



Review

Device Architecture for Visible and Near-Infrared Photodetectors Based on Two-Dimensional SnSe₂ and MoS₂: A Review

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Abstract: While band gap and absorption coefficients are intrinsic properties of a material and determine its spectral range, response time is mainly controlled by the architecture of the device and electron/hole mobility. Further, 2D-layered materials such as transition metal dichalcogenides (TMDCs) possess inherent and intriguing properties such as a layer-dependent band gap and are envisaged as alternative materials to replace conventional silicon (Si) and indium gallium arsenide (InGaAs) infrared photodetectors. The most researched 2D material is graphene with a response time between 50 and 100 ps and a responsivity of <10 mA/W across all wavelengths. Conventional Si photodiodes have a response time of about 50 ps with maximum responsivity of about 500 mA/W at 880 nm. Although the responsivity of TMDCs can reach beyond 10⁴ A/W, response times fall short by 3–6 orders of magnitude compared to graphene, commercial Si, and InGaAs photodiodes. Slow response times limit their application in devices requiring high frequency. Here, we highlight some of the recent developments made with visible and near-infrared photodetectors based on two dimensional SnSe₂ and MoS₂ materials and their performance with the main emphasis on the role played by the mobility of the constituency semiconductors to response/recovery times associated with the hetero-structures.

Keywords: device architecture; photodetectors; response speed; mobility; SnSe₂; MoS₂; heterostructures; graphene

1. Introduction

Photodetectors form vital components of many electrical and opto-electronic devices as they facilitate the conversion of light into an electric signal that can be processed by standard read-out electronics. They have found various applications such as in spectroscopy, broad-range infrared detection for night vision, fiber-optic communication, visible light detection for digital camera and video imaging and x-rays for biomedical imaging, etc. [1–4]. High response time photodetectors find applications mainly in telecommunication and have been aggressively pursued. The response time is mainly controlled by the charge carrier mobility of the semiconductor, electrode distance in the case of linear devices, and the depletion width in the case of non-linear devices, as illustrated from Figure 1a,b.

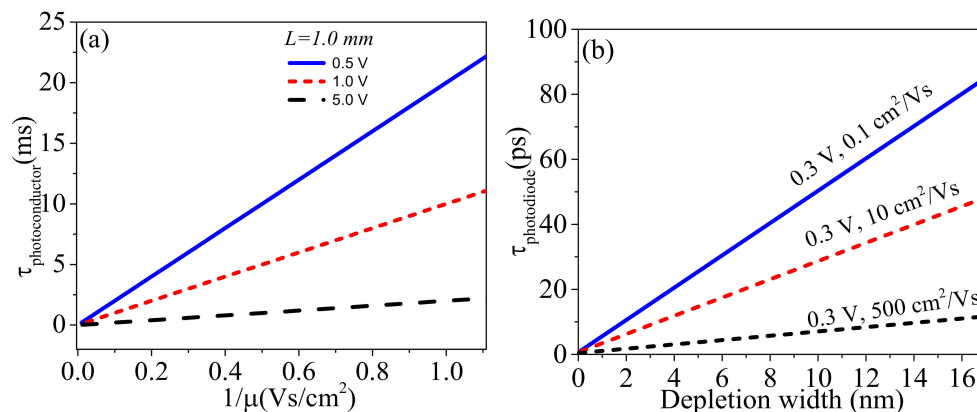


Figure 1. (a) Dependence of transit time on inverse of mobility for a photoconductor at constant electrode spacing $L = 1.00 \text{ mm}$ and different bias, (b) Dependence of transit time on depletion width for a photodiode at constant bias and different mobilities. The transit time strongly depends on mobility and bias for a photoconductor where as it strongly depends on the width of the depletion region and mobility for a photodiode. (a,b) are reproduced with permission from reference [5].

In the past, different kinds of photo-detector devices with different operating principles have been pursued. These include photoconductors [6], photodiodes [7], photoelectrochemical devices [8], phototransistors [9,10], and others which have been integrated with piezoelectric nano-generators for self-powered photodetection, which requires piezo-electric materials such as ZnO, GaN, and InN [11–14]. Conventional Si and InGaAs photodiodes have a response time of about 50 ps [6] and a responsivity of about 500 mA/W at maximum wavelength of 880 nm for Si photodiode and 1.2 A/W for InGaAs at 1550 nm [6]. However, they suffer from non-transparency, non-flexibility, limited spectral range, and relatively high manufacturing costs for InGaAs [1,15], although they can be applied for micron-scale imaging devices. Moreover, 2D layered semiconductors are a class of emerging materials with appealing properties such as transparency (at atomic level), strong light-matter interaction, good flexibility, and readiness in processing as well as in cost [6]. Graphene, the most studied 2D layered material composed of a sheet of carbon atoms just one atomic layer thick bonded together in a hexagonal honey comb lattice, was first isolated in 2004 [16]. Following the discovery, a number of its intriguing electronic, mechanical, optical and thermal properties have been studied [17]. The zero band gap and semi-metallic nature has allowed it to interact with light over a broad bandwidth from infrared (IR) to ultraviolet (UV) wavelengths and rendered it a promising material for various photodetectors over a wide spectral range. The response time of graphene can reach up to 50–100 ps [6,18], mainly due to its ballistic mobility of $2.5 \times 10^5 \text{ cm}^2/\text{Vs}$ [17]. However, its transparency allows it to absorb only $\approx 2.3\%$ of incident visible and IR light resulting into a reported responsivity of $<10 \text{ mA/W}$ which is undesirable for high performance photodetectors and its gapless nature leads to short photo-carrier lifetime which is unfavorable for efficient photocurrent generation [19–27]. Although attempts have been made to improve the responsivity by introducing electron trapping centers and band-structure engineering, such as building a graphene quantum dot-like structure, this has resulted in a low response time compared to the pristine one [1,28]. Other efforts of incorporating other materials like MoS₂/Graphene and Silicon/Graphene have similarly resulted in improved responsivity of 45.5 A/W at wavelength 642 nm and 85 mA/W at 1.55 μm respectively [29,30]. Transitional metal dichalogenides (TMDCs) are layered materials with properties that are band-gap dependent. These TMDCs have a general formulae MX_2 ($M = \text{W, Mo, Sn, Ga, In, \dots}$ etc.) and $X = (\text{Se and S})$ and include MoS₂ [31–38], WS₂ [33,39–41], MoSe₂ [38,42,43], WSe₂ [33,39–41,44], InSe [26,45–47], GaSe [48,49], and In₂Se₃ [50] among others, while SnSe₂ [49] is a layered material made of earth abundant Sn metal. These materials can absorb a wide range of photon energy from 0.3 (for BP) to 2.5 eV [51]. Despite the enormous strides made by researchers in the development of layered materials for opto-electronic nano-device applications with reported responsivity of $>10^4 \text{ A/W}$ [1,52–54], the response time does not compete with those of

the conventional photodetectors fabricated from graphene, Si, and InGaAs photodetectors. In this review, we present the recent developments with a case study of two dimensional SnSe₂ and MoS₂ and their related hetero-structures as photodetectors with the main emphasis on the role of mobility, electrode spacing, and depletion width to response/recovery time. Photodetectors have been developed on both flexible and non-flexible substrates. The response time is equally affected by choice of the substrate as substrates have different carrier density and mobilities. The article is organized as follows. We first introduce the mechanisms of photodetection and define the figures of merit for a photodetector. Then devices based on SnSe₂ and its related hetero-structures on glass, ITO, SiO₂/Si, and Si substrates will be discussed, followed by MoS₂ devices and their related heterostructures on Si/SiO₂, sapphire, GaN, GaAs, and p-/n-Si substrates. Finally, we conclude by suggesting methods of designing fast response time photodetector devices taking into consideration the overall performance of the device.

2. Photodetector Sensing Mechanisms

Photodetection devices rely on various sensing mechanisms and the devices are named accordingly. These mechanisms include photoconductive effect, photo-gating effect, photovoltaic effect, photo-thermoelectric effect, photo-bolometric effect, photo-electrochemical effect and piezo-phototronic effect [6,55–58]. We briefly discuss these mechanisms as follows.

2.1. Photoconductive and Photo-Gating Effect

The process of photoconduction involves generation of excess free charge carriers by a semiconductor absorbing photons with energy higher than the band gap which eventually results in reduction of its electrical resistance. A photoconductor in its basic design is shown in Figure 2a and consists of a semiconductor with two Ohmic metal contacts at opposite ends.

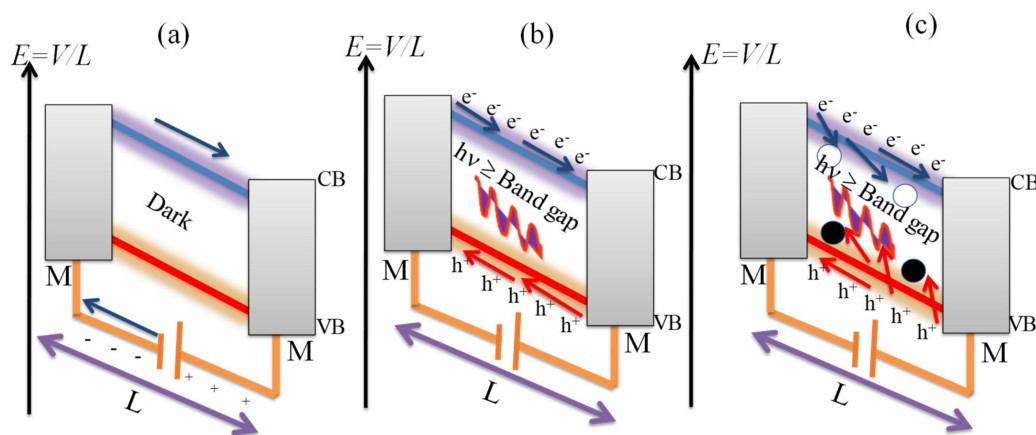


Figure 2. Schematic of a photoconductor between two metal contacts (M) (a) without illumination and (b) with illumination, (c) Schematic illustrating a photo-gating effect between a semiconductor and two Ohmic metal contacts (M) under illumination. The open and closed circles are defect states that can capture a hole or an electron eventually modulating the resistance of the semiconductor. (a,b) are adapted with permission from reference [6].

When the device is illuminated with photons of energy greater than the band gap, as shown in Figure 2b, electron-hole pairs are generated (excitons) and are separated by an applied bias. For a linear (Ohmic) photoconductor based on a metal-semiconductor-metal structure, the transit time (considered as the charge lifetime from generation until recombination or extraction), a measure of the response time of the photodetector is defined by a mathematical relation below [5,6,59];

$$\tau_{\text{transit}} = \frac{L}{\mu_{\text{drift}} \cdot E} \quad (1)$$

where μ is the mobility, L is the electrode spacing, and E is the applied field separating the free carriers. The consequence of this equation is the response time of a photoconductor highly depends on the carrier mobility of the semiconductor and electrode spacing. To achieve a short transit time requires that we use small electrode spacing and a high electric field as depicted in Figure 1a. The photoconductive gain defined as the ratio of the free photo-carrier lifetime to the transit time ($G = \tau_{\text{photocarriers}}/\tau_{\text{transit}}$) has the general expression as $G = (\tau_{\text{transit}} \times \mu \times V)/L^2$. Here, one type of carrier, e.g., a hole is usually captured in a trap state with lifetime $\tau_{\text{photocarriers}}$, while the other type of carrier is free to traverse the channel with transit time of τ_{transit} . Large gain results in when $\tau_{\text{photocarriers}} > \tau_{\text{transit}}$ [6]. Photogating effect is an example of photoconducting effect and is due to existence of a certain amount of localized or trapped states such as defects, impurities, or surface states within the band gap of a semiconductor as illustrated in Figure 2c. These trap states can capture either the photogenerated holes or electrons and localize them and eventually act as local gates that modulate the resistance of the semiconductor [52]. This effect is common in low dimensional systems, such as TMDCs and colloidal quantum dots, which possess a large surface-to-volume ratio and reduced screening effect [4,52].

2.2. Photo-Electrochemical Effect

The mechanism of photocurrent switching in a photo-electrochemical device is illustrated in Figure 3a,b and depends on various parameters which involve the redox properties of the semiconductor, availability of donors and acceptors in the electrolyte, applied potentials and energy of incident photons [60]. Upon photoexcitation of the semiconductor with energy greater than the band gap, a photocurrent is generated. By considering an electrode covered with the n-type semiconductor, the photo-generated electrons from valence band to conduction band can be transferred to the electrode if its potential is higher than the potential of trapped electrons. In the presence of electron donors and acceptors such as H^+ and $(\text{OH})^-$, interfacial electron transfer between the semiconductor and the electrolyte solution occurs. Anodic photocurrents require that an electron donor is easily oxidized by photo-generated holes and that the electrode potential enables electron transfer from the conduction band of the semiconductor to the electrode as illustrated in Figure 3a. Cathodic photocurrents occur when reduction of the electron acceptor by electrons from the conduction band and holes is more efficient than the mechanisms responsible for anodic photocurrent generation as illustrated in Figure 3b. The evolution of anodic photocurrents follows the kinetics presented in Figure 3c. The kinetics of cathodic photocurrent evolution is presented in Figure 3d [60–63].

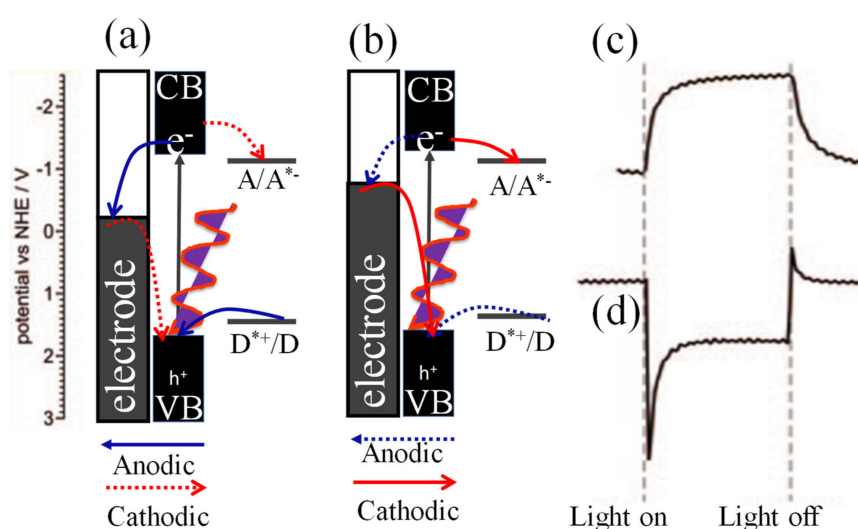


Figure 3. Mechanisms of anodic (a) and cathodic (b) photocurrent generation at the electrode covered with an n-semiconductor. Kinetics of anodic (c) and cathodic (d) photocurrent evolution. A/A^{*-} and D^{*+}/D are acceptor and donor levels. (a–c) are adapted with permission from reference [60].

2.3. Photovoltaic Effect

This can be categorized into two:

2.3.1. Metal-Semiconductor Configuration (Schottky/Rectifying Metal Contacts)

Here, a Schottky junction is a junction formed between a semiconductor and one of the metal electrodes [55] with a large barrier height and low doping concentration less than the density of states in the conduction or valence band [59]. The condition for the formation of a rectifying contact is based on the work function of the metal (ϕ_m) and semiconductor (ϕ_s) and depends on whether the semiconductor is n-type or p-type. The work function is defined as the energy difference between the Fermi and vacuum levels [59]. The potential barrier between the metal and the semiconductor is illustrated on the energy band diagram of Figure 4a,b and is the difference between the metal work function ϕ_m and the semiconductor electron affinity χ , and is given by

$$q \cdot \phi_B = q (\phi_m - \chi). \tag{2}$$

The electron affinity is defined as the energy difference between the conduction band edge and the vacuum level in the semiconductor [59]. For the case of an ideal contact between a metal and a p-type semiconductor, the barrier height (ϕ_B) is given by

$$q \cdot \phi_B = qE_g - q (\phi_m - \chi) \tag{3}$$

where E_g is the band gap of the semiconductor. For n-type semiconductor to form a Schottky diode, $\phi_m > \chi$. Similarly, $\phi_m < \chi$ for p-type semiconductor [57]. As an example, Figure 4c illustrates the possible metals with which bulk n-SnSe₂ forms Ohmic and Schottky diode. The work function of bulk SnSe₂ is between 5.0 and 5.3 eV [64,65].

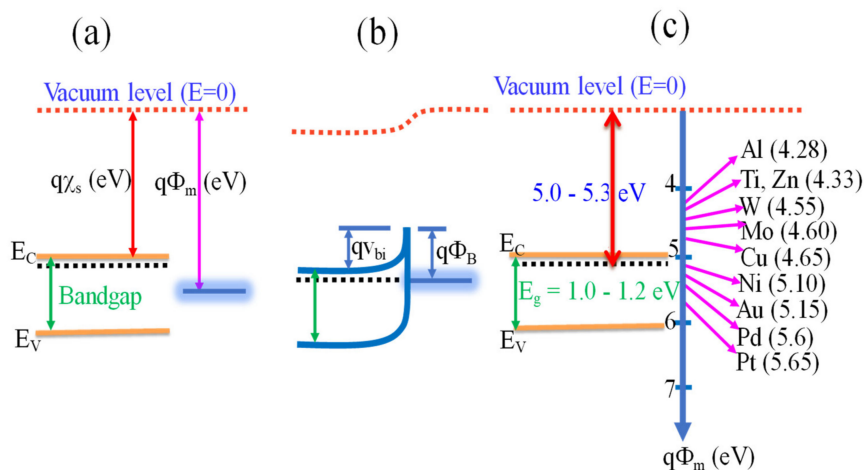


Figure 4. (a,b) Energy band diagram of isolated metal adjacent to isolated n-type semiconductor before equilibrium and metal-semiconductor in thermal equilibrium and (c) is possible metals with which SnSe₂ can form Schottky and Ohmic metal contacts.

2.3.2. Semiconductor-Semiconductor Configuration (p-n, n⁺-n⁺⁺, or p⁺-p⁺⁺ Junctions)

This effect is based on either a p-n, n-n⁺, or p-p⁺ junctions formed between either a p- and p⁺-type, n- and n⁺-type or a p- and n-type semiconductor. Here, p refers to a doped semiconductor where the majority of charge carriers are holes, n is where a majority of charge carriers are electrons while n⁺ and p⁺ have relatively larger carrier concentrations compared to n and p doped semiconductors. As an example, the formation of a p-n junction is illustrated in Figure 5a,b and is based on p-Si and n-SnSe₂. When the device is illuminated with photons of energy greater than the band gap, electron/hole

pairs are generated. The junction leads to charge carrier separation after the excitation process and electrons and holes drift in opposite directions towards the electrodes driven by the built-in electric field at the interface. The built-in electric field is normally produced at the depleted semiconductor region (Junction) where there is a significant difference in the work functions between the two materials. A photodiode displays rectifying current-voltage characteristics in the dark. The photodiode can function at two modes under illumination; i.e., photovoltaic (zero bias) and photoconductive mode (reverse bias). In photovoltaic mode, the photo-generated electron-hole pairs are separated by the built-in electric field and collected at opposite electrodes, which generates a short-circuit current (I_{SC}). The electrical output can be open-circuit voltage (V_{OC}). A photodiode working in photovoltaic mode has the lowest dark current leading to an improved detectivity and sensitivity. The magnitude of the reverse current increases when the device is illuminated. This is because photo-excited carriers are swept in opposite directions by the built-in electric field. The photovoltaic mode can also be used to convert the energy of the photons to electrical power (solar cell). In photoconductive mode, the external electric field is in the same direction as the built-in electric field which increases the separation efficiency of the electron-hole pairs and the response time. The charge carrier transit time for a p–n or Schottky junction depends on the width of the depletion region as well as charge carrier mobility, as depicted in Figure 1b, and is defined as [5,59]

$$\tau_{\text{transit}} = \frac{W}{v_{\text{drift}}} = \frac{W}{[\mu_{\text{drift}} E_0]}, \quad (4)$$

where

$$W = \sqrt{\frac{\varepsilon(V_{bi} - V_a)[N_a + N_d]}{2\pi e N_a N_d}} \quad (5)$$

is the width of the depletion region, V_{bi} , V_a , N_a , N_d , μ_{drift} , and E_0 are built-in potential, applied potential, concentration of acceptor atoms, concentration of donor atoms, electron-hole drift mobility and built-in electric field, respectively. The consequence of Equation (4) is that the transit time highly depends on the depletion width which is in the order of few nm and electron/hole mobility and as a result, the transit time is much faster for p–n or Schottky junction. We point out that over a certain value of the electric field, the drift velocity saturates. High frequency/speed operations require the depletion region to be thin to reduce transit time but on the other hand, to increase the responsivity or quantum efficiency the depletion layer must be sufficiently thick in order to allow a large fraction of the incident light to be absorbed. Thus, there is a trade-off between the response time and responsivity/quantum efficiency of a photodetector [59].

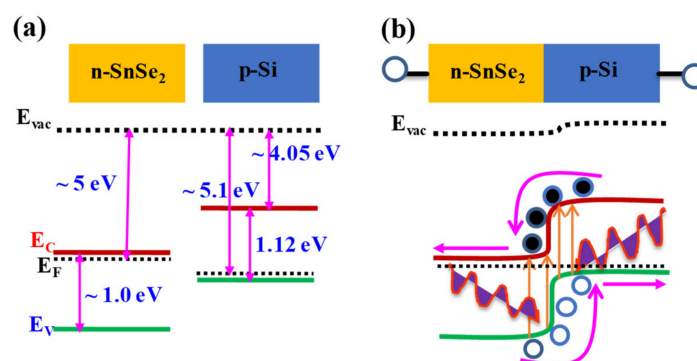


Figure 5. (a,b) A Energy band diagram of n-SnSe₂ and p-Si before at equilibrium. At equilibrium, the Fermi levels of n- and p-side line up as shown by the dashed lines. The absorption of a photon with energy $h\nu \geq$ band gap will generate electron-hole pairs. The electron-hole pairs are then separated and accelerated by the built-in electric field at the junction. (a,b) are reproduced with permission from reference [66].

2.4. Photo-Thermoelectric and Photo-Bolometric Effects

Photo-thermoelectric effect refers to the generation of a temperature gradient ΔT from charge carriers across a semiconductor channel upon photoexcitation as illustrated in Figure 6a. The temperature gradient ΔT is then converted into a photo-voltage difference ΔV_{PTE} called the Seebeck effect. The magnitude of ΔV_{PTE} is determined from $\Delta V_{\text{PTE}} = \Delta T(S_1 - S_2)$ where $S_{1,2}$ are Seebeck coefficients for metal and semiconductor respectively [6,52,67]. The heat gradient mainly stems from a localized illumination with a focused laser spot. The bolometric effect refers to the change in the resistance of a material induced by heating under uniform illumination as shown in Figure 6b [4,6]. The magnitude of this effect is associated with the conductance change of photosensitive materials with temperature ($dG/(dT)$) and the homogeneous temperature increase (ΔT) caused by laser heating [52,68]. A bolometer detects the incident photon power (dP) and measures the changes in temperature (ΔT). The key sensing parameters are the thermal resistance $R_t = dT/dP$ and the heat capacity C_h , which determines its response time, defined as $\tau = R_t \cdot C_h$ [69]. The change in conductance is mainly influenced by change in carrier mobility due to the associated temperature change and or change in the number of carriers contributing to the current [69].

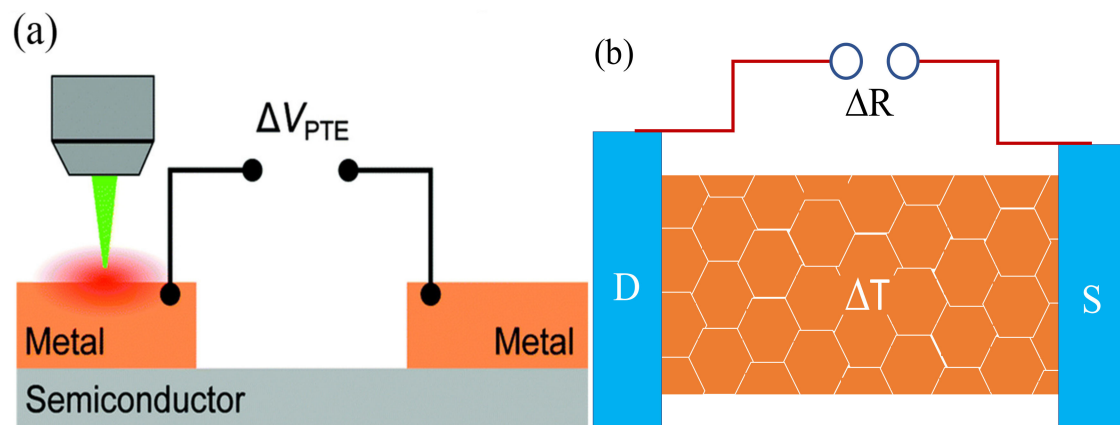


Figure 6. Photo-thermoelectric effect; Schematic of a semiconductor locally illuminated by a focused laser spot on one of the metal contacts to the semiconducting channel and (b) Schematic illustrating photo-bolometric effect. (a) Reproduced with permission from reference [6]. (b) Adapted with permission from reference [69].

2.5. Piezo-Phototronic Effect

Piezo-phototronic is a general term that refers to devices that use piezo-potential for controlling the carrier generation, transport, separation and/or recombination for improving the performance of opto-electronic devices [55]. It requires piezo-electric materials such as ZnO, GaN, and InN, that generate an electrical potential upon variations of applied pressure/stress [11–14,55–57].

The mechanism of operation of a piezotronic device is based on the fundamental concepts of the conventional Schottky contact and p–n junctions in semiconductor physics. The major difference is the presence of ionic charges introduced by piezoelectric polarization which can tune the carrier transport at the interface. The effect of piezopotential on metal-semiconductor contact under compressive and tensile strain is illustrated in Figure 7a,b. When a metal and an n-type piezoelectric semiconductor forms a Schottky contact (work function of metal is appreciably greater than electron affinity of the n-semiconductor) under compressive strain, as shown in Figure 7a, the negative piezoelectric polarization charges and the negative piezo potential induced at the semiconductor side can repel the electrons away from the interface, resulting in a further depleted interface and an increased local Schottky barrier height (SBH). If the piezoelectric semiconductor is under tensile strain as shown in Figure 7b, the positive piezoelectric polarization charges and the positive piezo potential created at the semiconductor side near the interface can attract the electrons toward the interface,

resulting in a less depleted interface and hence a decreased local SBH. The electron-hole pairs generated through photon excitation increases conductivity and reduces Schottky barrier height due to charge redistribution [55–57].

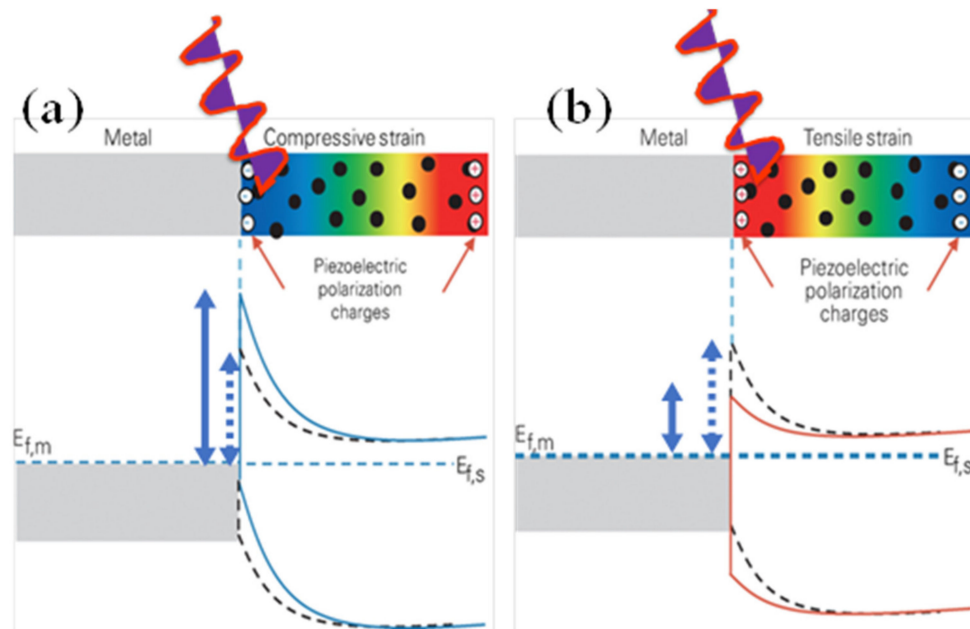


Figure 7. Schematic of energy diagram illustrating the effect of piezopotential on metal-semiconductor interface under illumination with photons of energy greater than the band gap; (a) Under compressive strain (b) Under tensile strain. The dotted and full double arrows indicate the changes in the SBH before and after the applied stress (piezopotential). The black dots represent the free-charge carriers in the bulk semiconductor. (a,b) are adapted with permission from reference [56].

3. Figures of Merit for Photodetectors

In this section, we define the general meaning to each figure of merit.

3.1. Responsivity (R)

The responsivity of a photodetector is defined as the ratio of the output photocurrent or photovoltage to the input optical power on the active region of the device. It is an indication of the achievable electrical signal under certain illumination power. A large responsivity indicates a large electrical output signal for a defined optical excitation power. It is usually expressed as $R = I/(P \times A)$, where A is the effective surface area, P is the power density, and $I = I_{\text{illumination}} - I_{\text{dark}}$ is the photocurrent. It is measured in A/W [4,70].

3.2. External Quantum Efficiency (EQE)

The external quantum efficiency (EQE) is the ratio of the number of electron-hole pairs with contribution to the photocurrent, n_e to the total number of incident photons n_{photons} . It can be expressed as $EQE = n_e/n_{\text{photons}} = R_\lambda hc/e\lambda$, where e is the elementary charge, h is Planck's constant, c is the speed of incident light and λ is the wavelength of incident light. The EQE is the measure of the optical gain G in the photodetector. $EQE > 1$ means, more than one charge carrier per impinging photon is measured. To achieve a large EQE in a photodetector, the optical absorption of the active layer should be high, while the carrier recombination and trapping before being collected should be minimized [4,10,23].

3.3. Response/Recovery Time

The response/recovery time of a photodetector is usually measured between 10% (90%) to 90% (10%) of the generated signal under modulated excitation intensity, either on the rising or falling edge. A photodetector with a small response time is usually desired for certain applications, like video-rate imaging and optical communication [6,27].

3.4. Noise Equivalent Power (NEP):

This is the minimum detectable optical power at which the electrical signal-to-noise ratio (SNR) in the detector is equal to unity, when bandwidth is limited to 1 Hz. NEP describes the sensitivity of a detector and is defined as the ratio of noise current to responsivity, $NEP = \frac{I_{noise}}{R}$ [1,4].

3.5. Detectivity (D^*)

The detectivity D^* is a useful parameter for comparing the detection performance of photodetectors with different materials and geometries. A higher detectivity indicates a better photodetector performance. It is defined as the reciprocal of the noise equivalent power (NEP), i.e., the minimum optical power which can be detected by the photodiodes, i.e.,

$$D^* = \frac{\sqrt{AB}}{NEP} = \frac{R\sqrt{AB}}{S_n} = \frac{R\sqrt{A}}{S_n} \quad (6)$$

where A is the device area and B is its bandwidth and S_n is the noise spectral density [1,4].

Photodetector devices require both flexible and non-flexible substrates. The most available and preferred substrates in the development of photodetectors are glass, p- and n-Si, Si/SiO₂ (thin insulating layer between 280 and 300 nm thick), Al₂O₃, GaN and GaAs. In the following section, we review photodetectors based on these substrates and the influence of mobility on response/recovery times of SnSe₂, MoS₂, and their related heterostructures.

4. Performance of Photodetectors Based on SnSe₂

Band gap, absorption coefficient, mobility, and device architectures play important roles in photon absorption, responsivity, charge carrier transport, and separation. SnSe₂ is n-type semiconductor with carrier concentration between 10¹⁷–10¹⁹ cm⁻³ [71]. The band gap varies between 0.9–2.04 eV [70–74], absorption coefficient of >10⁴ cm⁻¹, and mobility between 0.6–85 cm²/Vs [3,71,72,75–80]. The highest mobility of 85 cm²/Vs was extracted from an exfoliated SnSe₂ field effect transistor [80]. Most of the reported mobility is <10 cm²/Vs. Based on these parameters and SnSe₂ being a layered material whose band gap depends on the number of layers coupled with elements that are earth abundant, it can effectively absorb photons of energy ≥1 eV suitable for various device applications. However, the low mobility may hinder its applications in devices that require high frequency operations. In the following section, we discuss various attempts made by different groups in the development of SnSe₂ and its related heterostructures in photodetector applications.

Recently, our group have developed SnSe₂ thin films on soda lime glass substrate and tuned the band gap for IR photodetection [71]. The device was illuminated with a 1064 nm wavelength, as shown in Figure 8a, and a responsivity of ~2 mA/W, and an estimated response/recovery time ~7.76 and 2.5 s at a bias of 5 and 10 V, respectively, were reported (Figure 8b) [71]. It is interesting to note that the response time is in accordance with Equation (1) for a photoconductor. We further fabricated a hetero-structure based on SnSe₂ and PEDOT:PSS on soda lime glass substrate (Figure 8c) in order to utilize the built-in potential and narrow depletion width for fast response/recovery speed [5]. The response/recovery time improved to 1.33 and 1.22 s respectively with the device operating at zero-bias (Figure 8d) but the improve in response time resulted into a reduction in responsivity to 1.4–2.6 μA/W [5]. The fact that PEDOT:PSS and SnSe₂ suffer from low interfacial mobility, the response/recovery time was further improved to 57 ± 25/34 ± 15 μs by fabricating a hetero-structure based on n-SnSe₂/p-Si substrate

(Figure 8e,f) and a responsivity of 120 mA/W was obtained [66]. These studies clearly demonstrate the role played by mobility/device structure in the response/recovery speeds of a photodetector. From a mobility point of view, SnSe₂ may not compete favorably with conventional semiconductors, but its attractive layer dependent optical properties make it a viable candidate to be intergrated with conventional Si technology for various device applications.

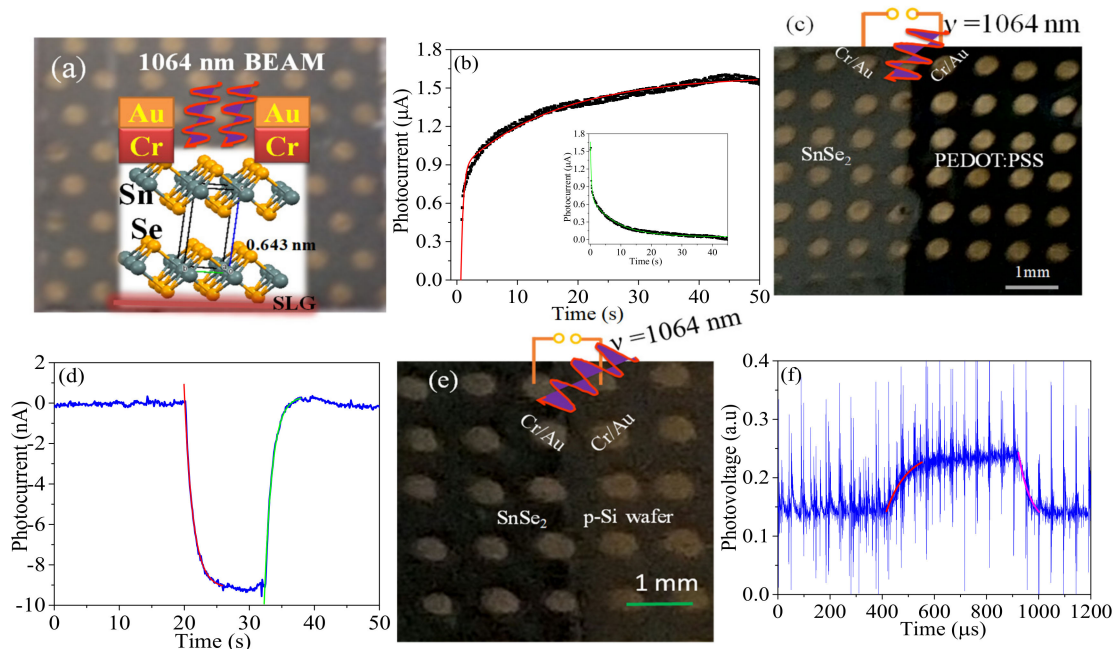


Figure 8. (a) Optical image of SnSe₂ on soda lime glass (SLG) substrate illuminated with 1064 nm wavelength, (b) Response/recovery bi-exponential fitted, (c) Optical image of SnSe₂/PEDOT:PSS hetero-structure on SLG substrate, (d) Single exponential fitted response/recovery speeds, (e) Optical image of SnSe₂/p-Si hetero-structure and (f) Single exponential response/recovery speed of SnSe₂/p-Si photodiode. (a,b) are reproduced with permission from reference [71], (c,d) are reproduced with permission from reference [5], (e,f) are reproduced with permission from reference [66].

A bilayer SnSe₂ was mechanically exfoliated from bulk single crystals onto SiO₂/Si substrate for photodetector and field effect transistor applications [81]. Their electrical analysis revealed a mobility of 4 cm²/Vs and an on/off ratio of 10³ at room temperature and the dark state. As shown in Figure 9a,b, the device was irradiated with a 633 nm laser beam and a fast response/recovery speed of 2.1 ± 0.3/3.2 ± 0.2 ms respectively with a responsivity of ~0.5 A/W were reported. The bilayer SnSe₂ device had a slower response time and higher responsivity compared to that of graphene and conventional Si and InGaAs photodetectors and can mainly be attributed to low mobility of SnSe₂. The responsivity of the bilayer device is comparable to that of conventional Si photodiodes reported at 500 mA/W at 880 nm [6], and can be attributed to high absorption coefficient of bilayer SnSe₂. Ultrathin SnSe₂ flakes of ~3 nm thick were synthesized onto mica substrate by chemical vapor deposition (CVD). The flakes were transferred onto SiO₂/Si substrates. An indirect band gap of 1.78 eV was extracted. Electrical analysis revealed a mobility of ~0.6 cm²/Vs and a high on/off ratio ~2.5 × 10³. The device was illuminated with a 530 nm radiation and a fast response/recovery time of 14.5/8.1 ms with an ultrahigh responsivity of 1.1 × 10³ A/W were reported as shown in Figure 9c,d [3]. The remarkable performance of the device was attributed to the high-quality, ultrathin morphology of the SnSe₂ flakes and the formation of the Schottky contact which increases the separation efficiency of photogenerated electron-hole pairs [3]. Using a similar CVD technique, SnSe₂ crystal flakes of ~50 nm thick were developed on Si/SiO₂ substrate for photodetector applications [82]. A direct band gap of ~2.0 eV and an indirect band gap of 1.0 eV were evaluated. Their electrical analysis revealed a mobility of ~0.1 cm²/Vs

and a high-on ratio of about 100 [82]. The device was irradiated with a 543 nm radiation as shown in Figure 9e. As shown in Figure 9f, a response/recovery time of 17/45 μs with a high responsivity 0.48 A/W were reported.

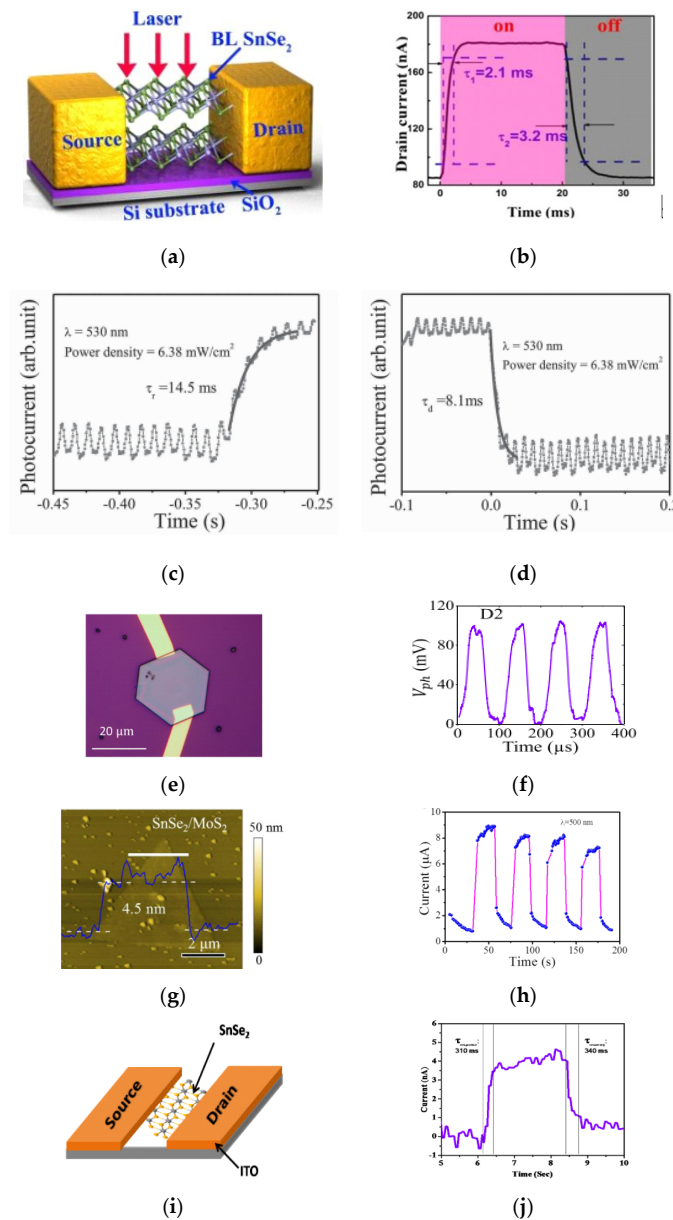


Figure 9. (a) Schematic representation of the photodetector device consisting of a bi-layered SnSe₂ on SiO₂/Si wafer with laser illumination. (b) Photocurrent dynamics of one period of the time-resolved photoresponse. The laser used in (b) is 633 nm with a power of 4 mW. (a,b) are reproduced with permission from reference [81]. (c,d) Rise and decay curves measured using an oscilloscope and fitted with a single-exponential function. (c,d) are reproduced with permission from reference [3]. (e,f) Optical image of electrode deposited onto a SnSe₂ crystal (thickness ≈ 50 nm) to make the optoelectronic device, Rise and fall time for the SnSe₂ photoconductor within the microsecond regime. (e,f) are reproduced with permission from reference [82]. (g,h) AFM image of a typical SnSe₂/MoS₂ hetero-structure, Time-dependent photoresponse of the photodetector under 500 nm light illumination. (g,h) are reproduced with permission from reference [83]. (i,j) Schematic of fabricated two probe device with few-layer SnSe₂ and typical single I - t curve for response and recovery time measurements. (i,j) are reproduced with permission from reference [84].

It is not clear how such a high response/recovery time can be related to a device of such low mobility although the good performance of the device was mainly attributed to the formation of a Schottky barrier between SnSe₂, SiO₂/Si and metal contacts [82]. From mobility point of view, WSe₂ has a higher mobility that ranges between 100–500 cm²/Vs [85–91], which is ten times more than that of SnSe₂ (<10 cm²/Vs) [71]. We can assert that the higher response time of the hetero-junction is mainly attributed to the higher mobility of WSe₂ in addition to the asymmetric band offsets at the two interfaces of the ITO/WSe₂/SnSe₂ hetero-junction that creates a large built-in potential across a small thickness of WSe₂ resulting into a large built-in field [92]. A vertical photodetector device based on a SnSe₂/MoS₂ van der Waals hetero-structure SiO₂/Si substrate, as shown in Figure 9g, has also been reported [83]. When the device was illuminated with a 500 nm radiation, a responsivity of 9.1×10^3 A/W and response/recovery time constants of 0.2/0.6 s were obtained [83]. The transient response is shown in Figure 9h. On the basis of the mobility of MoS₂ that has a range of values between 0.1 and 200 cm²/Vs depending on the method of synthesis and number of layers [93–104], one can assert that the relatively slow response time is due to low mobility of the constituent semiconductors compared to SnSe₂/WSe₂ hetero-structures. Compared to other findings, the authors attributed the response time to efficient charge transfer at SnSe₂/MoS₂ hetero-structures which were formed via epitaxial growth [83]. In a study that involves a transparent conducting oxide as a substrate, few-layer nano-sheets were developed by solvothermal approach with sheet thickness of about 3 nm [84]. A white light photodetector device was fabricated based on ITO/SnSe₂, as shown in Figure 9i. The response/recovery time constants of the fabricated device were estimated as 310/340 ms respectively with transient response shown in Figure 9j. Manoj et al. recently have developed SnSe₂ based photodetector on soda lime glass substrate for NIR (1064 nm) photodetection [105]. The device showed a responsivity of ~0.8 mA/W with rise and decay times of 276 ms/332 ms. The slow response times may due to the trap states present in the system. The details are summarized in Table 1.

Table 1. Wavelength range, responsivity, response/recovery speed of SnSe₂, and its related hetero-structures.

Device Structure	Detection Wavelength (nm)	Responsivity (A/W)	Response/Recovery Time	Ref
SLG/SnSe ₂ -Bulk	1064	$\sim 2 \times 10^{-3}$	7.76 s/2.5 s	[71]
SLG/SnSe ₂ /PEDOT:PSS-Bulk	1064	$\sim 1.4\text{--}2.6 \times 10^{-6}$	1.33 s/1.22 s	[5]
p-Si/n-SnSe ₂ -Bulk	1064	~0.12	57 ± 25/34 ± 15 μs	[66]
Bi-layer (BL) SnSe ₂ /SiO ₂ /Si	633	~0.5	2.1 ± 0.3/3.2 ± 0.2 ms	[81]
SnSe ₂ flakes/SiO ₂ /Si	530	$\sim 1.1 \times 10^3$	14.5/8.1 ms	[3]
SnSe ₂ /SiO ₂ /Si	543	~0.48	17/45 μs	[82]
ITO/WSe ₂ /SnSe ₂ /SiO ₂ /Si	785	~1100	10 μs	[92]
SnSe ₂ /MoS ₂ /SiO ₂ /Si	500	$\sim 9.1 \times 10^3$	0.2/0.6 s	[83]
ITO/SnSe ₂	white light	-	310/340 ms	[84]
SnSe ₂ /SLG	1064	$\sim 0.8 \times 10^{-3}$	276/332 ms	[105]

5. Performance of Photodetectors Based on MoS₂

We next look at MoS₂, the most studied layered transition-metal dichalcogenides with appealing optical properties that are layer dependent. A single layer is about 6.5 Å thick and has been extracted using scotch tape method by various research groups. Bulk MoS₂ has an indirect band gap of 1.2 eV whereas single-layer MoS₂ has a direct band gap of 1.8 eV [106]. The mobility of MoS₂ has a range of values between 0.1 and 200 cm²/Vs depending on the method of synthesis and number of layers [93–104]. In this regard, a phototransistor was fabricated based on single-layer MoS₂ by mechanical exfoliation onto Si/SiO₂ substrate as shown in Figure 10a [107]. A carrier mobility of 0.11 cm²/Vs was extracted for the bottom-gate FET device configuration [107]. The device was irradiated with broad band wavelength from 450 to 800 nm. The photocurrent was higher for wavelengths lower than 670 nm which was attributed to energy greater the corresponding band gap of mono-layer MoS₂, a photoresponsivity of ~7.8 mA/W and a response/recovery time constants 50 ms were reported with transient response

shown in Figure 10b [107]. The slow response time as compared to graphene was attributed to low carrier transport [107]. In another study, a photodetector was fabricated based on monolayer MoS₂ through mechanical exfoliation onto Si/SiO₂ substrate [32]. The spectral response revealed a direct band gap of ~1.8 eV [32]. From their electrical analysis, a mobility of 4 cm²/Vs was extracted from the back-gate FET device configuration. The device was excited with a focused laser of wavelength 561 nm and a responsivity of 880 AW⁻¹ at a wavelength of 561 nm was estimated. The response/recovery times were estimated as 4/9 s respectively [32]. We wish to point out that the response time reported is in accordance with the carrier mobility extracted from the device and the ultra-high responsivity can mainly be attributed to the high absorption coefficient of MoS₂.

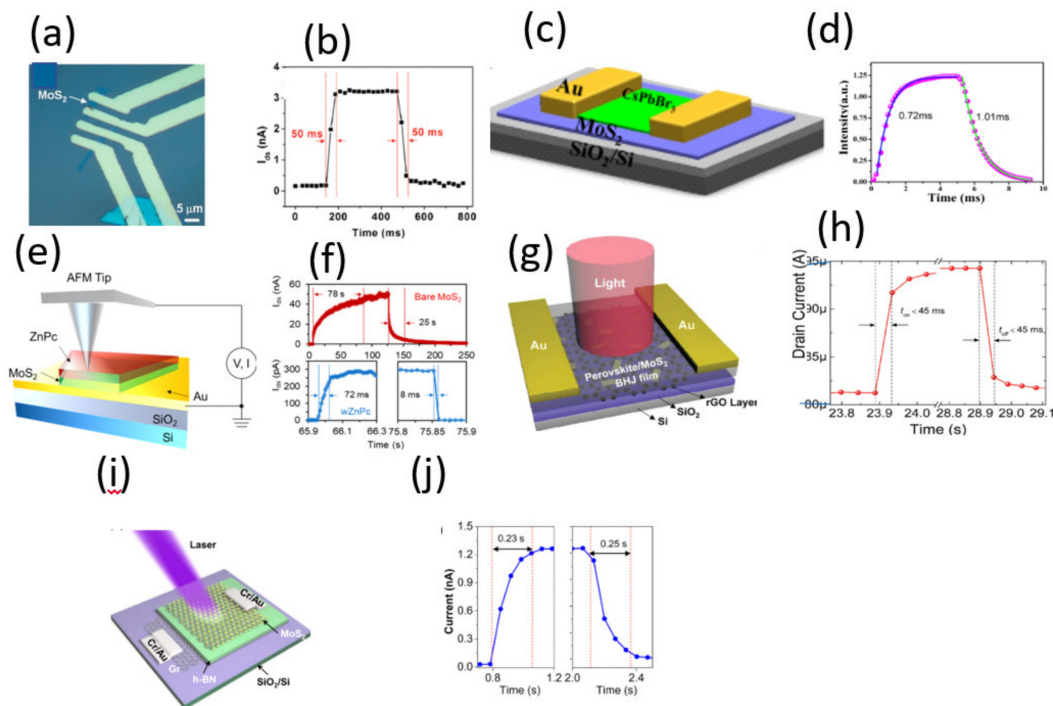


Figure 10. (a,b) Optical image of FET device made by single-layer MoS₂ and photo-switching rate. Reproduced with permission from reference [107]. (c,d) Schematic illustration of the hybrid MoS₂/CsPbBr₃ photodetector and temporal photocurrent response of the hybrid device. Reproduced with permission from reference [108]. (e,f) Schematic of the conductive AFM measurement and time-dependent photoresponse dynamics for a MoS₂ device after varied ZnPc treatments plotted on a linear scale. Reproduced with permission from reference [109]. Copyright 2018, American Chemical Society. (g,h) Device structure of the hybrid photodetector based on perovskite/MoS₂ BJJ on rGO and photo-switching characteristics of the perovskite/MoS₂ BJJ-rGO hybrid photodetector measured alternately in dark and under 660 nm light illumination (1 mW cm⁻², V_G = 0 V, V_{DS} = 2 V). Reproduced with permission from reference [110]. (i,j) Optical images of the device (scale bar: 10 μm), Dynamic photoresponse obtained from device. Reproduced with permission from reference [111]. QD-MoS₂ hybrids with interfacial interaction dominated by charge transfer (left) and by non-radiative energy transfer (right), Time-resolved analysis of the normalized rise times of photocurrent at V_G = -40 V and V_{DS} = 0 V under 488 nm laser with 37 μW. Reproduced with permission from reference [112]. (i,j) Schematic of the MoS₂/BN/graphene hetero-structure photodetector for photon absorber/selective hole tunneling layer/bottom electrode, respectively and Time-resolved photoresponse of the 7 h-BN nm device under 405 nm laser irradiation. Reproduced with permission from reference [113].

The response time could further be improved with devices that involve Schottky junctions and heterostructures using MoS₂ as one of the constituent semiconductors were fabricated. Some of the fabricated devices are discussed as follows.

An ultrasensitive and broadband MoS₂ photodetector on SiO₂/Si substrate driven by a ferroelectric poly(vinylidene fluoride/trifluoroethylene) P(VDF-TrFE) employed to suppress the dark current of the MoS₂ semiconducting channel has been developed [114]. The device was illuminated with a 635 nm radiation, and a responsivity of 2570 A/W with a fast response/recovery time of 1.8/2.0 ms respectively were estimated [114]. A FET mobility of 85 cm²/Vs was estimated from the differential *I-V* curves which is of the same order of pristine MoS₂ [114]. The fast response time could have resulted from the strong built-in electric field at the interface of ferroelectric P(VDF-TrFE) and MoS₂. In a study with device configuration shown in Figure 10c for MoS₂/CsPbBr₃ fabricated on SiO₂/Si substrate [108], a responsivity of 4.4 A/W at 442 nm radiation with a response/recovery times of 0.72/1.01 ms were reported with transient response shown in Figure 10d. The findings were compared with the response/recovery time exclusively based on perovskite CsPbBr₃ which was estimated 62.5/18.2 ms. By analyzing the carrier mobility of CsPbBr₃ that ranges between ~77 and 1000 cm²/Vs [115–118], which is much higher than that of MoS₂, we can assert that the difference in the rising and decay times between the perovskite photodetector with or without MoS₂ is due to the higher mobility of CsPbBr₃. The authors attributed this to the carrier transfer from perovskite to MoS₂ layer which induced trap passivation on the substrate [109]. Similarly, Huo et al. developed CsPbBr₃/MoS₂ heterojunction phototransistor on SiO₂/Si substrate. The device was illuminated with 442 nm radiation, a responsivity of 13.1 A/W and the response/recovery time were estimated as 2.5/1.8 ms respectively [119]. Next we analyze a study with device configuration zinc phthalocyanine (ZnPc)/MoS₂ developed on SiO₂/Si substrate shown in Figure 10e for a ultrafast photoresponse [109]. A comprehensive study that involves charge transfer at the interface between MoS₂/SiO₂/Si and ZnPc/MoS₂/SiO₂/Si was carried out [109]. For MoS₂/SiO₂/Si device, the photoresponse was observed to persist for a minute and for 40 min ZnPc-treated MoS₂, a fast rise and decay times of 72 and 8 ms respectively were estimated. The transient photo-responses are shown in Figure 10f. The mobility values of ZnPc is of order 10⁻³–10⁻⁴ cm²/Vs and this value may not play a major role in charge transportation [120–122]. The slow response in MoS₂/SiO₂/Si was attributed to inherent defect states in MoS₂ and the localized trap states at the MoS₂/SiO₂ interface in addition to inherent low mobility. The improved response time in ZnPc-decorated MoS₂ was regarded as a result of the suppressed slow hole trapping in the above localized states [109]. The same authors improved the responsivity of the device to 430 A/W by introducing Al₂O₃ passivation layer that could screen charge impurity scattering [109]. The response time of the device remained fast at ~100 ms after the passivation still 100 times faster than the bare MoS₂ device. The slightly slower response compared to ZnPc-treated MoS₂ was attributed to the increased number of the inherent deep trap centers in MoS₂ and at the interface that involves the minority hole trapping when Fermi level is tuned close to the conduction band [109]. A hetero-junction was fabricated, as shown in Figure 10g, based on CH₃NH₃PbI₃/MoS₂ on SiO₂/Si substrate combined with reduced graphene oxide (r-GO) as a hole transport layer for photodetector applications [110]. The device was illuminated with a 660 nm wavelength light and a responsivity of 1.08 × 10⁴ AW⁻¹ and as shown in Figure 10h, a fast response/recovery speed of shorter than 45 ms were reported [110]. The carrier mobility of CH₃NH₃PbI₃ varies between 1 and 100 cm²/Vs for polycrystalline and single crystals respectively [123–125]. The carrier mobility of r-GO varies between 1.8–83 cm²/Vs [126,127]. These values suggest that the mobility of CH₃NH₃PbI₃, r-GO, and MoS₂ are comparatively equal and therefore, as expected, there is no much significant change in response/recovery time constants on the basis of mobility, if any, it could be due to the presence of a narrow depletion width. Van der Waals hetero-structure photodiode based on GaSe/MoS₂ on SiO₂/Si substrate have been developed [111]. When the device was illuminated with a 532 nm radiation, a responsivity of ~3 A/W and a response time of 50 ms were obtained [111]. On the basis of mobility, the mobility of GaSe has been reported and varies between 0.4–50 cm²/Vs [128–132], which is of similar order to that of MoS₂ and in accordance with the observed response/recovery time constants. A study that applies graphene as an electrode with device structure MoS₂/h-BN/graphene was developed on SiO₂/Si substrate where h-BN is used as an insulating layer between graphene electrode and MoS₂ photo-absorber as shown in Figure 10i [113]. On the illumination of the device

with 405 nm of light, a responsivity of 180 AW^{-1} and a response/recovery speed of 0.23/0.25 s were reported with transient response shown in Figure 10j [113]. Yang et al. developed $\text{MoS}_2/\text{MoSe}_2$ hetero-junction on SiO_2/Si substrate [133]. A fast response time of $\sim 10 \text{ ms}$ and a responsivity of $\sim 350 \text{ A/W}$ were reported when the device was illuminated with a pulsed light of 633 nm and was faster by 1–2 orders of magnitude as compared to isolated MoSe_2 and MoS_2 devices [133]. In comparison to fabricated MoSe_2 phototransistors with a bottom-gate configuration on SiO_2/Si substrates [134], that possess a fast response/recovery speed of 1.7/2.2 ms at 650 nm radiation with a responsivity of $1.4 \times 10^5 \text{ A/W}$, we can assert that the fast response in the hetero-structure is due to better mobility of MoSe_2 as compared to that of MoS_2 . The mobility of MoSe_2 varies between $15\text{--}118 \text{ cm}^2/\text{Vs}$ [38,135–137], and is comparatively higher than that of MoS_2 . A high-performance MoS_2/CuO nano-sheet hetero-junction shown in Figure 11a was developed on SiO_2/Si substrate for photodetector applications [138]. As shown in Figure 11b, a fast photoresponse/recovery speed of $\sim 34.6/51.9 \text{ ms}$ with a responsivity of $\sim 157.6 \text{ A/W}$ were found at 570 nm illumination [138]. The mobility of CuO reported between 0.01 and $8 \text{ cm}^2/\text{Vs}$ [139–141] is lower than that of MoS_2 suggesting no significant effect in response/recovery time constants of the hetero-structure, if any, it could be due to a strong built-in electric field at the junction. Chen et al. developed MoS_2 photodetectors enhanced by graphene QDs as shown in Figure 11c on SiO_2/Si substrate [142]. With a 405 nm radiation, a responsivity of $\sim 10^4 \text{ A/W}$ and a response time of 70 ms and recovery speed of 1.23 s, 10.97 s were reported [142]. The transient response is shown in Figure 11d. The response/recovery speed is low yet graphene QDs are known to have a high electron mobility [143,144]. The authors attributed the slow response to either defects or charge impurity states inside the band gap or by the presence of trap states between MoS_2 and the underlying SiO_2 layer, which usually occurs for MoS_2 grown by CVD [142].

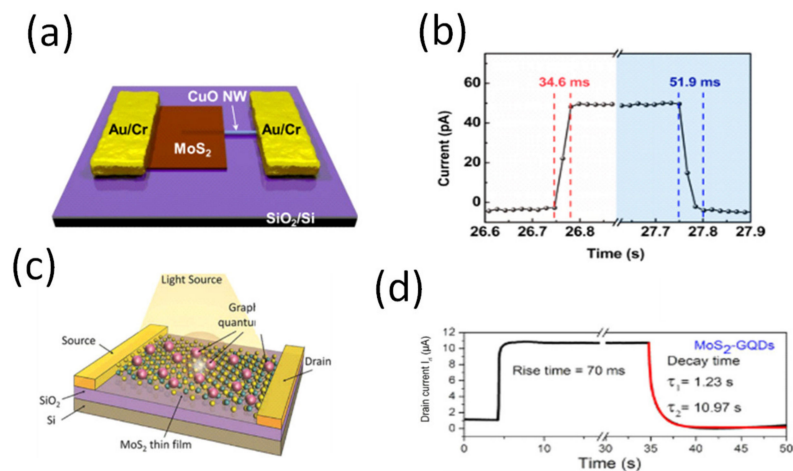


Figure 11. (a,b) Schematic representation of a MoS_2/CuO nano-sheet on 1D hetero-junction photodiode, photo-responsive rise and decay times of a MoS_2/CuO hetero-junction photodiode under light illumination of $\lambda = 570 \text{ nm}$ at $P_{\text{light}} = 1.4 \text{ mW}$ and a bias voltage of -2 V . Reproduced with permission from reference [138], Copyright 2016, American Chemical Society (c,d) Schematic of a MoS_2 -GQDs hetero-structure phototransistor, Time-dependent photoresponse of MoS_2 -GQDs device. Reproduced with permission from reference [142].

A low noise and fast photoresponse of few-layered MoS_2 passivated by $\text{MA}_3\text{Bi}_2\text{Br}_9$ on SiO_2/Si substrate has been developed as shown in Figure 12a. With a 530 nm light, a responsivity of 112 A/W and the response/recovery speed of 0.3 ms were obtained. The transient response is shown in Figure 12b [145]. Our literature search could not reveal the mobility of $\text{MA}_3\text{Bi}_2\text{Br}_9$ but the response time suggests that it is of the same order to that of MoS_2 . The $\text{MA}_3\text{Bi}_2\text{Br}_9$ passivation was responsible for fast and strong photo-response of the MoS_2 [145]. A 3D rGO- MoS_2 /pyramid Si hetero-junction with device structure as shown in Figure 12c was fabricated for ultrahigh detectivity

and ultra-broadband photodetection on Si/SiO₂ substrate [146]. The device was illuminated with a 808 nm radiation and a responsivity of 21.8 A/W and a response/recovery of 2.8/46.6 μs were found [146]. The transient response is shown in Figure 12d. To elucidate on the fast response time, two devices were compared one with RGO–MoS₂/pyramid Si and the other with MoS₂/pyramid Si device (rise time/decay time = 32.6/87.8 μs). We wish to mention here that the carrier mobility of r-GO varies between 1.8 and 83 cm²/Vs [126,127]. The authors attribute the faster response time to the insertion of RGO in the device that improved conductivity of MoS₂ film [146]. A self-powered broadband, high-detectivity and ultrafast photodetectors based on Pd–MoS₂/Si hetero-junction with device structure shown in Figure 12e was developed [147]. The device was illuminated with a broad band radiation (300–1100 nm), a responsivity of ~654.0 mA/W at 950 nm and a response/recovery speed of 2.1/173.8 μs were reported [147]. The transient response is shown in Figure 12f. The mobility of n-Si is about 1500 cm²/Vs which is several orders of magnitude higher than that of MoS₂ [59]. We believe, it is this high electron mobility and a strong built-in electric field at the interface that are response for the fast response time. A gate-tunable carbon nanotube–MoS₂ hetero-junction p–n diode has been developed on SiO₂/Si substrate as shown in Figure 12g [148]. The device was illuminated with a 650 nm radiation, a responsivity exceeding 0.1 A/W with a response time of 15 μs (limited by the instrument set-up) was reported [148]. The transient response is shown in Figure 12h. On the basis of mobility, carbon nano tubes (CNTs) are reported to have a high mobility of >10⁴ cm²/Vs [149,150], several orders of magnitude higher than that of MoS₂. We believe it is this extraordinary high mobility of CNTs that is primarily response for the fast response of the device. A high-performance photovoltaic detector based on MoTe₂/MoS₂ van der Waals hetero-structure shown in Figure 12i was fabricated on SiO₂/Si substrate [151]. The device was illuminated with a 637 nm radiation, a responsivity of 46 mA/W and a response/recovery speed of 60/25 μs was reported [151]. The transient response is shown in Figure 12j. The carrier mobility of MoTe₂ is of ranges between 0.3–4000 cm²/Vs [152–156], depending on the preparation conditions. This value suggests that the mobility of MoTe₂ is greater than that of MoS₂ and this explains the higher response time observed in the heterostructure in addition to the strong built-in electric field. Henning et al. fabricated a mixed-dimensional single and multilayer MoS₂/p-silicon nanowire hetero-junction with device structure shown in Figure 12k on SiO₂/Si substrate and studied the charge separation at the junction. [157] They carried out time-resolved photocurrent measurements on four types of devices: p-Si nanowire/n-MoS₂ monolayer devices, n-Si nanowire/n-MoS₂ monolayer devices, n-MoS₂ monolayer metal/semiconductor/metal (MSM) photoconductors, and p-Si nanowire/n-MoS₂ multilayer devices. [157] The transient response is shown in Figure 12l. For p-Si nanowire/n-MoS₂ monolayer devices, n-Si nanowire/n-MoS₂ monolayer devices and MSM photoconductors devices, a response/recovery speed of 110 μs was estimated at instrumental resolution of 12 μs. When the instrumental resolution was increased to 200 kHz, the response and recovery times were estimated as 1.4/1.6 μs respectively. For a multilayer MoS₂/Si nanowire, the response/recovery times were estimated as 0.7/1.1 μs respectively. Their findings suggest that the response time depends externally on the mobility of silicon nanowire which is of several orders higher than that of MoS₂ and strong electric field created as a result of the depletion width at the interface [157]. Similarly, Wang et al. fabricated Pd-single layer MoS₂ Schottky junction on SiO₂/Si substrate [158]. A responsivity of 0.88 A/W and a response/recovery speed of 24.2/24.5 ms respectively were estimated when the device was illuminated with a 425 nm radiation [158]. Wang et al. developed a MoS₂/CdTe p–n hetero-junction on Si/SiO₂ substrate with device schematic shown in Figure 12m for broadband response up to 1700 nm, a responsivity of 36.6 mA/W and a response/recovery speed of 43.7/82.1 μs at a pulsed light illumination of 780 nm radiation was reported [159]. The transient response is shown in Figure 12n. Here, the carrier mobility of CdTe is of order 10³ cm²/Vs [160–164], which is several orders higher than that of MoS₂ and as expected, the hetero-structure has a high response time. Hao et al. developed MoS₂/SiO₂/Si p-i-n junction device structure [165]. The device was illuminated with 650 nm radiation and the response/recovery times were estimated as 16.2/160.5 μs respectively. As expected, the response time in this p-i-n device structure is controlled by the higher mobility of Si.

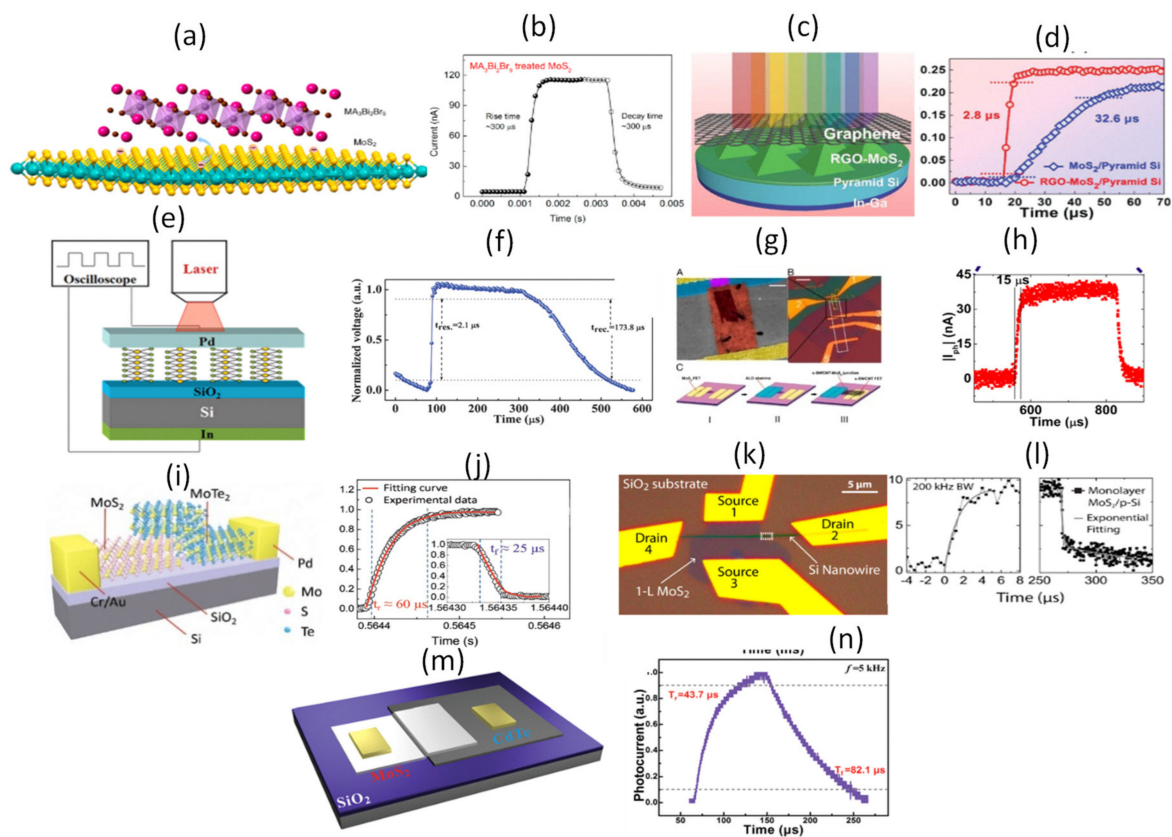


Figure 12. (a,b) Schematic diagram of MoS₂ and MA₃Bi₂Br₉ hetero-structure, Rise and decay time of MA₃Bi₂Br₉-treated MoS₂ device irradiated by 530 nm LED with a power density of 3.3 mW/cm². Reproduced with permission from reference [145]. (c,d) Schematic illustration of the structure of 3D RGO–MoS₂/pyramid Si hetero-junction photodetector, rise edges, Reproduced with permission from reference [146]. (e,f) Schematic illustration of the setup for measuring the response time of the Pd–MoS₂/Si device, Single normalized modulation cycle measured at 2000 Hz. Reproduced with permission from reference [147]. (g,h) False-colored SEM image of the hetero-junction diode. (Scale bar, 2.5 μm.) The yellow regions at the top and bottom are the gold electrodes. The patterned alumina (blue region) serves as a mask for insulating a portion of the SL–MoS₂ flake (violet region). The pink region is the patterned random network of s-SWCNTs (p-type) in direct contact with the exposed part of the SL–MoS₂ flake (n-type) to form the p–n hetero-junction diode (dark red), Time- dependent photoresponse of the p–n hetero-junction showing fast rise and decay times of ~15 μs, Reproduced with permission reference [148]. (i,j) Schematic diagram of the MoTe₂/MoS₂ van der Waals hetero-structure, Time resolved photoresponse of the hetero-structure at V_{sd} ≤ 0.51 V. The inset is falling edge of the response, Reproduced with permission from reference [152]. (k,l) Optical micrograph of a device in which a MoS₂ monolayer was transferred onto a p–Si nanowire followed by contact fabrication, Transient photocurrent of monolayer MoS₂/Si nanowire p–n hetero-junction (black squares) measured with a high-speed current preamplifier (200 kHz bandwidth) at V_D = –8 V. Rise and fall times are fit with single and bi-exponentials (gray lines), and equal t_{rise} = 1.4 μs and t_{fall} = 1.6 μs (80 μs), respectively. Reproduced with permission from reference [159]. (m,n) Schematic illustration of a MoS₂/CdTe hetero-junction device, Rising and falling edges for estimating the rise time and the fall time at 5 kHz. Reproduced with permission from reference [165].

Tang et al. developed MoS₂ nano-sheet photodetectors with ultrafast response on SiO₂/Si substrate [166]. The device was illuminated with 532 radiation, a responsivity of 59 A/W and a response time of 42 μs were reported [166]. The authors fabricated the device with metals of different work functions such as Pd, Cr, Au and Ti. The fast response time can be attributed to the presence of a strong electric field at the interface between MoS₂ and the metal contacts that forms

a Schottky junction. Oliva et al. developed van der Waals MoS₂/VO₂ hetero-junction on SiO₂/Si substrate for photodetector applications [167]. The device was illuminated with a 500/650 nm radiation, a maximum photoresponsivity of 1.25 A/W at 550 nm and a response time of 3.5 ms were reported [167]. By considering the mobility of VO₂ reported between 0.07–2.65 cm²/Vs [168–173]. This analysis suggests that the mobility of VO₂ has little effect on response/recovery speed of MoS₂/VO₂ hetero-structure, if any, it could be due a strong built-in electric field at the interface of the constituent semiconductors. Liu et al. fabricated a MoS₂/GaN NW p–n junction on SiO₂/Si substrate for photodetector applications [174]. The device was illuminated with a 550 nm radiation, a responsivity of 443.3 A/W and a response/recovery speed of 5 ms were reported [174]. The response time is 20 times faster probably due to anisotropic dependence of mobility for GaN nano-wire [175]. Lei et al. fabricated a MoS₂/black phosphorous (BP) heterostructure photodetector [176] with responsivity of ~153.4 mA/W under 1550 nm illumination with rise and decay times of 15/70 μs. The fast response times can be attributed to the high mobility of BP. The detailed response/recovery speed and their device structures are summarized in Table 2.

Table 2. Wavelength range, responsivity, response/recovery speed of MoS₂ on SiO₂/Si substrate and its related hetero-structures.

Device Structure	Detection Wavelength (nm)	Responsivity (A/W)	Response/Recovery Time	Ref
Single layer MoS ₂	450	~7.5 × 10 ⁻³	50/50 ms	[107]
P(VDF-TrFE) driven MoS ₂	635	~2570	1.8/2.0 ms	[114]
MoS ₂ /CsPbBr ₃	442	~4.4	0.72/1.01 ms	[117]
CsPbBr ₃ /MoS ₂	442	~13.1	2.5/1.8 ms	[119]
ZnPC treated MoS ₂ with Al ₂ O ₃ passivation layer	532	~430	100 ms	[109]
CH ₃ NH ₃ PbI ₃ /MoS ₂ with r-GO as HTL	660	~1.08 × 10 ⁴	<45 ms	[110]
GaSe/MoS ₂	532	~3	50 ms	[111]
BL MoS ₂			174/166 ms	[112]
CdSe QDs/BL MoS ₂	400–670	~50–700	159/172 ms	[109]
Core/shell CdSe/ZnS QDs/BL MoS ₂			185/172 ms	[109]
MoS ₂ /h-BN/graphene	405	~180	0.23/0.25 s	[113]
MoS ₂ /MoSe ₂	633	~350	10 ms	[133]
MoS ₂ /CuO	570	~157.6	~34.6/51.9 ms	[138]
MoS ₂ /Graphene QDs	405	~10 ⁴	0.07/1.23 s	[142]
MA ₃ Bi ₂ Br ₉ passivated few-layer MoS ₂	530	~112	0.3/0.3 ms	[145]
3D r-GO-MoS ₂ /pyramid Si	808	~21.8	2.8/46.6 ms	[146]
Pd-MoS ₂ /Si	950	~0.654	2.1/173.8 μs	[147]
CNT-MoS ₂ p-n junction	650	~0.1	~15/15 μs	[148]
MoTe ₂ /MoS ₂	637	0.046	60/25 μs	[151]
Si NW-n-MoS ₂	500	-	1.4/1.6 μs	[157]
Pd-Single layer MoS ₂	425	~0.88	24.2/24.5 ms	[158]
MoS ₂ -CdTe	780	~0.0366	43.7/82.1 μs	[159]
MoS ₂ /SiO ₂ /Si p-i-n junction	650	-	16.2/160.5 μs	[165]
MoS ₂ nanosheet	532	~59	42 ms	[166]
MoS ₂ /VO ₂	550	~1.25	3.5 ms	[167]
MoS ₂ -GaN NW p-n junction	550	~443.3	5 ms	[174]
MoS ₂ /PbS	400–1500	6 × 10 ⁵	0.3–0.4 s	[53]
Graphene/MoS ₂	635	5 × 10 ⁸	-	[54]
WS ₂ /MoS ₂	532	2340	-	[177]
MoS ₂ /BP	1550	~0.153	15/70 μs	[176]

We now shift our attention to hetero-structure devices based on MoS₂ that are fabricated with one of the semiconductors taken as p/n-doped Si. The carrier mobility of n or p-doped Si ranges between 500–1500 cm²/Vs [59]. Such devices are expected to have a fast response/recovery speeds in the order of few μs or ns and for defectless devices, it should approach ps limit. For example, Wang et al. developed polycrystalline n-MoS₂ of 150 nm thick on p-Si substrate with device structure shown in Figure 13a via magnetron sputtering for self-driven visible-near infrared photodetection [178]. The device was illuminated with a 808 nm radiation, a responsivity of 300 mA/W and ultra-fast response/recovery speed of 3/40 μs were reported [178]. The transient response for half-cycle is shown

in Figure 13b. The high speed and self-driven response was partly attributed to the existence of a strong built-in electric field at the MoS₂ and Si interface [178]. Zhang et al. developed n-MoS₂/n-Si vertical multilayered hetero-junction shown in the schematic of Figure 13c for high-speed visible-near-infrared photodetectors by two step thermolysis [179]. The device was irradiated with light of wave-length 650 nm radiation, and a responsivity of 11.9 A/W and a response/recovery speed of 30.5/71.6 μs were reported [179]. The transient response is shown in Figure 13d. The high-speed response was attributed to good quality synthesized MoS₂ films and the reliable contact quality at the interface. Cong et al. developed vertically standing few layer MoS₂/p-Si hetero-junction photodetector with schematic shown in Figure 13e [180]. The device was illuminated by lasers of different wavelengths (405, 532, 671, 808, and 980 nm) as the illumination source. The 532 nm laser was used for response/recovery time measurements. An ultrafast response/recovery times of 16/176 ns respectively were estimated. The transient response for once cycle is shown in Figure 13f. The ultrafast response times can be attributed to the excellent quality of V-MoS₂/Si hetero-junction with strong light absorption and quick carrier transport speed in the unique vertically oriented few-layer MoS₂ nano-sheets and large built-in electric field at the interface of V-MoS₂ and Si [180]. Qiao et al. developed a vertically layered MoS₂/Si hetero-junction with device structure shown in Figure 13g for an ultrahigh and ultrafast broad band photoresponse from 350–1100 nm [181]. A responsivity of up to 908.2 mA/W and a response/recovery speed estimated as ~56/825 ns were reported. The transient response for one cycle is shown in Figure 13h. Kim et al. developed a high-performing MoS₂-embedded Si photodetector with MoS₂/n-Si/p-Si device architecture [182]. An ultra-fast fast response/recovery speed of 33/30 μs respectively were measured when the device was illuminated with a wavelength of around 515–520 nm. Wu et al. developed MoS₂/Si nanowire array hetero-junction shown in Figure 13i [183]. The device was illuminated with a 650 nm radiation, a responsivity of 53.5 A/W and a fast response/recovery speed of 2.9/7.3 μs respectively were reported [183]. The transient response is shown in Figure 13j. The same authors developed MoS₂/Si hetero-junction shown in Figure 13k for broadband photodetectors from deep ultraviolet to near infrared [184]. The device was illuminated with a 780 nm radiation, a responsivity of 23.1 A/W and a response/recovery speed of 21.6/65.5 μs were reported [184]. The transient response is shown in Figure 13l. Dhyan et al. developed n-MoS₂/porous silicon device structure [185]. The device was illuminated with light of wavelength between 550 and 850 nm, a responsivity of 9 A/W and a fast response/recovery speed of 9/7 μs were reported [185]. Their findings were compared with planar Si-MoS₂ hetero-junction which had a response/recovery speed of 35/8.1 μs respectively. Their analysis clearly suggest that charge collection efficiency in the porous hetero-structure is quite high compared to the planar device [185]. The same authors (Dhyan et al.) developed a high-speed Si/MoS₂ p–n hetero-junction photodetector [186]. The device was illuminated with a 580 nm radiation, a responsivity of 8.75 A/W at 580 nm and a response/recovery speed of 10/19 μs was reported [186]. Kim et al. fabricated MoS₂ layers on p-Si substrate by sputtering method for efficient photoelectric application [187]. The device was illuminated with a 455 nm radiation, a responsivity of 0.03 A/W and a response/recovery speed of 38.78/43.07 μs were reported [187]. Recently, Guo et al. [188] used a broadband photodetector of MoS₂/p⁺-Si with a responsivity of ~0.746 A/W under 808 nm illumination with rise and decay times of ~178/198 μs. The fast response times can be attributed to the inbuilt potential at the interface and also to the comparatively high mobility of p⁺-Si. The details are summarized in Table 3.

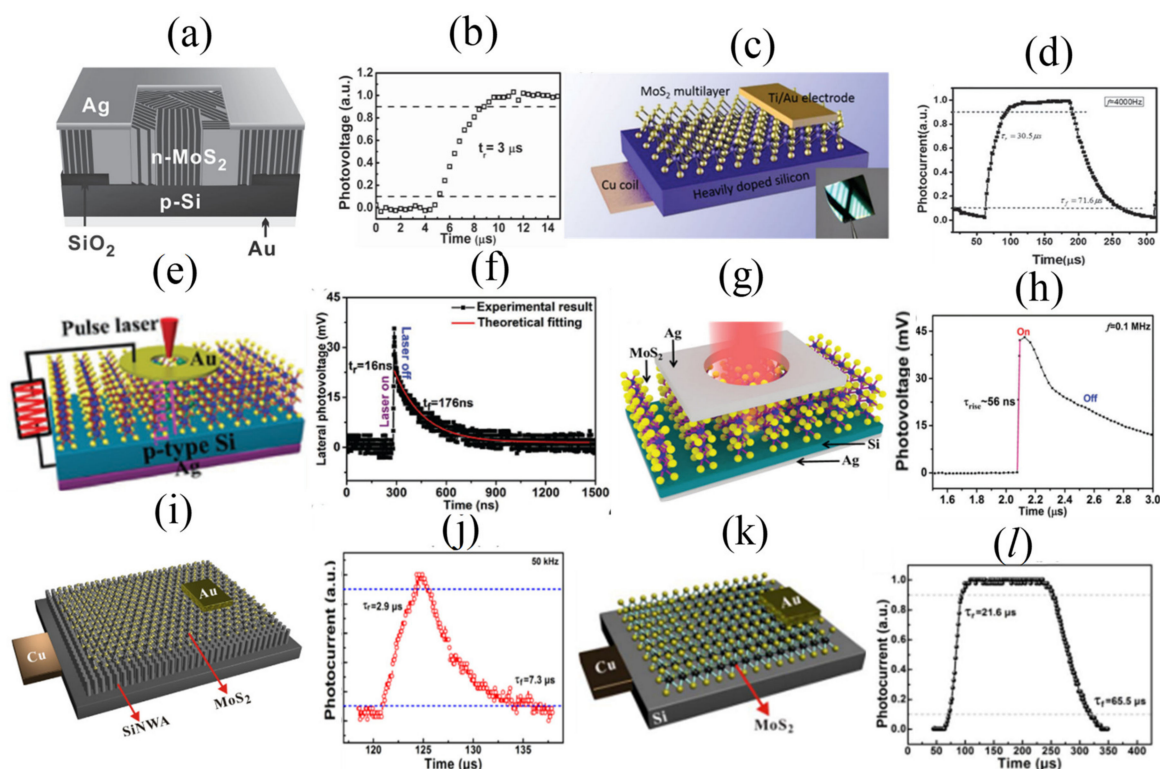


Figure 13. (a,b) Schematic illustration of the MoS₂/Si hetero-junction-based photodetector, enlarged rise edge of the photoresponse curve. Reproduced with permission from reference [178]. (c,d) Schematic representation of the vertical multilayered MoS₂/Si hetero-junction, magnified and normalized plots of one response cycle. Reproduced with permission from reference [179]. (e,f) Schematic of V-MoS₂/Si hetero-junction, time-dependent lateral photovoltage for one pulse illumination (pulse width of 100 fs), Reproduced with permission from reference [180]. (g,h) Schematic illustration of the photoresponse of the V-MoS₂/Si hetero-junction device, Time-dependent photovoltage at frequencies of 0.1 MHz (100 fs-pulse-width pulsed laser), Reproduced with permission from reference [181]. (i,j) Schematic diagram of a MoS₂/Si NWA hetero-junction device fabrication. Rising and falling edges for estimating rise time (τ_r) and the fall time (τ_f) at 50 kHz, Reproduced with permission from reference [183]. (k,l) Schematic illustration of a MoS₂/Si hetero-junction device, Rising and falling edges for estimating the rise time (τ_r) and the fall time (τ_f). Reproduced with permission from reference [184].

Table 3. Wavelength range, responsivity, response/recovery speed of MoS₂ on p/n doped Si substrate.

Device Structure	Detection Wavelength (nm)	Responsivity (A/W)	Response/Recovery time	Ref
n-MoS ₂ /p-Si	808	~0.3	3/40 μ s	[178]
n-MoS ₂ /n-Si	650	~11.9	30.5/71.6 μ s	[179]
V-MoS ₂ /p-Si	532	-	16/176 ns	[180]
MoS ₂ /Si	808	~0.9082	~56/825 ns	[181]
MoS ₂ /n-Si/p-Si	515–520	-	~33/30 μ s	[182]
MoS ₂ /Si NW array	650	~53.5	2.9/7.3 μ s	[183]
MoS ₂ /Si	780	~23.1	21.6/65.5 μ s	[184]
n-MoS ₂ /p-porous Si	550	~9	9/7 μ s	[185]
Si/MoS ₂ p-n junction	580	~8.75	10/19 μ s	[186]
MoS ₂ /p-Si	455	~0.03	38.78/43.07 μ s	[187]
MoS ₂ /p ⁺ -Si	808	~0.746	~178/198 μ s	[188]

As a benchmark, we compare the response time of graphene, commercial Si and InGaAs photodiodes with those of SnSe₂, MoS₂, and their related hetero-structures as summarized in Figure 14.

Mittendorff et al. fabricated ultrafast graphene-based broadband THz detector on Si/SiO₂ substrate. The response time was estimated as 50–100 ps in the wavelength range from 30 μm to 220 μm [18]. Commercial Si and InGaAs photodiodes are reported to have a response time of about 50 ps [6].

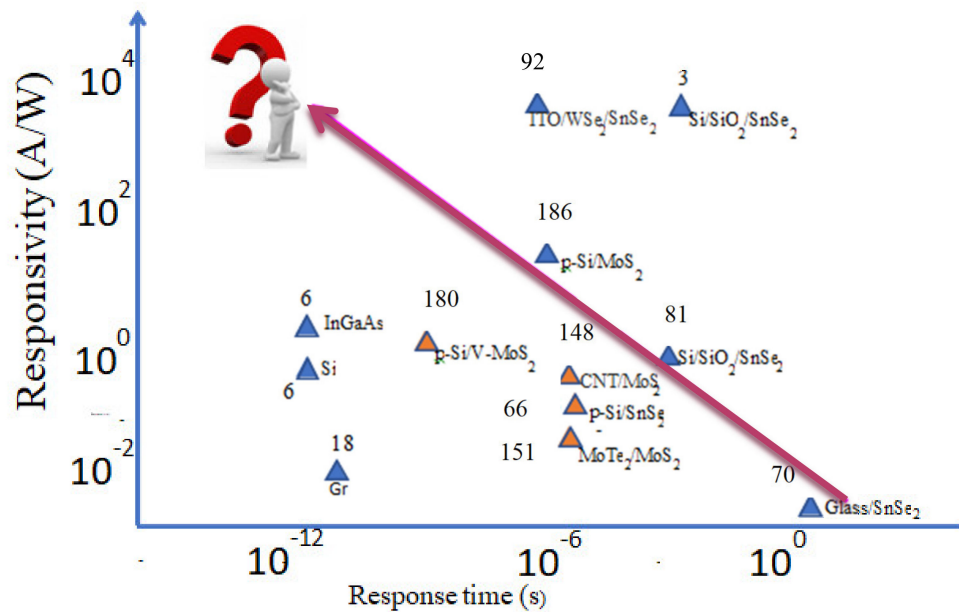


Figure 14. Reported response time and responsivities in comparison to Graphene, Si and InGaAs photodetectors.

6. Summary and Perspective

We have reviewed and analyzed the factors that influence the response time of photodetectors based on 2D SnSe₂, MoS₂ and their related hetero-structures in relation to their constituent carrier mobility, built-in electric field at the interface and compared the findings to graphene and conventional materials of Si and InGaAs. Our systematic analysis suggests that the visible and NIR responsivity of 2D SnSe₂ and MoS₂ on insulating substrates (substrates with very low carrier mobility) is greater than those of graphene, Si and InGaAs, their response time is in the order of few milliseconds to seconds and hinder their applications in devices that require high speed. Other efforts have been made by developing hetero-structures with materials/substrates of higher mobility, including graphene and Si, and this has resulted in an increase in response time to a few μs, maintaining the overall performance of the device. We would like to mention here that this review has focused on 2D SnSe₂, MoS₂, and their related hetero-structures and there are a variety of other visible and NIR-active 2D-materials and their related hetero-structures which we have not discussed in this article but which have similar advantages and draw backs to those of SnSe₂ and MoS₂. On a challenging note, the ultrahigh responsivity reported here requires the application of a certain bias voltage which would eventually increase the operational cost of the devices. To minimize this would require developing hetero-structures or Schottky diodes with a strong built-in electric field although it saturates within certain region of the interface. This requires identifying materials with a large difference in the work-functions. In realizing a response time that matches that of graphene, Si, and InGaAs, keeping the overall performance of the device remains a grand challenge. In addition, identifying materials with high visible transparency, higher mobility, a high infrared optical absorption coefficient, and developing hetero-structures with low defect states at the interface also remains challenging. To harness the advantages of these 2D-layered materials and realize the desired goals in potential device applications, incorporating them with other materials of higher mobility with a large difference in the work-functions and minimizing the interfacial defect states seems to be the important way forward for the further development of these class of photodetectors.

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