photo FET application, which showed a high detectivity of $5 \times 10^{11}$ Jones and a response speed of 25 $\mu$s [149].

Wu et al fabricated CsPbI$_{3-x}$Br$_x$ QDs/monolayer MoS$_2$ phototransistor [150]. Monolayer MoS$_2$ was grown by an oxygen-assisted CVD process and transferred onto SiO$_2$/Si substrate by PMMA-assisted transfer method, while mixed halide all-inorganic perovskite QDs were synthesized by hot injection method. The schematic illustration of the device is presented in figure 15(a), and the inset of the figure shows the TEM image of perovskite QDs. The favorable type II band alignment facilitating interfacial photogenerated charge separation and efficient carrier injection into the MoS$_2$ result in superior photodetection of the hybrid system. Under light illumination, electron–hole pairs were generated in the highly light-absorbing perovskite QDs layer and then separated by the built-in field produced at the interface of the heterojunction (figure 15(b)). The photogenerated electrons were transferred to the MoS$_2$ side, while holes moved to the QDs side, suppressing radiative recombination. The transfer characteristics of the phototransistor under 532 nm illumination with different optical powers is presented in figure 15(c). The photocurrent increases with illumination power. The photo FET exhibits a high responsivity of $7.7 \times 10^4$ A W$^{-1}$ and a specific detectivity of $5.6 \times 10^{11}$ Jones at a V$_G$ of 60 V (figure 15(d)). The photocurrent rise and fall times were observed to be 0.59 and 0.32 s, respectively, at V$_d = 0.1$ V (figure 15(e)). Though their device showed very high responsivity, the temporal response was relatively slow.

Zhang and co-workers tuned optoelectronic properties of all inorganic CsPbBr$_3$ perovskite NCs by regulating solution concentration and surface ligand content for perovskite NCs/MoS$_2$ hybrid PDs [151]. The device configuration is presented in figure 15(f). The band structure and photogenerated electron transfer from perovskite NCs to MoS$_2$ are presented in figure 15(g). The variation of EQE and detectivity of
the device with perovskite QDs concentration is shown in figure 15(h). With the increase of concentration of perovskite solution, the light absorption capability was effectively increased, which results in a more effective process of exciton dissociation and charge transport at the interface leading to an improved photocurrent response of the PD. The responsivity, EQE, and detectivity were observed to be $6.40 \times 10^5 \text{ mA W}^{-1}$, $1.50 \times 10^5 \%$, and $3.38 \times 10^{11} \text{ Jones}$, respectively. Perovskite QDs concentration–dependent photocurrent growth and decay times are presented in figure 15(i). Owing to the efficient separation and transport of the charge carriers at the interface, the response rate became faster in the hybrid system. For the synthesis of perovskite NCs/QDs, different surfactants, ligands such as oleic acid and oleylamine are used, which are beneficial for surface passivation and elimination of defects [152, 153]. However, an excessive amount of ligands blocks charge transfer across the interface by forming an insulating layer in a device. The author investigated the regulation effects of surface ligands with different purifying treatment cycles. With the increase of purifying cycles, the perovskite NCs may be degraded and aggregated, which results in poor device performance (figures 15(j) and (k)).

2D MoS$_2$ has also been used in different perovskite solar cells as a charge transport layer, which efficiently increases carrier separation and transport, leading to high PCE [154–157]. In a recent work, Ghosh et al demonstrated a monolayer MoS$_2$/CsPbBr$_3$ NCs type-II vertical heterojunction PD [158]. All inorganic CsPbBr$_3$ NCs were synthesized by room-temperature, two-step solid-state synthesis method. Large area monolayer MoS$_2$ film was grown directly on a SiO$_2$/Si substrate by the CVD method. The schematic of the vertical heterojunction PDs is shown in figure 16(a); the inset of the figure depicts the cross-sectional FESEM image of the heterojunction device. Based on the band alignment, MoS$_2$ and CsPbBr$_3$ form a type-II heterojunction. Under illumination, electron and hole pairs are generated in CsPbBr$_3$ NCs, and these electrons are transported to MoS$_2$. Thus, separation and transport of excitons suppressing radiative recombination in the perovskite lead to the high photocurrent of the device with fast photoresponse speed. The quenching of the PL intensity of perovskite in the heterostructure with MoS$_2$ device as compared to the pristine CsPbBr$_3$ NCs film provided evidence that indeed there is transfer and separation of photogenerated carriers from perovskite NCs to MoS$_2$ (figure 16(b)). TRPL spectra related to perovskite NPs also confirmed charge transfer, as shown in the inset of figure 16(b). Under the reverse bias, the photocurrent of the device was increased significantly, as shown in figure 16(c), which was attributed to the suitable band bending at the heterojunction enabling superior charge separation and transport. The comparison of spectral photoresponsivity of the heterojunction PDs and pristine MoS$_2$ PDs are shown in figure 16(d). The vertical heterojunction PD exhibit responsivity of $6.7 \text{ A W}^{-1}$ at 400 nm of incident light, which is significantly higher.
Figure 17. (a) Schematic of device structure of the hybrid CH$_3$NH$_3$PbI$_3$/WS$_2$ PD fabricated on a sapphire substrate. (b) The energy level of CH$_3$NH$_3$PbI$_3$/WS$_2$ heterostructure. (c) $I$–$V$ characteristics of the hybrid PD measured in dark and under illumination with white-light at different intensities. (d) Variation of photocurrent and responsivity with incident light intensity. (e) Temporal photoresponse of the device showing photocurrent growth and decay times. [159] John Wiley & Sons. © 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (f) $I$–$V$ curves of the CH$_3$NH$_3$PbI$_3$/WS$_2$ hybrid PD under dark and white-light. The inset shows the schematic diagram of CH$_3$NH$_3$PbI$_3$/WS$_2$ hybrid PD. (g) Variation of responsivity and detectivity of the device with illumination intensity. (h) Spectral responsivity and detectivity of CH$_3$NH$_3$PbI$_3$/WS$_2$ hybrid PD. (i) Temporal photoresponse of CH$_3$NH$_3$PbI$_3$/WS$_2$ and bare CH$_3$NH$_3$PbI$_3$ PD. Reprinted with permission from [162]. Copyright (2018) American Chemical Society.

than the pristine MoS$_2$ PD. The responsivity of the heterojunction PD as a function of incident light intensity is shown in the inset of figure 16(d). The variation of the photocurrent as a function of incident light intensity is shown in figure 16(e). The rise time and fall time of the heterojunction PD were observed to be 5.48 and 24.01 µs, respectively (figure 16(f)) which is significantly faster than the pristine MoS$_2$ PD. Thus, the high photoresponsivity and fast photoresponse speed were attributed to the superior charge separation and transport in the type–II heterojunction device. The detection range of the PD was 300–540 nm. The above conclusion was supported by the density functional calculation of the electronic structure of the pristine and hybrid systems.

2.2.2. Perovskite/WS$_2$ photodetector

Monolayer WS$_2$ having a higher direct bandgap than monolayer MoS$_2$, has receive major attention for various optoelectronic applications. In 2016, Ma et al first integrated monolayer WS$_2$ with perovskite to improve the photodetection performance in planar PDs [159]. Monolayer WS$_2$ was deposited by the CVD method on clean C-plane sapphire substrates, while CH$_3$NH$_3$PbI$_3$ perovskite films were deposited onto the monolayer WS$_2$ using sequential vapor deposition and vapor annealing method. A schematic device structure of the hybrid CH$_3$NH$_3$PbI$_3$/WS$_2$ planar PD fabricated on a sapphire substrate is presented in figure 17(a). The energy band structures of the CH$_3$NH$_3$PbI$_3$/WS$_2$ hybrid system is illustrated in figure 17(b). In the presence of light, electrons-hole pairs are generated in the photoactive perovskite layer, and due to the difference between their Fermi levels, these photogenerated charges are transferred to monolayer WS$_2$, which reduces the carrier recombination and enhances the photodetection performance [160, 161]. With the increase in the power of incident light, photocurrent increases and the symmetric $I$–$V$ curves of CH$_3$NH$_3$PbI$_3$/WS$_2$ PD indicate Ohmic–like contacts between the photoconductors and the electrodes.
In the hybrid device with WS$_2$, dark current was observed one order of magnitude lower than the pristine perovskite PD due to defects and interfacial carrier transfer lowering the charge concentration in the depletion region. The device exhibits a responsivity of 17 A W$^{-1}$ under a bias of 5 V and a light intensity of 0.2 $\mu$W cm$^{-2}$ (figure 17(d)). The maximum on/off of the device was observed to be $3 \times 10^5$. The rise and decay times of photocurrent were measured to be 2.7 and 7.5 ms, respectively (figure 17(e)).

Li et al. studied the light-matter interactions and mechanism of photogenerated carrier transfer from CH$_3$NH$_3$PbI$_3$ to atomically thin WS$_2$ by conductive and photoconductive atomic force microscopy [162]. They further fabricated CH$_3$NH$_3$PbI$_3$/WS$_2$ hybrid PD. Under illumination at 532 nm, they observed a significant increase in photocurrent in CH$_3$NH$_3$PbI$_3$/WS$_2$ heterostructure compared to the bare CH$_3$NH$_3$PbI$_3$ regions from the spatially resolved photocurrent images. The $I$–$V$ characteristics of CH$_3$NH$_3$PbI$_3$/WS$_2$ heterojunction PD show photovoltaic behavior with an open-circuit voltage of ~0.3 V and photocurrent was increased with the increase in illumination intensity (figure 17(f)). The PD exhibited a responsivity of 76.7 mA W$^{-1}$ and a detectivity of $3.40 \times 10^9$ Jones under illumination intensity of 0.09 mW cm$^{-2}$ and 2.5 V of applied bias (figure 17(g)). The incident light wavelength-dependent responsivity and detectivity match well with the absorbance of the photoactive perovskite layer (figure 17(h)). With the illumination of 405 nm, photosensitivity and detectivity of the PD were measured to be 3.9 mA W$^{-1}$ and 1.81 $\times$ 10$^9$ Jones, respectively. The photocurrent rise and decay times were found to be 13.5 and 18.7 ms, respectively, which is faster than the bare perovskite PD due to the efficient photogenerated carrier separation at the interface between WS$_2$ and perovskite (figure 17(i)). Thus, the photosensitivity of the device was quite low compared to the other reported devices.

Zhang et al. demonstrated phototransistors using CH$_3$NH$_3$PbI$_3$ perovskite-WS$_2$ NS composite as optical absorber and rGO as a channel layer [163]. The photo FET exhibits photosensitivity of 678.8 A W$^{-1}$, specific detectivity of 4.99 $\times$ 10$^{11}$ Jones and EQE value of 2.04 $\times$ 10$^{-2}$. The superior photodetection performance of the device is attributed to electron trapping by the WS$_2$ NSs resulting in photo-generated electron–hole separation and high carrier mobility of the rGO channel. Thus, the device showed very high photosensitivity compared to the other reported perovskite/WS$_2$ based PDs.

Erkılıc et al. reported perovskite/WS$_2$ heterostructure by an all vapor-phase growth approach for photodetection [164]. First, monolayer WS$_2$ was deposited on c-plane sapphire substrate by the CVD method. Next, the CH$_3$NH$_3$PbI$_3$ film was deposited on the monolayer WS$_2$/sapphire substrate by a two-step vapor phase method, in which first inorganic PbI$_2$ layer was deposited on the monolayer WS$_2$, followed by organic CH$_3$NH$_3$I intercalation, as illustrated in figure 18(a). Illustration of the CH$_3$NH$_3$I/WS$_2$ heterostructure photo FET is presented in figure 18(b). Due to the type-II heterojunction formation in CH$_3$NH$_3$I/WS$_2$ hybrid, photogenerated carriers were separated avoiding the radiative recombination, which led to enhanced photocurrent of the device. The $I$–$V$ curves of the PD in dark and under 532 nm laser illumination at various incident powers with gate voltage of 60 V are presented in figure 18(c). The PD shows good photoswitching behavior at $V_{DS} = 10$ V, $V_G = 30$ V and illumination intensity of 4.4 mW cm$^{-2}$ (figure 18(d)).

Xu et al. reported 1D single-crystalline CsPbBr$_3$ nanowires/mechanically exfoliated 2D WS$_2$ nanoflakes vdWs heterostructure planar PD [165]. Schematic illustration of CsPbBr$_3$/WS$_2$ hybrid PD is presented in figure 18(e), while the energy band structure of the heterojunction is shown in figure 18(f). Under photoexcitation, the light-induced carriers in CsPbBr$_3$ are transferred to WS$_2$, suppressing radiative recombination, which effectively increases the photodetection performances of the device. The PD exhibits responsivity of 57.2 A W$^{-1}$ and EQE of 157.9 at 2 V (figure 18(g)). Figure 18(h) shows the variation of photocurrent on/off ratio and detectivity of the device as a function of incident light power. The highest on/off ratio was observed to be $10^{8.83}$ and detectivity was calculated to be $1.36 \times 10^{11}$ Jones for CsPbBr$_3$/WS$_2$ PD. The hybrid PD shows good photoswitching behavior at an applied bias of 0.5 V and 76.3 nW of incident optical power (figure 18(i)). The photocurrent rise and decay time were observed to be ~2 ms (figure 18(i)). Due to the piezo-phototronic effect of CsPbBr$_3$ nanowire, they observed strain-gated photocurrent and responsivity of the hybrid device on a flexible PEN substrate. Note that this work reported the highest detectivity among the perovskite/WS$_2$ based PDs.

### 2.2.3. Perovskite/other TMDs photodetector

Although most of the reported perovskite/2D TMDs hybrid PDs are based on MoS$_2$ and WS$_2$, there are few reports on other TMDs (MoSe$_2$, WSe$_2$, PtSe$_2$, PdSe$_2$) based hybrid perovskite PDs. Lu and co-workers reported CH$_3$NH$_3$PbI$_3$/monolayer WSe$_2$ hybrid PD [166]. Monolayer WSe$_2$ flakes were grown on transparent sapphire substrates by the CVD method. The device architecture of CH$_3$NH$_3$PbI$_3$/monolayer WSe$_2$ hybrid PD is illustrated in figure 19(a). From DFT based electronic structure calculation, it was shown that monolayer WSe$_2$ and CH$_3$NH$_3$PbI$_3$ form a type-II heterojunction which facilitates superior photocarrier separation (figure 19(b)). The wavelength-dependent responsivity of the device is shown in figure 19(c),
Figure 18. (a) Schematic of CH$_3$NH$_3$PbI$_3$/WS$_2$ heterostructure synthesis by all vapor-phase growth approach. (b) Schematic illustration of CH$_3$NH$_3$PbI$_3$/WS$_2$ PD. (c) I–V curves of the heterostructure PD in dark and under illumination with different intensities. (d) Temporal photoresponse of the device. Reprinted with permission from [164]. Copyright (2019) American Chemical Society. (e) Schematic illustration of CsPbBr$_3$ nanowire/WS$_2$ planar PD. (f) Band structure of CsPbBr$_3$/WS$_2$ illustrating transfer of photogenerated carriers from CsPbBr$_3$ to WS$_2$. (g) Variation of responsivity and EQE with incident power. (h) On/off ratio and detectivity as a function of incident power. (i) Time-dependent photocurrent of the device at an applied bias of 0.5 V under incident power of 76.3 nW. (j) Temporal response of photocurrent showing photocurrent rise and decay times. Reprinted from [165], Copyright (2019), with permission from Elsevier.

which matches with the UV–visible absorption spectrum of the light-absorbing perovskite layer. The maximum responsivity, detectivity, and EQE were reported to be $1.1 \times 10^2$ A W$^{-1}$, $2.2 \times 10^{11}$ Jones, and $2.5 \times 10^4\%$, respectively. Thus, the performance of the device was quite good.

Hassan et al chemically synthesized MoSe$_2$–CsPbBr$_3$ nanohybrids using 4-aminothiophenol bifunctional ligand for photodetection [167]. The schematic of device architecture of MoSe$_2$–CsPbBr$_3$ nanohybrids PD is presented in figure 19(d). Under light illumination, electron and hole pairs are generated in perovskite NCs, and due to type-II band alignment, these electrons are transferred to MoSe$_2$ flakes suppressing the radiative recombination (figure 19(e)). Among the bare MoSe$_2$ NSs, CsPbBr$_3$ NCs and MoSe$_2$–CsPbBr$_3$ the hybrid PD shows the best performance with the highest photocurrent due to enhanced carriers separation and transport (figure 19(f)).

Newly discovered PtSe$_2$, a promising member of TMDs, has been theoretically expected to have high carrier mobility and tunable bandgap [168–170]. However, the multilayer 1T-phase PtSe$_2$ exhibits semi-metallic properties with a broad light absorption spectrum [171]. Zhang et al fabricated self-powered Cs-doped FAPbI$_3$ perovskite/multilayered PtSe$_2$ vertical heterojunction broadband PD, as illustrated in figure 20(a) [172]. Multilayer semimetallic PtSe$_2$ film was synthesized by selenization of sputter deposited Pt film. To fabricate the heterojunction PD, FA$_{0.85}$Cs$_{0.15}$PbI$_3$ perovskite precursor solution was spin-coated on PtSe$_2$ film. Due to the built-in electric field, photogenerated electrons and holes near the heterojunction interface were rapidly separated, which eventually increased the photocurrent. The device exhibits a responsivity of 117.7 mA W$^{-1}$, EQE of 14.9% and maximum on/off ratio of $5.7 \times 10^3$ at zero bias (figure 20(b)). The heterojunction device shows a broadband photoresponse in the UV–NIR region due to the strong light absorption of the perovskite/PtSe$_2$ hybrid system in a broad wavelength region (figure 20(c)). The photocurrent rise and fall times were measured to be 78 and 60 ns, respectively (figure 20(d)).

Similar to PtSe$_2$, PdSe$_2$ is a newly explored promising TMD for optoelectronic applications [174–176]. Zeng and co-workers reported self-powered Cs-doped FAPbI$_3$ perovskite/PdSe$_2$ Schottky heterojunction broadband vertical PD, as illustrated in figure 20(e) [173]. The responsivity and detectivity of the heterojunction PD were observed to be 313 mA W$^{-1}$ and $2.72 \times 10^{13}$ Jones under illumination intensity of 35.1 $\mu$W cm$^{-2}$ at 0 V, respectively (figure 20(f)). The PD exhibits good photodetection capability over the entire UV-NIR region (figure 20(g)). The photocurrent growth and decay times were observed to be 3.5 and
Figure 19. (a) Schematic illustration of CH$_3$NH$_3$PbI$_3$/WSe$_2$ heterostructure PD. (b) Band structure of CH$_3$NH$_3$PbI$_3$/WSe$_2$ heterostructure. (c) Wavelength dependent photoresponsivity of the device. [166] John Wiley & Sons. © 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (d) Schematic illustration of MoSe$_2$–CsPbBr$_3$ nanohybrids PD. (e) Energy band diagram MoSe$_2$–CsPbBr$_3$ heterostructure. (f) I–V characteristics of bare CsPbBr$_3$ NCs and MoSe$_2$ NSs and MoSe$_2$–CsPbBr$_3$ nanohybrids PD. Reprinted with permission from [167]. Copyright (2020) American Chemical Society.

Figure 20. (a) Schematic illustration of the perovskite/PtSe$_2$ heterojunction PD. (b) Responsivity and EQE of the PD as a function of incident light intensity. (c) Spectral responsivity and detectivity of the heterojunction PD. (d) Temporal photoresponse showing rise and fall times of the PD. Reprinted with permission from [172]. Copyright (2018) American Chemical Society. (e) Schematic illustration of FA$_{0.85}$Cs$_{0.15}$PbI$_3$/PdSe$_2$ heterojunction PD. (f) Variation of responsivity and detectivity of the heterojunction PD with incident light intensity. (g) Wavelength dependent EQE of the device. (h) Temporal photoresponse showing rise and fall times. Reproduced from [173]. CC BY 4.0.

4 µs, respectively (figure 20(h)). Note that the photoresponsivity and detectivity of perovskite/PdSe$_2$ based PDs are superior to that of the perovskite/PtSe$_2$ based PDs.

A summary of the comparative performance parameters of the perovskite/TMDs based PDs is presented in table 2. It may be observed that the TMD based heterostructure PDs have relatively lower responsivity than the graphene based PDs.

2.3. Perovskite/black phosphorous photodetector
Recently 2D layered BP have been extensively studied for their application in PDs due to their unique optoelectronic properties with high carrier mobility [177–179]. Zou et al reported Schottky barrier-controlled mixed halide CH$_3$NH$_3$PbI$_3$–xCl$_x$ perovskite/few-layer BP phototransistor [180]. Few-layer BP flakes were synthesized by mechanical exfoliation from bulk BP crystals and further transferred to the highly doped SiO$_2$/Si substrates. Mixed halide CH$_3$NH$_3$PbI$_3$–xCl$_x$ precursor solution was spin-coated on top
Table 2. Summary of the performance parameters of metal halide perovskite/TMD based PDs.

| Device structure                                      | Responsivity (A/W) | Detectivity (Jones) | Rise time/fall time (s) | EQE (%) | Reference |
|-------------------------------------------------------|--------------------|---------------------|-------------------------|---------|-----------|
| CH$_3$NH$_3$PbI$_3$/MoS$_2$/APTES photo FET           | 2.11 × 10$^4$      | 1.38 × 10$^{10}$    | 6.17/4.5                | —       | [132]     |
| CH$_3$NH$_3$PbI$_3$/MoS$_2$ heterostructure PD        | 3.3 × 10$^5$       | 7 × 10$^{11}$       | <0.025/0.05             | —       | [133]     |
| CH$_3$NH$_3$PbI$_3$-MoS$_2$ BHJ/rGO                  | 1.08 × 10$^4$      | 4.28 × 10$^{13}$    | <45 × 10$^{-3}$         | 2.0 × 10$^6$ | [134]     |
| CH$_3$NH$_3$PbI$_3$/MoS$_2$ heterojunction PD         | 68.11               | —                   | 2.15/0.9                | —       | [135]     |
| CH$_3$NH$_3$PbI$_3$/2D BP/MoS$_2$ self-powered photodiode | 11                  | 1.3 × 10$^{12}$     | 1.50/2.40 × 10$^{-4}$   | 80      | [139]     |
| RP perovskite/MoS$_2$ self-powered PD                 | 0.121               | 8.09 × 10$^9$       | 0.034/0.038             | 38.2    | [140]     |
| Triple cation perovskites/MoS$_2$ flexible PD         | 342                 | 1.14 × 10$^{12}$    | 0.027/0.021             | —       | [141]     |
| 2D perovskite/MoS$_2$ hybrid PD                       | 16.8                | 1.06 × 10$^{13}$    | 0.006/0.004             | 3.3 × 10$^3$ | [142]     |
| CsPbBr$_3$ nanosheets/2D MoS$_2$ PD                   | 4.4                 | 2.5 ×10$^{10}$      | 7.2/10.1 × 10$^{-4}$    | 302     | [145]     |
| CsPbBr$_3$ QDs/MoS$_2$ photo FET                      | 1.02                | —                   | —                       | —       | [146]     |
| CsPbBr$_3$ QDs/MoS$_2$ photo FET                      | 4.68 × 10$^4$      | —                   | 0.0075/0.0080           | 1.4 × 10$^3$ | [148]     |
| CH$_3$NH$_3$PbI$_3$/MoS$_2$ QDs photo FET            | 920                 | 5 × 10$^{11}$       | 15/25 × 10$^{-6}$       | —       | [149]     |
| CsPbI$_3$–Br$_x$ QDs/MoS$_2$ photo FET               | 7.7 × 10$^4$       | 5.6 ×10$^{12}$      | 0.59/0.32               | 10$^0$  | [150]     |
| CsPbBr$_3$ NCs/MoS$_2$ hybrid PD                      | 6.40 × 10$^5$      | 3.38 × 10$^{11}$    | 0.092/-                 | 1.50 × 10$^5$ | [151]     |
| CsPbBr$_3$ NCs/MoS$_2$ vertical heterojunction PD    | 24.3                | 3×10$^{12}$         | 5.48/24.01 × 10$^{-6}$  | 7.5 × 10$^4$ | [158]     |
| CH$_3$NH$_3$PbI$_3$/WS$_2$ hybrid PD                  | 17                  | 2 × 10$^{12}$       | 2.77/7.5 × 10$^{-3}$    | —       | [159]     |
| CH$_3$NH$_3$PbI$_3$/WS$_2$ hybrid PD                  | 0.076               | 3.40 × 10$^{9}$     | 13.5/18.7 × 10$^{-3}$   | —       | [162]     |
| CH$_3$NH$_3$PbI$_3$–WS$_2$ nanocomposite/rGO photo FET | 678.8               | 4.99 × 10$^{11}$    | <0.060/0.780            | 2.04 × 10$^5$ | [163]     |
| CH$_3$NH$_3$PbI$_3$/WS$_2$ photo FET                  | 43.6                | —                   | —                       | —       | [164]     |
| CsPbBr$_3$ nanowire/WS$_2$ hybrid PD                  | 57.2                | 1.36 × 10$^{14}$    | 2 × 10$^{-3}$/-         | 157.9   | [165]     |
| CH$_3$NH$_3$PbI$_3$/WC$_2$ hybrid PD                  | 1.1 × 10$^2$       | 2.2 × 10$^{13}$     | —                       | 2.5 × 10$^6$ | [166]     |
| Cs-doped FAPbI$_3$/PbSe$_2$ self-powered PD           | 0.118               | 2.91 × 10$^{12}$    | 7.8/6.0 × 10$^{-8}$     | 14.9    | [172]     |
| Cs-doped FAPbI$_3$/PdSe$_2$ self-powered PD           | 0.313               | 2.72 × 10$^{13}$    | 3.5/4 × 10$^{-6}$       | ~60     | [173]     |
of the BP layer to fabricate the photo FET. Using a low work function Al metal as the source electrode, a Schottky barrier was introduced in the photo FET between the source electrode and the perovskite-modified BP channel, as illustrated in figure 21(a). They tuned the photoresponse speed of the PD by changing the drain voltage, which was attributed to a field-assisted de-trapping process of electrons in the perovskite layer close to the Schottky barrier. The device exhibits a high responsivity of $1.9 \times 10^8$ A W$^{-1}$ under operational voltages of $V_{gs} = 30$ V and $V_{ds} = -2$ V (figure 21(b)). The responsivity of the device was observed to be $5.2 \times 10^5$ A W$^{-1}$ at 895 nm of the incident light. The photoresponse in the NIR region beyond the absorption edge of the perovskite was observed which is attributed to the excitation of carriers from the valence band to the trap states within the perovskite bandgap. The temporal photoresponse of the device at an operational voltage of $V_{gs} = 30$ V and $V_{ds} = -2$ V is presented in figure 21(c). The photocurrent rise time and decay time were measured to be 8 and 17 ms, respectively. The normalized photoresponsivity of the PD in the broad wavelength region from UV to NIR is presented in figure 21(d).

Chen and co-workers synthesized all inorganic CsPbBr$_3$ NCs/BP NSs heterostructures (figure 21(e)) via a in situ hot-injection method for photodetection [181]. Due to the strong bonding between CsPbBr$_3$ NCs and BP NSs, photogenerated carriers were separated and transferred efficiently, which yielded good the performance of the device. The device architecture is presented in figure 21(f), while figure 21(g) shows $I–V$ characteristics of the device at different illumination intensity. Reproduced from [181] with permission of The Royal Society of Chemistry.

2.4. Perovskite/MXene photodetector

A new class of 2D transition metal carbides, carbonitrides and nitrides, named MXenes, has emerged as a potential candidate for optoelectronics [182–184]. MXenes serve various roles in PDs, such as photo absorbers, transparent electrodes, Schottky contacts, and plasmonic materials [182]. Deng and co-workers reported CsPbBr$_3$ NSs/conductive Ti$_3$C$_2$T$_x$ MXene flexible PD for photo communication, as illustrated in figure 22(a) [185]. Under illumination, electron and hole pairs were generated in the perovskite layer and transferred to MXene electrodes (figure 22(b)). The temporal photoresponse of the PD with different thicknesses of the MXene electrode under 450 nm light illumination at a bias of 10 V is presented in figure 22(c). The device exhibits photoresponsivity of 44.9 mA W$^{-1}$, detectivity of $6.4 \times 10^8$ Jones, and a maximum on/off ratio of $2.3 \times 10^3$. Photocurrent growth and decay times were measured to be 48 and 18 ms, respectively (figure 22(d)). The PD maintains the excellent flexibility and stability after 1500 bending cycles.

Pan et al reported visible light and x-ray PD based on CsPbBr$_3$ NCs/Ti$_3$C$_2$T$_x$ MXenes nanocomposite [186]. 2D MXene NSs were synthesized through the conventional acid etching and delamination of Ti$_3$AlC$_2$. Perovskite NCs were grown on MXene NSs. Figure 22(e) depicts the TEM image of the nanocomposite. The temporal photoresponse of the PD is shown in figure 22(f), while the inset of the figure shows the schematic of the device. However, the on/off ratio and responsivity of the device were very low.
3. Conclusions and outlook

Herein, we have systematically reviewed the recent developments in metal halide perovskite/2D materials hybrid PDs. Due to their unique optical properties, high carrier mobility, and superior mechanical flexibility, atomically thin 2D materials have been widely investigated in PDs, which show a very fast photoresponse speed along with high responsivity. However, the performances of these 2D material based PD are restricted by the low light-harvesting capability of monolayer and few-layer material. On the other hand, halide perovskite has an excellent capability of light-harvesting with a large absorption coefficient. Thus, the integration of 2D TMD materials with high light-harvesting perovskite materials could lead to considerable enhancement in the light absorption and result in high responsivity along with fast photoresponse due to high carrier mobility in 2D TMDs. In the reported PDs, halide perovskites serve as a light-harvesting active layer, while ultra-thin 2D materials serve various roles, such as charge transport layer/channel, Schottky contacts, photo absorbers, etc. Among the reported perovskite/graphene PDs, most of the reports are on perovskite/graphene-based photo transistors and Schottky junctions PDs due to the metallic behavior of graphene with zero bandgap. In this type of photo FET, perovskite absorbs the incident light, and graphene acts as a carrier transporting channel material. These photo FET working on the strong photogating effect can exhibit very high photoresponsivity ($10^9$ A W$^{-1}$). Zero-dimensional perovskite QDs/NCs-graphene-based photo FETs also have been reported. Compared to organic–inorganic hybrid perovskite, all-inorganic perovskite NCs/QDs shows enhanced ambient stability. All inorganic perovskite NCs/graphene photo FET exhibits a responsivity as high as $8.2 \times 10^5$ A W$^{-1}$ and detectivity of $2.4 \times 10^{16}$ Jones. 2D perovskite/graphene photo FET and photo FET on flexible substrates are also reported, which shows excellent stability and flexible durability, respectively. TiO$_2$/graphene/perovskite self-powered PD can work without any applied bias due to the built-in potential across the heterojunction. The photoresponsivity of 2D perovskite/MoS$_2$ self-powered hybrid PD was reported of $0.12$ A W$^{-1}$. Among the reported perovskite/2D TMDs based PDs, most of the reports are on perovskite/MoS$_2$ photo FET and type-II heterojunction PDs. The maximum responsivity of perovskite/MoS$_2$ photo FET has achieved $10^4$ A W$^{-1}$, while the maximum responsivity was observed about $10^{13}$ Jones. Perovskite film, QDs or NCs/monolayer MoS$_2$ type-II heterostructure PDs can detect light without external bias due to the built-in potential across the heterojunction. The photoresponsivity of 2D perovskite/MoS$_2$ self-powered hybrid PD was reported of $0.12$ A W$^{-1}$. CsPbBr$_3$ NCs/monolayer MoS$_2$ type-II vertical heterojunction PD showed very fast photoresponse of few $\mu$s. WS$_2$ and WSe$_2$ based perovskite PDs exhibit maximum photoresponsivity of 679 and $1.1 \times 10^2$ A W$^{-1}$, respectively. The maximum specific detectivity of WS$_2$ and WSe$_2$ based perovskite PDs was observed about $10^{14}$ and $10^{11}$ Jones, respectively. Newly discovered PdSe$_2$ and PtSe$_2$ are a promising
member of the TMDs family with semi-metallic properties. Perovskite–PdSe₂ and PtSe₂ based self-powered hybrid PDs show very fast photoresponse ($\sim 10^{-8}$ s). Other 2D materials such as BP, MXene–perovskite PDs have also been reported with inferior performance, and thus needs further exploration.

Despite the remarkable progress in this field of perovskite/2D material PD, there are many unresolved issues and challenges that need to be addressed, and there are plenty of room for further improvement towards commercial applications of perovskite/2D material based devices. Few major issues related to perovskite optoelectronic devices are: (a) poor ambient stability in the presence of oxygen and water molecules, (b) ion migration resulting poor storage stability of the devices (c) toxicity of lead, (d) reproducible methods of device fabrication. Though perovskite optoelectronic devices show excellent performances, perovskite-based devices usually degrade when they are exposed to thermal-stress, light, electric fields, oxygen, or moisture. Compared to organic–inorganic hybrid perovskite, all inorganic and 2D halide perovskite show enhanced ambient stability. Thus, future studies would be required for stability improvement. The thermal energy and local polarization can generate fast and slow ion migration in perovskite semiconductors. By grain boundary passivation, fast ion migration in perovskite such as the migration of iodide defects can be suppressed, whereas the slow ion migration is still an important issue that hinders the low-term stability of the device. Further investigation would be required to gain deeper insights into degradation and ion migration in metal halide perovskites/2D material based PDs. There have been some successful strategies to improve the stability of solar cells. However, such studies are lacking for perovskite based PDs. The toxicity of lead on humans and the environment has been a challenge for the development and commercialization of perovskite-based PDs. However, lead-free perovskite (including tin, copper, and germanium etc based) possesses poor ambient and storage stability compared to lead-based perovskites. Thus, further investigation is required on lead-free perovskite/2D material based PDs. Among the reported perovskite/2D material PDs, in most of the reports, perovskite films/nanostructures were fabricated by solution-processable technique. However, reproducibility and large scale fabrication for commercial applications are the major issues by this technique. Thus, alternative fabrication method of perovskite films/nanostructures should be explored.

The perovskite 2D material interface plays an important role in device performance. The trap states present at the interface can lead to poorer photodetection performance. The passivation of trap steps by different physical and chemical methods can substantially enhance the performance of the devices. For synthesis of perovskite nanostructures, different surfactants are used. Residue of this surfactant can result in slow photoresponse speed. Thus, investigation of alternative synthesis and processing method of high quality perovskite nanostructures should be required. The performance of the perovskite/2D material PDs can also be further improved by the engineering of device structure and interface layers. The integration of perovskite with different newer 2D materials should be explored for photodetection. The spectral photoresponsivity of perovskite/2D material PDs are limited to UV–visible region. However, development of NIR PDs using perovskite/2D material heterostructure is very important for different technological interests. The spectral response range of PDs can be extended by utilizing mixed-halide perovskite materials with smaller optical bandgap and by combining up-conversion materials with perovskite materials. Thus, further investigation would be required on perovskite/2D material PDs for NIR photodetection. Flexible PDs own extensive research attention in the field of wearable and portable devices. Further investigation is required on perovskite/2D material flexible PD. Plasmonic nanostructures at perovskite/2D material interface can help in superior light-harvesting of the PDs. Further studies are required on the fabrication of perovskite/2D material vertical heterojunction self-powered PDs.

There has been a lot of recent interest in the synthesis of 2D perovskites for various optoelectronic applications. The term 2D refers to the intrinsic 2D crystal structures of Ruddlesden–Popper perovskites or perovskite NSs and nanoplates with ultralow thickness. Recent studies have demonstrated the feasibility of constructing heterostructures of 2D perovskites with other 2D-layered materials [187, 188]. It would be interesting to explore the integration of 2D perovskites with other inorganic or organic semiconductors and functional oxide perovskites for PD applications [189]. Single crystal perovskite nanostructures shows interesting polaron physics and exciton–photon coupling that could be explored for heterostructure PD applications. Thus, perovskite/2D material based hybrid PDs are just emerging and require more knowledge on properties, device optimization for different technological interest. We expect that this review will stimulate more dedicated investigations of this emerging field for commercialization and practical applications of the hybrid devices.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).
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