Recent progress in optoelectronic applications of hybrid 2D/3D silicon-based heterostructures

Jingshu Zhou^{1,2}, Kaiyao Xin¹, Xiangkai Zhao^{3*}, Dongmei Li³, Zhongming Wei^{*} and Jianbai Xia¹

ABSTRACT Silicon-based semiconductor technology has made great breakthroughs in the past few decades, but it is reaching the physical limits of Moore's law. In recent years, the presence of two-dimensional (2D) materials was regarded as an opportunity to break the limitation of traditional silicon-based optoelectronic devices owing to their special structure and superior properties. In consideration of the widely studied hybrid integration of 2D material detectors and 3D silicon-based systems, in this paper, the basic properties of several 2D materials used in photodetectors are summarized. Subsequently, the progress in silicon photonic integrated photodetectors based on 2D materials is reviewed, followed by the summarization of the device structure and main performances. Then, the combination of some other traditional and 2D devices is discussed as a supplement. Finally, the prospective development of the hybrid 2D/3D silicon-based heterostructures is expected.

Keywords: silicon, 2D/3D, hybrid heterostructure, optoelectronic application

INTRODUCTION

In the middle of last century, low-dimensional materials had entered the scientists' sight. After the experimental demonstration of graphene realized by Novoselov and Geim [1], the exploration of low-dimensional materials stepped into an explosive stage. In addition to in-depth investigations on graphene, not only a large number of new low-dimensional materials have been discovered, but the old bulk materials have been injected with new vitality. Research in recent years has shown that low-dimensional materials possess many unprecedented merits, such as strong light-matter coupling [2], adjustable bandgap [3], high carrier mobility [4], low dark current [5], appreciable signal-to-noise ratio [6], ultra-low energy dissipation [7], and fast response/recover speed [8]. These advantages have made low-dimensional materials potential to outperform traditional 3D bulk materials in the next generation of optoelectronics. Therefore, it is of great promise for low-dimensional materials to serve in diverse applications especially optoelectronic fields with outstanding performance.

Two-dimensional (2D) material-based devices have been widely used in photovoltaics, semiconductor, electrode and biological monitoring applications due to their unique proper-

ties. At the same time, there is less exploitation space left for conventional substrates, including Si [9,10], Ge [11,12], GaN [13], InP [14], and mica [15], since the mature industrial technology and natural unlayered crystal structures. 2D materials, while unlikely to replace the conventional materials, can complement existing technologies. Microelectronic devices will continue to develop in the direction of faster, smaller and cheaper. Graphene, transitional metal dichalcogenides (TMDCs) and other new 2D materials, with unique optical, electrical and magnetic properties and new quantum physical phenomena, have potential applications in information, micro-nano optoelectronics and other aspects. So, the cross-dimensional heterostructures which could combine the advantages of both low-dimensional materials and conventional substrates will be the best choice in the future. To date, various hybrid 1D/2D [16,17], 2D/2D [18,19], and 2D/3D [20,21] heterostructure devices have been successfully fabricated. The 2D/3D heterojunction photodetectors have attracted the most attention, which has been considered as the most feasible scheme. Because on one hand, the light absorption efficiency in 2D/3D heterostructures would be much stronger than in ultrathin 1D (2D)/2D stacking, laying solid foundation for generating photocarriers. On the one hand, the 2D/3D heterostructure devices could exhibit excellent properties that 2D devices and 3D devices cannot show alone. On the other hand, 2D/3D heterojunction photodetectors can be easily achieved by directly integrating 2D-layered materials onto mature 3D commercial substrates such as Si and GaAs *via* interlayer van der Waals interaction. The van der Waals gap in 2D materials has the potential to accommodate intercalated atomic and molecular species. In fact, epitaxial graphene on SiC has been a model system for studying intercalation, and a decoupling graphene/SiC buffer layer typically causes charge carrier scattering and n-type doping in epitaxial graphene [22]. For example, annealing epitaxial graphene at high temperatures in a H₂ environment promotes hydrogen intercalation between the buffer layer and the SiC substrate, resulting in passivation of the Si dangling bonds. Among diverse traditional platforms, optoelectronics based on silicon substrate are compatible with complementary metal-oxide-semiconductor (CMOS) processing, which can achieve mass production with the help of mature microelectronics technology. In addition, there are many advantages of silicon wafers, including low cost, easy accessibility, reliability and ability to achieve high integration. Hence, it is expectable as well as promising to realize the integration of

¹ State Key Laboratory of Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, China

² School of Electronic, Electrical and Communication Engineering, University of Chinese Academy of Sciences, Beijing 100049, China

³ Optoelectronic System Laboratory, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 10083, China

* Corresponding authors (emails: zmwei@semi.ac.cn (Wei Z); zhao_xiangkai@semi.ac.cn (Zhao X))

low-dimensional materials on silicon-based substrates. Until now, intensive efforts have been devoted to low-dimensional material/Si heterojunction devices. These silicon-based heterostructural devices have been applied in many fields, such as photodetecting, infrared imaging, polarization monitoring, and flexible device producing.

It is generally accepted that low-dimensional material/Si heterostructures are also easy to fabricate. For example, by transferring graphene on Si, An *et al.* [23] obtained an ultrasensitive graphene/Si heterojunction photodetector and realized a high responsivity of 0.33 A W^{-1} . For another example, by directly growing MoS_2 on Si substrate, Wang *et al.* [24] fabricated a MoS_2/Si heterojunction. Fig. 1 shows the schematic structure of the Si-based cross heterostructures based on different types of 2D materials. Herein, this review will be focused on the Si-based 2D/3D heterostructures to display the properties of cross-dimensional devices, along with some low-dimensional materials as complement.

PREPARATION METHODS

In general, there are two strategies in the preparation of low-dimensional materials, top-down and bottom-up. The top-down method means a reduction of thickness from the bulk material to monolayer or few layers such as exfoliation. The main approaches of the top-down method are mechanical exfoliation (ME) and mechanically assisted exfoliation. ME that helped Novoselov *et al.* [1] to discover graphene is the easiest way to get low-dimensional materials, and it was the milestone for the fabrication of low-dimensional materials. For the case of bottom-up method, chemical vapor deposition (CVD) is one of the most widely used methods for that high-quality low-dimensional materials can be controllably grown. Meanwhile, molecular beam epitaxy (MBE) and other epitaxial technologies are also usually used to synthesize low-dimensional materials. Subse-

quently, a brief introduction of these common methods will be described in this section.

Mechanical exfoliation

The ME was first created and utilized by Novoselov *et al.* [1]. They got few-layer graphene by repeated peeling from bulk graphite on transparent tapes, which ignited the flame of investigation on 2D materials and provided a cheap, easy but feasible way to access. Even now, the ME is a common means for researchers to get other kinds of low-dimensional materials.

Many assistant tools were designed to help exfoliate low-dimensional materials. Sonication-assisted exfoliation and shear-force-assisted liquid phase exfoliation (LPE) [26], ball milling-assisted exfoliation [27] are typical and commonly used in mechanically assisted exfoliation. The main principle of LPE is to take the bulk material as the raw chemical, adding to a specific solvent or surfactant to form a pretreatment solution. Then the monolayer or few-layer material from the bulk surface will be peeled with the help of high-power ultrasonic equipment. The LPE method takes advantage of cavitation and high shear force from the ultrasonic wave. The cavitation makes the bulk material be crushed, while the shear force makes the solvent form a micro-jet impact, which promotes the separation between the layers of the bulk material. Early in 2008, Coleman's group [28] added graphite powders into organic solvents, a mixed solution of *N,N*-dimethylformamide and *N*-methylpyrrolidone, and then treated the mixture by means of ultrasound and centrifugation, obtaining the graphene with few defects at last, and the production efficiency of graphene was nearly 0.01 mg mL^{-1} after 30 min ultrasonic treatment. LPE features easy operation and high production efficiency, so that it is expected to be a potential path in industrial production of low-dimensional materials, especially graphene. On the other hand, LPE can be used to obtain some few-layer or single-layer materials that are difficult

Figure 1 (a) Diverse types of 2D materials and their response ranges. (b) Optical image of 2D materials/Si heterostructures. The 2D materials are combined with Si substrate and the top electrode is attached to the oxide layer. (c) Schematic diagram of a 2D/Si heterostructure device. (d) Sectional view of a 2D/Si optoelectronic device. (b) is reprinted with permission from Ref. [25]. Copyright 2021, Springer Nature Limited.

to peel by ME. Besides, many other famous 2D materials, including h-BN [29], some kinds of TMDCs [30], and black phosphorus (BP) [31], could also be obtained by LPE. To enhance the efficiency and increase the production efficiency of low-dimensional materials, tip sonication with a high power density and high shear rates could be used. Ball-milling-assisted exfoliation, adept at producing large-scale samples, is another mechanical method to exfoliate low-dimensional materials, which also takes advantage of shear force. And several materials (graphene [32], h-BN [33], MoS₂[34]) have been successfully obtained by using this method.

Epitaxial methods

Although relatively high-quality materials could be obtained through aforementioned approaches, the acquisition of the products is almost uncontrollable and the final size of the crystal is restrained. In order to get large-scale or even wafer-scale monolayer, it is generally accepted that epitaxial growth will be the most appropriate as well as feasible way. Because compared with the exfoliation, the nucleation and growth processes in epitaxial growth are under precise control, resulting in accurate arrangement of the thickness and atomic configuration, which acts as the fundamental for obtaining large-scale products. In this part, we will introduce some epitaxial methods to prepare low-dimensional materials.

Chemical vapor deposition

CVD is the most popular technique for preparing high-purity and high-performance films. Some heterostructures can be formed by growing low-dimensional materials directly on the substrate via CVD technology [35,36]. In the process of CVD, one or more vapor source atoms or molecules are carried by carrier gas in the chamber, and the desired film will be formed on the substrate surface through chemical reaction. CVD is usually used to grow graphene, h-BN, MX₂ (M = Mo, W, Re and so on, X = S, Se, Te), and some kinds of metal oxides. In the CVD process, the dosage ratio of raw materials plays a critical role. For example, Zhou *et al.* [37] demonstrated an effective CVD strategy, using a Te:metal oxide:metal chloride weight ratio of 1:1:1, to synthesize few-layer and monolayer WTe₂ and MoTe₂ on a large scale [38]. However, it is not generally suitable for all low-dimensional materials. For example, BP, b-As_xP_{1-x} and metal oxides cannot be obtained through CVD, requiring complementary fabrication methods by which the shortcomings of CVD could be overcome.

Molecular beam epitaxy

MBE is a technology to epitaxially grow materials on the crystalline substrate in an ultrahigh-vacuum environment. MBE can precisely control the thickness, purity, doping density and composition of dopants. Nevertheless, the popularization of MBE in fabrication of 2D materials is limited by its high cost and complex operation [39]. To satisfy different demands of various materials, other epitaxial methods are exploited. For example, considering the difficulty of preparing large-scale as well as atomically thin BP film by ME and CVD, Xu *et al.* [40] reported the direct growth of large-scale crystalline BP films on insulating silicon substrates. With polyphosphide Au₃SnP₇ serving as the nucleation seeds, large-size BP film appeared from epitaxial nucleation to controllable lateral growth, following a newly developed gas phase growing strategy. As reported, the field-

effect and Hall mobility reached 1000 and 1400 cm² V⁻¹ s⁻¹ at room temperature, respectively.

Atomic layer deposition

In low-dimensional heterostructures, insulating layer would usually be introduced to make a potential barrier. Atomic layer deposition (ALD) is a special CVD technique used to deposit atomically thin films onto a substrate. By ALD, controllable thickness, uniformity, composition of the film and compatibility to most substrates can be realized at the same time. Furthermore, ALD can be used to prepare ultrathin films of many materials, such as oxides, nitrides, sulfides, carbides, fluoride and even polymers [41,42]. It is noteworthy that the ALD technology is usually carried out in vacuum at relatively low temperatures, with the help of which ALD could be applied to sensitive surface but free of pinholes, cracks and defects. Some high-K oxide layers serving as dielectric layers, such as Al₂O₃, HfO₂, and SiO₂ [43,44], are typically prepared by ALD.

Electron beam evaporation

Electron beam evaporation (EBE) method is a kind of vacuum evaporation coating method, which uses electron beam to evaporate materials by heat in vacuum environment [45]. The evaporated materials will be transported to the substrate and condensed into thin films on the substrate. In the electron beam heating device, the heated material is placed in the water-cooled orange vortex, which can avoid the reaction between the evaporating material and the orange wall, thus leading to final products with high purity and quality. Moreover, multiple crucibles can be installed in the same equipment to realize simultaneous or sequential evaporation and deposition. Even the materials with high melting point can be turned into high-quality thin film by EBE, which makes it a widely popular method in preparation of conductive glass and other optical films.

FIGURE OF MERITS

Photodetectors are the devices in which electronic signals would be monitored after absorption of external photons. Researchers have put forward several kinds of mechanics to explain the photovoltaic conversion for various photodetectors. The most common mechanisms are photoconductive effect (PCE) and photovoltaic effect (PVE). When the photon energy of incident light is larger than the bandgap energy, the electrons will be transited to excited state, leading to the emergence of non-equilibrium carriers, thus increasing the total carrier concentration. In this case, an external electric field should be exerted to separate the photon-generated electron-hole pairs, by which the difference between photocurrent and dark-current could be distinguished. Particularly, photogating effect (PGE) could be seen as a special circumstance of PCE in the case of photodetectors based on ultrathin low-dimensional materials. It utilizes the trapped stage as a localized floating gate to modulate the channel conductance. On the contrary, for the PVE photodetectors, the photogenerated carrier will be separated by a built-in electric field, and the photocurrent could be formed without extra power. Moreover, photothermoelectric effect (PTE) and bolometric effect (BE) are usually used to explain the photoelectric process which is dominated by thermodynamics principle.

Although it seems that various photodetectors work following

different mechanisms, there are some universal standards to estimate the quality of these photodetectors. In the following part, we will introduce some primary indicators.

Responsivity (R)

Responsivity represents the ability of photoelectric conversion of photodetectors, which is defined as the ratio of the generated photocurrent (I_{ph}) or output voltage (V_{ph}) to the average power of the incident light (P_{in}) on the photodetector. It can be calculated by the following equation:

$$R_i = \frac{I_{ph}}{P_{in}S}, \text{ or } R_v = \frac{V_{ph}}{P_{in}S}, \quad (1)$$

where S is the illuminated area, the units of the R_i (responsivity based on photocurrent) and R_v (responsivity based on voltage) are $A W^{-1}$ and $V W^{-1}$, respectively.

External quantum efficiency (EQE)

It is well-known that when incident light is shined on the device, not all photons will be used to generate nonequilibrium carriers. EQE denotes the ability for a photodetector to transform the photonic signal into an electrical signal, which is calculated as the number of electron/hole pairs detected per incident photon, given by the following equation:

$$EQE = R h c / e \lambda, \quad (2)$$

where h is Planck's constant, c is the light velocity, e is the electron charge and λ is the excitation wavelength. In some situations, the value of EQE may be larger than 1 because the photo-generated carrier lifetime is longer than the transit time, and it will be calculated several times for one photo-generated carrier.

Noise equivalent power (NEP) and detectivity (D^*)

NEP is the minimum detectable power of a photodetector and D^* is inversely proportional to NEP. Both D^* and NEP represent the detection ability of a photodetector. In general, people prefer detectivity than NEP to express the property of a detector, which can be given by:

$$D^* = \frac{A^{1/2} R}{(2eI_d)^{1/2}}, \quad (3)$$

where A is the device area. In this equation, we can see a low dark current (I_d) and high responsivity will help to achieve a larger D^* .

Photoconductive gain (G)

For photodetectors, the photocurrent originates from the detected photons, so the photoconductive gain is described by:

$$G = \frac{\tau_{life}}{\tau_{trans}}, \quad (4)$$

where τ_{life} is the carrier's lifetime, and τ_{trans} is carrier's drift time. From the equation, we can see the G implies the number of carriers that a single incident photon excites. Long carrier life means high gain, but also longer response time.

On/off ratio

On/off is defined as the ratio of the open state current (I_{on}) to the off state current (I_{off}) of the device. For an ideal switch, the off current will be zero and the on current will only be limited by the external circuit.

DEVICES BASED ON DIFFERENT 2D MATERIALS

To date, devices based on various low-dimensional materials have been fabricated including graphene, TMDCs, elemental semiconductors (like BP, Te), main group metal chalcogenides (MMCs), thin film of unlayered materials and some heterostructures. These nanomaterials could cover the wide range of electromagnetic spectrum, showing many novel properties that traditional devices do not have. The new performances have attracted scientists' eyesight, and the new generation devices may play an important role in the future electronic and optoelectronic field. In this section, we will introduce these well-performed devices from the perspective of materials.

Graphene-based materials

Graphene is a single low-dimensional sheet of carbon atoms with a honeycomb lattice structure, in which its carbon atoms attach each other to form a hexagonal shape, and its layers combine *via* van der Waals interaction. The single-layer and few-layer graphene was firstly obtained by Novoselov *et al.* [1] *via* a scotch tape method, and then they took a closer look at the properties of graphene. As they reported, graphene has many good properties that not previously discovered in other semiconductor materials, such as zero bandgap, ultrahigh carrier mobility (about $10,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at room temperature, $200,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at low temperature), and high conductivity.

The pristine graphene-based optoelectronic devices

Due to its outstanding properties, quite a few scientists put a lot of effort into graphene, and among the vast number of low-dimensional materials, the studies of graphene are the most, which also have implication for other low-dimensional materials. In 2012, a graphene metal-oxide-semiconductor field-effect transistor (MOSFET) with an intrinsic cutoff frequency exceeding 400 GHz was successfully demonstrated [46]. The encouraging development misled scientists, so that graphene had been designated as the successor of Si in mainstream electronics. However, as the research of graphene continues, the graphene/Si heterostructure-based devices can only attain an I_{on}/I_{off} ratio of at most a few tens due to their zero bandgap, which limits the further application into integrated circuits. To solve this problem, scientists have designed many schemes to open a gap in graphene. Nevertheless, the as-made opened-gap graphene-based devices had worse performance than the primary ones. Therefore, to explore the graphene-based devices in a commercial use, there are a lot of problems to be solved in the near-to medium-term future.

A lot of graphene heterostructure-based photodetectors have been finished. Graphene-based optoelectronic devices present ultra-broadband photoresponse due to its zero bandgap. As early as in 2010, the IBM group designed a graphene/Si heterostructure-based photodetector [47], whose response range covered 514 to 2400 nm, and the as-made photodetector also had good performances with the maximum external photoresponsivity of 6.1 mA W^{-1} at a wavelength of $1.55 \mu\text{m}$. In a recent research, THz graphene-based detectors have been fabricated and they have a potential to be used in outer space exploration. Lara-Avil *et al.* [48] synthesized graphene sheets that were uniformly doped to the Dirac point and then were used to construct graphene/Si heterostructure-based photodetectors. With material resistance dominated by quantum localization and thermal relaxation governed by electron diffu-

sion, the photodetectors showed highly sensitive and wide-band coherent detection of signals from 90 to 700 GHz and maybe across the entire terahertz range. Then researchers conducted THz mixing experiments, a gain bandwidth of $f_0 = 8$ GHz was demonstrated. And at a low temperature (36 K), the gain bandwidth would exceed 20 GHz. These breakthroughs in detector may accelerate the development of heterodyne astronomy. And for some photodetectors, graphene could be an excellent electrode material due to its high conductivity and transparency [49].

The improvements for graphene-based devices

For most graphene/Si heterostructure-based devices, they have high leakage current noise, relatively high dark currents and low light absorption, which limit the further development of high-performance photodetectors. To reduce the dark current, researchers have tried some ways, one of which is to introduce a small gap to suppress the dark current, for that a small gap functioned like a quantum well helps suppress the dark current. For example, Ryzhii *et al.* [50] designed several prototype devices to improve the performances of graphene-based photodetectors. The first design was based on bilayer graphene in which the energy gap can be changed by the external electric field with a maximum detectivity over 10^{11} Jones. The second one was based on tailored graphene nanoribbons [51] in which the band gap can be changed by controlling the width of nanoribbons with a responsivity of about 6.4 A W^{-1} . Based on these two prototypes, they developed a variety of high-performance THz infrared graphene detectors in recent years [52,53].

On the other hand, a p-i-n structure by introducing a thin interfacial insulative layer is a good way to improve the band structure of graphene/Si heterostructure. Li *et al.* [54] reported a design that introduces an interfacial oxide layer between the graphene and n-silicon; the dark current of graphene/n-Si heterojunction has been reduced by two orders of magnitude at zero bias. At room temperature, the graphene/n-Si photodetector with interfacial oxide exhibits a specific detectivity up to 5.77×10^{13} Jones with a responsivity of 0.73 A W^{-1} as well as a high on/off ratio of $\sim 10^7$. Fig. 2a–d show the p-i-n structure and its effect on dark current suppression. In a latest report [11], a thin layer of HfO_2 was introduced in the graphene/Si heterostructure to enhance the interfacial gating effect. The as-made graphene/ HfO_2 /Si heterostructure photodetectors attained better performances with a broad response range from 473 to 1150 nm, a high responsivity of 45.8 A W^{-1} , fast response speed of $\tau_{\text{rise}} = 9 \mu\text{s}$, $\tau_{\text{decay}} = 20 \mu\text{s}$. Fig. 2e, f show the schematics and photo-response characteristics of the graphene/ HfO_2 /Si photodetector.

Decorating metal plasmonic nanoparticles (arrays) on the surface of graphene is a good way to improve the photocurrent. The plasmonic nanoparticles could enhance the field on the surface of metal, leading to an effective improvement to the photo gain of the photodetectors. Ferrari and coworkers [55] fabricated a plasmonic-enhanced metal/single-layer graphene (SLG)/Si Schottky photodetector with a responsivity of 0.37 A W^{-1} (operating at $1.5 \mu\text{m}$) and an internal quantum efficiency (IQE) of 7% (see Fig. 3a, b). To improve the response speed, Selvi *et al.* [56] investigated a graphene-silicon-on-insulator (GSOI) Schottky junction. In this junction, the thickness of

Figure 2 p-i-n structures to suppress dark current. (a) Schematic view of a graphene/n-Si heterojunction photodetector with an interfacial oxide layer. The silicon wafer with a 300-nm SiO_2 layer was patterned by photolithography and wet-etching of thermally grown oxide to prepare square windows. The energy-band diagrams of the heterojunction of (b) graphene/n-Si and (c) graphene/ SiO_2 /Si. ϕ_G and $\phi_{\text{n-Si}}$ are the work functions of graphene and n-Si, respectively; ϕ_b is the ideal barrier height. V_{bi} is the built-in potential barrier. V_{ox} is the potential drop across the oxide layer. δ is the thickness of oxide layer. E_g is the band gap of silicon (1.12 eV) and E_F is the energy of the Fermi level. (d) Oxide-thickness-dependent response of the heterojunction photodetectors. (a–d) are reprinted with permission from Ref. [54]. Copyright 2015, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim. (e) Schematic view of the graphene/ HfO_2 /Si photodetector with a pair of 10/50 nm Ti/Au source-drain electrodes. Incident photons create electron-hole pairs in Si. These photogenerated carriers are separated by the built-in field and the accumulation of the holes at the HfO_2 /Si interface gates the graphene through capacitive coupling. (f) Transfer curves of the photodetector at various light powers. The inset is the optical image of the photodetector. (e, f) are reprinted with permission from Ref. [11]. Copyright 2021, American Chemical Society.

Figure 3 (a) Schematic M-SLG-Si Schottky photodetector. SOI: silicon-on-insulator. BOX: buried oxide. (b) Finite element simulated optical intensity profile of a surface plasmon polariton waveguide mode supported by an M-SLG-Si structure. (c) Schematic image of an on-chip photodetector structure. (d) Normalized temperature profiles for a varying chemical potential for (d) high-disorder (chemical potential for $\Delta = 60$ meV) and (e) low-disorder (chemical potential for $\Delta = 40$ meV) graphene sheets. (a, b) are reprinted with permission from Ref. [55]. Copyright 2016, American Chemical Society. (c–e) are reprinted with permission from Ref. [8]. Copyright 2021, Optical Society of America.

the photo-active silicon layer was reduced to 10 μm , suppressing the rate-limiting diffusion currents, so that significant speed improvements were obtained in both a GSOI grating device and a GSOI planar device. The rise and decay times of the former were 4 and 10 to 15 ns, while those for the latter were 10 and 20 to 70 ns, respectively. Recently, AlAloul and Rasras [8] used CMOS-compatible plasmonic metal TiN to enhance the responsivity of graphene detector on Si substrate (as shown in Fig. 3c). The as-mentioned TiN/graphene/Si heterostructure not only shows a high responsivity of 1.4 A W^{-1} , but also shows a fast response speed for that its bandwidth exceeds 100 GHz. In this device, the photodetection process is predominantly determined by the PTE at zero bias, and the PTE causes a very transient Fermi-Dirac distribution of hot carriers, and they decided to use a chemical potential and a carrier temperature to describe the photodetector. The temperature profiles are shown in Fig. 3d, e.

Further exploration to graphene

On the one hand, few-layered graphene-based devices show excellent performances. Table 1 summarizes the performances of some representative devices mentioned in this paper. On the other hand, researchers have found some interesting phenomena in bilayer graphene. For bilayer graphene-based devices, a special stacking method called Bernal stacking could achieve a field-induced band gap which could improve the properties of graphene. In Bernal stacking bilayer graphene, half of the atoms of one layer are located exactly above the atoms of the second layer, while the other half of the atoms lie precisely above the empty center of the hexagons of the other layer. According to the theoretical calculation of Iannaccone *et al.* [57], the field-induced gap is limited to about 0.15 eV, which leads to a high

on/off ratio (about 100). What's more, the twisted angle has a great influence on the properties of the bilayer graphene, Cao *et al.* [58–61] have published a series of papers on magic-angle twisted graphene. They fabricated a small-angle twisted bilayer-bilayer graphene (TBBG) structure which consists of two sheets of Bernal-stacked bilayer graphene. Then they further studied the phases and states in the twisted bilayer graphene system [59]. The TBBG showed a rich phase diagram and tunable correlated insulator states. These insulator states exhibited high sensitivity to the twist angle and the application of electric displacement field, the latter reflecting the inherent polarizability of Bernal-stacked bilayer graphene. Furthermore, this research can also provide a guide to the study of electronic correlations in other twisted van der Waals heterostructures.

Graphene is now extensively utilized for electrode materials of supercapacitors because of its high conductivity and resistance to corrosion and the graphene heterostructure-based devices have also exhibited good performances, such as broad photo-response range, high responsivity, and ultrafast response. Different graphene-based devices have been widely used in various fields. However, we can see from the above part, these devices have many disadvantages, e.g., high-speed devices present weak photoelectric performances, while the devices with high optoelectronic properties exhibit poor response speed. More importantly, how to prepare large-scale graphene with high quality is an unavoidable problem on its road to commercial application. Improving the CVD method to prepare large-area graphene will be a development trend in the future. On the other hand, Cao's several articles [58–61], published in *Science* and *Nature*, show that graphene still has great potential to probe. Constructing nanostructure of graphene may be a research direction to explore its miraculous properties.

Table 1 Comparison of the critical parameters for graphene-based photodetectors

Devices	Wavelength (nm)	Responsivity (A W^{-1})	Detectivity (Jones)	Response time (μs)	Ref.
Graphene/ SiO_2/Si	514–2400	6.1 mA W^{-1}	–	Less than 60 ps	[47]
Bilayer graphene/ SiO_2/Si	THz	4.6	10^{11}	Fast	[50]
Nanoribbon graphene/ SiO_2/Si	THz	6.4	–	Fast	[51]
Graphene/oxide/n-Si	410–950	0.73	5.77×10^{13}	320/750	[54]
SLG/Si Schottky	514,1550	0.37	10^{12}	Fast	[55]
Graphene/ HfO_2/Si	473–1150	45.8	–	9/20	[11]
Graphene/ HfO_2/Ge	473–1550	9.03	–	11/38	[11]
Graphene/ $\text{HfO}_2/\text{a-MoS}_2$	473–2712	5.36	10^9	68/89	[11]
TiN-graphene/Si	1550	1.4	5×10^{10}	Less than 10 ps	[8]
SLG/Ge Schottky heterojunction	–	51.8 mA W^{-1}	1.38×10^{10}	–	[62]
GSOI	<1100	0.26	7.83×10^{10}	Several ns	[56]

TMDC-based devices

The common chemical formula of TMDCs is MX_2 , where M is a transition metal ($\text{M} = \text{Mo}, \text{W}, \text{Re}$, and so on) and X is a chalcogen ($\text{X} = \text{S}, \text{Se}, \text{Te}$). TMDCs possess a layered crystal structure in which the individual layers are bonded together *via* weak van der Waals interactions. TMDCs have many excellent properties, such as favorable flexibility and high mobility. TMDCs have a very similar structure to graphene and were considered as a substitute of graphene. Although TMDC materials have a lower mobility than graphene, they have a tunable gap and a higher light absorptivity [63,64]. Hence, TMDCs may be more suitable materials for optoelectronic devices compared with graphene. In fact, different TMDC materials have different characteristics. In this section, hence, we will introduce basic properties of some TMDC materials, followed by versatile applications. And as a most typical TMDC material, MoS_2 will be the focus of a large section.

MoS_2 -based devices

As a typical low-dimensional layered TMDCs, MoS_2 has been extensively studied due to its exciting properties. MoS_2 is a typical isotropic crystal whose bandgap is thickness-dependent. The bandgap of bulk MoS_2 is 1.2 eV which is an indirect gap, while single-layer MoS_2 is a direct-gap material with a bandgap of 1.8 eV [65]; this phenomenon attract scientists' interest and they focus more attention on few-layer MoS_2 -based devices. For example, Wang *et al.* [24] reported the construction of few-layer MoS_2/Si heterojunction-based photodetectors. The MoS_2/Si device showed a wide photoresponse range of 450–1050 nm, ultrafast response speed of 3 μs , and high detectivity of 10^{13} Jones. Although MoS_2 -based photodetectors have excellent performances, the intrinsic bandgap of MoS_2 (whose cutoff wavelength edge is about 1 μm) limits the detection range of the photodetectors. The deviation from the perfection of the atomic lattice, which can effectively tune the bandgap and response range, may provide an appropriate way to broaden the detection range of the MoS_2 photodetectors. In 2016, Xie *et al.* [66] fabricated an ultrabroadband multilayer MoS_2/Si photodetector by controlling the defects of S atoms. They designed MoS_2 with an element ratio of S/Mo ions of 1.89, and found the bandgap was decreased to 0.26 eV in a theoretical calculation, and the cutoff wavelength could reach about 4 μm . The as-fabricated photodetectors showed a wide response region up to 4.7 μm , and were

investigated in the wavelength range from 445 (blue light) to 2717 (mid-infrared (MIR)) nm. At a cost, the responsivity and detectivity dropped to 50.7 mA W^{-1} and 1.55×10^9 Jones, respectively. However, due to the lack of high-quality p-n junction and large wafer scale fabrication process, the MoS_2 -based photodetectors are limited for commercial applications. Recently, large-scale preparations for TMDC/Si heterostructure-based devices have been realized. Dhyani *et al.* [20] successfully fabricated a large-area MoS_2/Si heterostructure-based photodetector with a simple, reproducible and scalable fabrication process. This photodetector was compatible to CMOS circuits and had excellent performances, such as high responsivity (up to 8.75 A W^{-1}), high detectivity (about 1.4×10^{12} Jones) and fast response time (about 10 μs), and the transient measurements of the phototransistors showed excellent stable and reproducible dynamic response to high-frequency laser.

On the other hand, Wang *et al.* [67] successfully demonstrated an ultrasensitive and broadband photodetector in another way. In this device, they made pioneering use of poly(vinylidene fluoride-trifluoroethylene) (P(VDF-TrFE)), an organic ferroelectric material, and the remnant polarization of P(VDF-TrFE) provided a high local electrostatic field, making the few-layer MoS_2 channel a fully depleted state which significantly increased the sensitivity of the detector. Possessing these novel properties, the photodetector attained a responsivity of up to 2570 A W^{-1} , along with the detectivity of 2.2×10^{12} Jones, as well as a broad response range of 450–1550 nm. It is worth noting that the $\text{MoS}_2/\text{P(VDF-TrFE)}$ heterostructure exhibited pronounced photovoltaic activity, enabling their applications as self-driven photodetectors operated at zero bias voltage. It means that the low energy-cost photodetectors can be considered for applications in portable and flexible devices. This successful sample provides a new way, utilizing hybrid heterostructures and nanocomposites, to boost the performances of MoS_2 photodetectors. In fact, diverse MoS_2 hybrid heterostructures could increase carrier mobility and incident light absorption. For example, Xiao *et al.* [68] reported a 3D reduced graphene oxide (RGO)- MoS_2 /pyramid Si heterostructure-based photodetector which was made by a simple solution-processing method. Fig. 4 shows a schematic illustration of the structure of the 3D heterostructure-based photodetector, current-voltage characteristics of the device, a comparison of absorption spectra of planar and pyramid Si, RGO, and RGO- MoS_2/Si heterojunction devices and

Figure 4 (a) Schematic illustration of the RGO-MoS₂/pyramid Si heterojunction-based photodetector. (b) A comparison of the absorption spectra of planar and pyramid Si, RGO, and RGO-MoS₂/Si heterojunction devices. (c) Photosensitivity of the RGO-MoS₂/pyramid Si heterojunction-based device in the wavelength region between 350 and 1100 nm. (d, e) Photocurrent switching behavior of the heterojunction device measured under light illuminations at (d) 1310 and (e) 1550 nm at zero bias voltage. (f) Photocurrent switching behavior of the heterojunction device measured under 1870, 1940, 2200, 3460 and 4290 nm (near-infrared (NIR)-MIR) light illuminations. (g) Photoresponsivity and specific detectivity curves of the heterojunction device measured as a function of wavelength from 1310 to 4290 nm at a fixed laser power intensity of 50 mW under zero bias voltage. (h) R and D^* curves of the heterojunction photodetector measured as a function of laser power intensity at 808 nm under zero bias voltage. (i, j) Comparison of the photocurrent switching behavior of RGO-MoS₂/pyramid Si and RGO-MoS₂/planar Si heterojunction devices measured at (i) 808 nm, 1 mW laser power intensity, and (j) 1550 nm, 10 mW laser power intensity. (k) A comparison of the wavelength coverage by the RGO-MoS₂/pyramid Si heterojunction-based photodetector along with other traditional semiconductor-based photodetectors. The RGO-MoS₂/pyramid Si hybrid heterojunction-based photodetectors operated from 350 to 4.3 μm (UV to MIR) ultra-broadband spectral range due to the bandgap narrowing caused by the S vacancy defects in MoS₂ crystals. (a-j) Reprinted with permission from Ref. [68]. Copyright 2018, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim. (k) Reprinted with permission from Ref. [69]. Copyright 2020, The Royal Society of Chemistry.

diverse performances of the 3D heterostructure photodetector. From the figure, we can see, at zero bias voltage, the photo-voltage of RGO-MoS₂/Si heterostructure photodetector increases with the enhancement of incident light power intensity, indi-

cating the self-powered operation of the photodetector. Furthermore, this 3D heterostructure-based photodetector exhibited excellent performances because of the special structure, such as an extreme broad spectrum range from ultraviolet (UV)

(350 nm) to MIR (4300 nm); moreover, a very high detectivity of 3.8×10^{15} Jones and a photoresponsivity of 21.8 A W^{-1} were achieved at the illumination of 808-nm incident light.

With the continuous study of MoS_2 , researchers found MoS_2 possesses three phases: 2H- MoS_2 , 3R- MoS_2 and 1T- MoS_2 , where H, R, and T represent hexagonal, rhombohedral and tetragonal symmetry, respectively, and “1”, “2”, and “3” represent the number of S single bonds, Mo single bonds, and S layers within a single unit cell, respectively [70]. Different phases have different structures and properties. Fig. 5a–f show crystal structures and Raman spectra of the three phases and the other MoX_2 have similar structures. Common semiconducting MoS_2 in nature is 2H phase, while the metallic 1T- MoS_2 is metastable and can be synthesized by utilizing the interaction of alkali metal ions. What we have discussed in the previous part is also 2H MoS_2 [71]. As we can see from the previous part, the 2H- MoS_2 -based photodetectors have the disadvantage of low photoresponse which

severely limits their broader applications in optoelectronic fields. The metallic 1T- MoS_2 [72] shows ultrafast response time which complements 2H- MoS_2 very well. Recently, Wang *et al.* [73] fabricated vertically stacked 2H- MoS_2 /1T@2H- MoS_2 /Si photodetectors which integrated the positive traits of 2H- MoS_2 and 1T- MoS_2 . The hybrid MoS_2 /Si heterostructure showed fewer dangling bonds and better lattice matching which ensured an excellent performance with responsivity of 1917 A W^{-1} , detectivity of 7.55×10^{11} Jones, and EQE of 448,384% at 532 nm wavelength light illumination with gate bias of 20 V. The very high EQE value is caused by Schottky barrier modulation. As a comparison, the maximum R of 1T@2H- MoS_2 -based photodetector is about 10^{-4} A W^{-1} which is even lower than that of the 2H- MoS_2 -based transistor (maximum value of 56 A W^{-1}), indicating that the 1T@2H- MoS_2 layer only provides high-speed carrier and the effect of the 1T@2H- MoS_2 layer is negligible in the photodetection performance. The schematic diagram is show

Figure 5 (a) 2H, (b) 3R, and (c) 1T phases of MoS_2 . Crystals viewed along the a -axis, where the unit cell is shown as a gray box (3R rhombohedral unit cell shown in red). (a–c) are reprinted from Ref. [74]. Copyright 2021, American Chemical Society. (d) Raman spectrum of 2H MoS_2 . ML: monolayer; FL: few-layer. Reprinted from Ref. [75]. Copyright 2021, American Chemical Society. (e) Raman spectrum of 3R MoS_2 . Reprinted from Ref. [76]. Copyright 2017, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (f) Raman spectrum of 1T MoS_2 . Reprinted from Ref. [77]. Copyright 2018, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim. (g) 3D graphic demonstration showing the typical photodetector structure on the left. The zoomed-in region in the red box on the right presents transported electrons moving in different layers. (h) 3D view of photoresponsivity mapping for this photodetector under different gate bias and source-drain bias values, at an illumination power density of 2.35 mW cm^{-2} . (i) Schematic band diagram of the semiconducting 2H- MoS_2 and metal-like 1T@2H- MoS_2 diodes. (g–i) are reprinted from Ref. [73]. Copyright 2020, American Chemical Society.

in Fig. 5g.

Other Mo- and W-dichalcogenide-based devices

Unlike MoS₂, MoSe₂ is a semiconducting material with an indirect bandgap of 1.1 eV in bulk form, which increases gradually to 1.55 eV and becomes direct bandgap in monolayer and bilayer. The monolayer MoSe₂-based FET shows an n-type behavior and its average mobility is about 50 cm² V⁻¹ s⁻¹, while the mobility of bulk MoSe₂ is about 100 cm² V⁻¹ s⁻¹ [78,79]. In 2019, a MoSe₂/p-Si junction was successfully demonstrated for position-sensitive detector [80]. The MoSe₂/Si showed a lateral PVE (LPE) which exhibited a good linear dependence on the laser spot position. When there was a small displacement of the laser spot, the LPE caused a relative change in the corresponding current. As a consequence, the MoSe₂/p-Si junction showed a large positional sensitivity of 563 mV mm⁻¹. Moreover, MoSe₂ has a higher optical absorption and a narrower bandgap; hence, the MoSe₂/Si is also a good alternative used for infrared detection. Jie's group [81] fabricated graphene/MoSe₂/Si photodetectors. In this heterostructure, the graphene layer was used

for transparent electrode to collect carriers. Fig. 6a, d and e show the structure of the detector and the carrier collecting effect of graphene transparent electrode. The as-mentioned photodetector was sensitive to the light from 365 to 1310 nm with ultrafast response (about 0.3 μs for both raise and fall time). Meanwhile, the detectivity and responsivity of the photodetector were 7.13×10^{10} Jones and 0.27 A W⁻¹, respectively. Ko *et al.* [82] introduced a SiO₂ layer to few-layered MoSe₂/Si heterostructure to form a back-gated phototransistor. The photoresponsivity could be easily controlled to reach a maximum value of 238 A W⁻¹ under NIR light excitation, while achieving a high specific detectivity $D^* = 7.6 \times 10^{11}$ Jones, a high EQE of 37,745%. In a recent report, Xu *et al.* [40] synthesized a stacked layered MoSe₂ on an n-type Si substrate. The layered MoSe₂ was uniform and with high quality and the device exhibited excellent performances. The n-n MoSe₂/Si heterojunction showed a broad response spectrum up to 1500 nm. The responsivity and detectivity can reach up to 0.58 A W⁻¹ and 1.4×10^{14} Jones, respectively. These performances are much better than those of most previous MoSe₂-based photodetectors which are mainly deter-

Figure 6 (a) Schematic illustration of the graphene/MoSe₂/Si heterojunction photodetector with graphene transparent electrode. (b) Schematic and photograph of the WS₂/p-Si heterojunction device. (c) The photograph of WS₂/Si array device. Simulated total current density distributions of the MoSe₂/Si photodetectors (d) with and (e) without graphene top electrode at forward bias of +1 V, respectively. (f) The band diagram of WS₂/Si heterostructure. (g) Absorption spectra of Ag-WS₂ nanocomposite compared with WS₂. (h) Normalized photocurrent of the WS₂/Si heterojunction as a function of the polarization angle. (i) Spatial resolved photocurrent mapping of a 4 × 4 heterojunction device array. (a, d, e) are reprinted with permission from Ref. [81]. Copyright 2016, The Authors. Published by WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (b) is reprinted with permission from Ref. [84]. Copyright 2018, American Chemical Society. (c, f, h and i) are reprinted with permission from Ref. [85]. Copyright 2019, American Chemical Society. (g) is reprinted with permission from Ref. [86]. Copyright 2020, Elsevier B.V.

mined by the high-quality MoSe₂ film. In addition, the response speed approaches 0.12 μ s, meaning the device may be used in high-speed circuits. Almost the same time, John *et al.* [83] fabricated a MoSe₂/Si p-n junction photodiode whose spectral range covered 405 to 2500 nm, and the maximum responsivity of 522 mA W⁻¹ was attained at the illumination of 1100 nm incident light. The MoSe₂ nanosheets were synthesized by a hydrothermal method, causing many defects in the lattice which was the main reason for high optical absorption of the short-wave infrared light. Furthermore, the response speed of the devices was also fast, for that the rise time was 18 μ s and the fall time was 14 μ s in the infrared region.

MoTe₂ has an indirect bandgap of 1.0 eV and a high carrier mobility of 200 cm² V⁻¹ s⁻¹ in its bulk form, which also has strong light absorption so that it is usually used for electrode materials in photovoltaic cells; while in its monolayer form, MoTe₂ has a direct bandgap of 1.07 eV. In 2016, Octon *et al.* [87] observed a fast photoresponse time (~160 μ s) in few-layered MoTe₂-based photodetectors with a high responsivity of 6 A W⁻¹. However, this MoTe₂-based photodetector has a narrow spectral range. Recently, Lu *et al.* [88] fabricated a few-layer MoTe₂/Si vertical heterojunction which has broad response band (from 300 to 1800 nm) and a very fast response speed. The n-type MoTe₂ nanosheet was synthesized on an n-type Si substrate *via* a pulsed laser deposition (PLD) method, and graphene was selected as the transparent electrode to ensure an ohmic contact with MoTe₂ and a high optical absorption. The n-n heterostructure photodetector exhibited a responsivity of 0.19 A W⁻¹, a large detectivity of 6.8×10^{13} Jones and a high current on/off ratio of 3.2×10^6 at 980 nm. It is noted that the device possesses an ultrahigh speed of a rise time of 150 ns and a fall time of 350 ns. They used laser with different pulse frequencies (100 Hz–5 MHz) to illuminate the device and recorded the signal. The 3-dB optical bandwidth was estimated to be up to 1.0 MHz and the electrical bandwidth was calculated to be 0.12 GHz. The ideality factor (*n*) of the MoTe₂/Si heterojunction was 1.9, in contrast to the much larger value of 3.5 for graphene/Si Schottky junction. In addition, the turn-on voltage of the device was 0.15 V and the saturation current was 20 μ A at a reverse voltage of -10 V. Just like MoS₂, MoTe₂ also has two common phases: 1T phase and 2H phase, while 1T-MoTe₂ is metallic and metastable. In a latest study, Lin *et al.* [89] inserted 1T-MoTe₂ into 2H-MoTe₂ to improve the photoresponse properties of MoTe₂-based photodetector. The 1T-MoTe₂ was introduced to enhance the interlayer reaction of the MoTe₂ nanosheet. The responsivity was enhanced about 10 times compared with single 2H-MoTe₂-based photodetector, and the response time decreased from about 190 to about 90 μ s.

WX₂ materials have similar properties to MoX₂, and many researchers study these materials from different perspectives to dig inner details of them. For example, Zheng *et al.* [90] tried to combine few-layered WSe₂ with different substrates (including Si/SiO₂, polyimide (PI) and glass) to develop different heterostructures. The WSe₂ thin films were prepared on various substrates by using PLD and indium tin oxide (ITO) was deposited as transparent electrodes. And the response range of the as-fabricated devices was 370–1064 nm, while the responsivity could reach up to 0.92 A W⁻¹, along with the maximum EQE of 180%. Also, Pataniya's group [91] designed a WSe₂/Si heterojunction, with an ideality factor of 1.21 and a faster response time of 32 ms (compared with Zheng's 0.9 s).

The WS₂/Si heterojunction also exhibited excellent performances. In 2018, Kim *et al.* [84] fabricated a WS₂/p-Si vertical heterostructure. This photodetector showed a broad response range which covers from UV to NIR (360–850 nm) and an ultrafast photoresponse (1.1 μ s) along with a specific detectivity of 1.5×10^{10} Jones; however, the responsivity is low (4 mA W⁻¹). It is worth noting that the area of WS₂ is very large to wafer scale along with ultrahigh response speed, indicating a potential application in the integrated circuits design. In a latest report, Ag nanoparticles were decorated on WS₂ nanosheets *via* an LPE to form a metal/TMDCs nanocomposite [86]. Just like the TiN/graphene heterostructure, the Ag/WS₂ structure would provide an improved plasmonic effect which can enhance the light absorption and photodetection ability of the device. Fig. 6g shows the enhanced light absorption of the nanocomposite. The as-made Ag-WS₂/Si exhibited a higher responsivity of 2.09 A W⁻¹, a specific detectivity of 6.6×10^{11} Jones, and a high EQE of 487%. In a recent study, Wu *et al.* [85] extended the detection range of WS₂/Si heterojunction detector to MIR (Fig. 6c). They explained the interlayer excitation was generated by the small gap between valence band maximum of Si and conduction band minimum of WS₂ (Fig. 6f). Therefore, the detectors could be sensitive to MIR light with a wavelength below the bandgaps of WS₂ and Si for that the detector would be sensitive to the 3043 nm infrared light. In addition, they made a 4 × 4 array to enhance the optoelectronic performance of the detector (Fig. 6i). Moreover, the photodetector exhibit a high *I*_{on}/*I*_{off} ratio of >10⁶. In addition, the WS₂/Si heterojunction shows sensitivity to polarized light and a high peak-to-valley ratio of 2.8 (Fig. 6h).

Group-ten dichalcogenide-based devices

In recent years, more and more studies of group-ten TMDCs are emerging due to their unique structures and electrical properties [92–94]. For group-ten TMDCs/Si heterostructures, MSe₂ (M = Pd, Pt) is more commonly used to construct the hybrid structures. For example, researchers directly grow PdSe₂ film on Si and pyramid Si substrates to form heterojunctions, respectively [95,96] (see Fig. 7a, b). These two heterojunctions exhibit similar performances and some parameters may be enhanced by the structure of pyramid Si. Fig. 7c shows the electric field energy distribution of the PdSe₂/pyramid-Si. The responsivity of PdSe₂/pyramid-Si is about 450 mA W⁻¹ compared with the PdSe₂/Si's 300 mA W⁻¹, and the detectivity of PdSe₂/pyramid-Si is also a little higher. The on/off ratios of them are both at a 10⁵ magnitude. It is noted that the PdSe₂/Si heterostructure detector has an ultrabroad response range covering from deep ultraviolet (DUV) (200 nm) to MIR (3044 nm), while the PdSe₂/pyramid-Si has a successful trail on infrared imaging (see Fig. 7d–f). Aftab *et al.* [97] combined n-PdSe₂ with p-type Si substrate by an ME technique. For fine electrodes, e-beam lithography was performed to produce Al₂O₃ (25 nm thick) films. The as-fabricated photodetector showed higher responsivity of 47.2 A W⁻¹ and lower detectivity of 8.13×10^9 . These two methods to make heterostructures cause different results, which may be due to the inconsistent crystal quality and the degree of bonding between the PdSe₂ films and the substrates.

PtSe₂ has a prominent layer-dependent electronic behavior, i.e., the monolayer PtSe₂ is semiconducting while the bulk form is semi-metallic. In a latest report, the wafer-scale [98] PtSe₂ film was directly grown on Si substrate to form a photocathode *via* a

Figure 7 Group-ten TMDCs-based devices. (a) Schematic illustration of PdSe₂/Si detector. Reprinted with permission from Ref. [95]. Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (b) Schematic diagram of the PdSe₂/pyramid Si with a wavelength of 980 nm. (d) Digital photograph of the template “tree” and “house”. The corresponding 2D current mapping of image: (e) tree under the illumination of 980 nm light, (f) house under the illumination of 1300 nm light. (b–f) are reprinted with permission from Ref. [96]. Copyright 2019, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (g) The energy band diagram of p-Si, PtSe₂ thin films with a thickness of 0.8, 1.4, 2.2, and 3.0 nm. The 4L (3.0 nm) PtSe₂ is semimetallic. Reprinted with permission from Ref. [98]. Copyright 2021, American Chemical Society. (h) The digital photograph of the PtSe₂/Si flexible device. Reprinted with permission from Ref. [99]. Copyright 2019, American Chemical Society.

CVD method. This as-made photocathode exhibited an outstanding conversion ability for direct solar-to-hydrogen (H₂) production due to the photoelectrochemical conversion. And the produced H₂ would protect the heterostructure substrate from oxidation. What's more, they also demonstrated the atomic-layer-dependent band alignments at the PtSe₂/Si interface. They found and figured out the work function of PtSe₂ with different thicknesses and then constructed corresponding band diagrams for the PtSe₂/Si heterostructure (as shown in Fig. 7g). In addition, the PtSe₂/Si heterostructure is also reported to be used for flexible devices in another study [99]. The as-mentioned flexible device has not only excellent Schottky junction properties, such as high-current rectification ratio ($>10^3$), small ideality factor ($n \sim 1.9$), and temperature-dependent variation of Schottky barrier heights, but also good anti-deformation characteristics—after hundreds of bends, the photovoltaic characteristics of the device are still the same as before. Fig. 7h shows the flexible photovoltaic 2D PtSe₂/Si devices.

Re-dichalcogenide-based devices

ReX₂ also have good performances especially in polarization detection. Liu *et al.* [100] demonstrated a ReS₂ polarization sensitive photodetector on a Si/SiO₂ substrate. The detector showed an excellent polarization property with high responsivity of $\sim 10^3$ A W⁻¹ and on/off ratio of 10^5 . Different from previous photodetectors, this ReS₂/SiO₂/Si photodetector did not require the use of extra components (gratings etc.) due to the anisotropic nature of the ReS₂. In fact, the performances of p-n junction made by n-ReS₂/p-Si are related to the thickness of ReS₂; the p-n heterojunction with thin ReS₂ layer showed weak photosensing performance but fast response, while the p-n junction with thick multilayer ReS₂ showed a larger photocurrent with an overall degradation of the response time. A mixed-thickness ReS₂/Si showed a balance between responsivity and speed, with a high photoresponsivity of 33.47 A W⁻¹ and a fast response time of 80 μ s [101]. In addition, the TMDC/TMDC heterostructures were also realized for ReS₂. Ahn *et al.* [102]

Figure 8 (a) Schematic illustration of the linear polarization-sensitive photodetection; (b) optical microscopy of MoTe₂/ReS₂ heterojunction; (c) energy level diagram of ReS₂/MoTe₂ van der Waals heterojunction, indicating the clear type-II band alignment. Dark and photoinduced current-voltage characteristics of a photodiode in (d) logarithmic scale and (e) linear scale under NIR (1310, 980, and 808 nm), red (638 nm), and green (532 nm) light illuminations. (f) Two different mode responsivities as a function of illumination wavelength. (g) Dark and photoinduced current-voltage characteristics under different laser diode (808 nm) illumination powers. (h) Photocurrent versus optical power density. (i) Plot of linear polarization-sensitive normalized photocurrent changes in polar coordinates. Reprinted with permission from Ref. [102]. Copyright 2021, American Chemical Society.

fabricated a MoTe₂/ReS₂ heterostructure for a high performance photodiode with an additional function of linearly polarized photodetection (as shown in Fig. 8). The heterostructure photodiode shows an ideality factor of 1.3, a rectification ratio of 10⁴, broad spectral response up to NIR light of 1310 nm, and relatively high responsivity of 0.54 A W⁻¹. There is no existing report about the ReSe₂/Si and ReTe₂/Si devices, though these two kinds of Re-dichalcogenide-based devices may possess more interesting performances.

Brief summary

Research activities of typical TMDC material-based photodetectors are emerging on a fast pace. This review only discussed some typical TMDC materials. Table 2 summarizes the performances of some representative TMDC-based devices mentioned in this paper. However, the large family of low-dimensional materials can be explored for developing more heterostructure photodetection devices. For example, other low-dimensional TMDCs such as TiX₂, CoX₂, NiX₂, SnX₂, NbX₂, TaX₂ (X = S, Se, Te) [103–108], can be explored for developing new high-performance photodetectors. TMDCs are also advancing as multifunctional materials for future wearable electronic and optoelectronic devices as an inexpensive alternative to the costly manufactured semiconducting materials currently employed in the electronics industry. Low-dimensional atomic layered material-based hybrid van de Waals heterostructures hold great potential for self-powered flexible photodetectors in the healthcare industry especially for biomedical imaging, point-of-care wearable biosensors for monitoring vital signs and meta-

bolic parameters, and for patients. In addition, TMDCs are very promising nanomaterials for developing high-performance flexible photodetectors and for integration of such devices into the next generation of wearable technologies. The integration of cross-dimensional heterostructure TMDC photodetectors with other electronic, photonic and optoelectronic functional devices remains challenging.

Black phosphorus devices

Besides graphene and TMDCs, BP is another kind of low-dimensional material that attracts much attention. BP is a rare allotrope of phosphorus with direct bandgap and its layers stack together *via* van der Waals interaction. The bandgap of BP is thickness-dependent, which decreases from 1.7 eV (for monolayer BP) to 0.3 eV (for bulk BP) [109], covering the band from visible to MIR, just filling the gap of graphene and TMDCs. BP also has tunable bandgap and moderate switching ratio, good regulation and other excellent characteristics. For the perspective at a atomic level, each phosphorus atom is connected with three phosphorus atoms, causing BP crystal to form two special directions: the armchair and the zigzag directions along the *x* and *y* axes, respectively. So BP has prominent in-plane anisotropy. This special structure brings anisotropic electronic band dispersion, hence bringing anisotropic electronic and optoelectronic properties. In addition, for single/few-layered BP, the hole mobility can reach as high as 1000 cm² V⁻¹ s⁻¹. The most unique property of BP is the strong in-plane anisotropy. In its D_{2h} point group orthorhombic crystal system, the carriers along the longitudinal sawtooth pattern is ten times more effective than

Table 2 Comparison of the critical parameters for some TMDC-based photodetectors

Device structure	Wavelength (nm)	Responsivity ($A W^{-1}$)	Detectivity (Jones)	Response time (μs)	Ref.
Few-layer MoS_2/Si heterojunction	450–1050	0.3	10^{13}	3	[24]
MoS_2 (S defects)	445–4700	5.07×10^{-3}	1.55×10^9	–	[66]
p-Si/n- MoS_2 heterojunction	350–950	8.75	1.4×10^{12}	10/19	[20]
$MoS_2/P(VDF-TrFe)/SiO_2/Si$	450–1550	2570	2.2×10^{12}	1.8×10^3	[67]
3D RGO- MoS_2 /pyramid-Si	350–4500	21.8	3.8×10^{15}	–	[68]
2H- $MoS_2/1T@2H-MoS_2/SiO_2/Si$	Green light	1917	$\sim 10^{11}$	–	[73]
Graphene/ $MoSe_2/Si$	365–1310	0.27	7.13×10^{10}	0.3	[81]
$MoSe_2/SiO_2/Si$	785	238	7.6×10^{11}	–	[82]
p- $MoSe_2/n-Si$ junction	405–2500	0.52	6.7×10^9	18/10	[83]
$MoTe_2$	685	6	–	160	[87]
1T- $MoTe_2/2H MoTe_2$	980	–	–	99	[89]
n- $MoTe_2/n-Si$ junction	300–1800	0.19	6.8×10^{13}	0.15	[88]
WSe_2/ITO	370–1064	0.92	–	9×10^5	[90]
WSe_2/Si junction	390–1088	0.12	–	–	[91]
Wafer-scale- WS_2/Si	350–850	4×10^{-3}	1.5×10^{10}	1.2	[84]
Ag- WS_2/Si	390–670	2.09	6.6×10^{11}	2×10^5	[86]
WS_2/Si array	200–3043	8.3	4.6×10^{14}	16/29	[85]
$PdSe_2/Si$	200–3044	0.3	$\sim 10^{13}$	38/44	[95]
$PdSe_2$ /pyramid Si	980–1650	0.45	9.97×10^{13}	–	[96]
ME- $PdSe_2/Si$	400–1200	47.2	8.13×10^9	9.2/17.3	[97]
$ReS_2/SiO_2/Si$	450–800	–	–	160	[100]
p- ReS_2/Si	–	33.47	–	90	[101]
$MoTe_2/ReS_2$	532–1310	0.54	10^{10}	2.4	[102]

that along the transverse structure. This property makes it possible to use BP for the design of new types of electronic and photonic devices.

BP/Si waveguide photodetectors

However, the studies of BP/Si heterojunctions are scarcely reported. A few related reports are the studies of BP and silicon waveguides. These hybrid Si/BP waveguide photodetectors have similar constructions that are mostly focused on the NIR-MIR region (see Fig. 9a, b). For the hybrid BP/Si waveguide photodetector, the anisotropy of BP thin film also plays an important role in the electronic and optical properties of the photodetectors. Different thicknesses and different orientations will influence the photodetectors' performances. For example, Yin *et al.* [110] demonstrated a silicon photonic integrated BP detector operating at 2 μm wavelength. Then, they calculated the light absorption coefficient and electric-field profiles of BP with different thicknesses at different orientation angles (the θ is 0 when the orientation is along with the armchair direction) (see Fig. 9c, d). In this BP/Si waveguide heterostructure, BP film can cover the top and side walls of SOI ridge waveguide well, thus enhancing the light absorption of the transverse electro fundamental mode (TE_0) in the ridge waveguide. The device exhibited a responsivity of 306.7 $mA W^{-1}$ and a 3 dB bandwidth of 1.33 GHz at a low bias voltage of 0.4 V. In another BP/Si waveguide heterostructure [111], the BP film was integrated on the grating coupler, and the silicon waveguide and grating coupler ensured the longitudinal light propagation and out-of-plane coupling, thus enhancing the interaction between light and BP. In this report, they also found the responsivity would be influenced by the thickness and orientation (as shown in Fig. 9f,

g). In addition, the photoresponse would be tuned by gate and drain bias (as shown in Fig. 9e), indicating the changes in band structures.

Protection to BP

Although BP has shown excellent optoelectronic properties, its instability has become one of the greatest concerns and hinders its further development. Some studies have been shown that BP flakes will quickly degrade and lose their electrical properties after a few hours/days at atmosphere conditions. Researchers have tried some strategies to enhance the stability of BP, including covering a passive layer (h-BN, Al_2O_3 , etc.) to protect the BP layer [112]. Recently, the group of Zhang [113] tried a new material, hydrophobic polyionic liquid poly(1-hexyl-3-vinylimidazolium) hexafluorophosphate salt (PIL-TFSI), as the protecting layer to form a self-healable photodetector. In this PIL/BP photodetector, the PIL-TFSI layer provided a significantly enhanced stability as well as a fluorination to BP. In addition, this detector showed a self-healing capability due to the self-healing nature of PIL-TFSI. The typical on/off signals can still be detected in this photodetector even after 50 cycles. Herein, the organic reagent PIL-TFSI may be a promising material to enhance the stability of BP-based devices in future applications.

Brief summary

BP-based devices have shown excellent optoelectronic properties; taking advantage of BP's adjustable bandgap property, they exhibit excellent performances in the short-to-mid IR range. Especially when combined with Si, the devices not only have the high-speed characteristic of Si-based detector, but also exhibit

Figure 9 (a) Microscopy image of a BP photodetector on Si waveguide. (b) Schematics of the waveguide-integrated on-chip system with BP photodetector. (c) Light absorption coefficient α as the BP orientation angle θ varies from 0° to 360° . The thickness of BP is 20, 30, 40, 50, 60, 70 nm. (d) Calculated electric field profiles of the TE_0 mode supported in the hybrid silicon/BP waveguides at $2 \mu\text{m}$. (e) Contour plot of gate and drain-dependent photocurrent under $237 \mu\text{W}$ illumination at $3.78 \mu\text{m}$. The optimized photocurrent is obtained with negative drain bias. (f) Spectral responsivities and (g) NEPs of the BP/Si waveguides with different thicknesses and orientations. (a, b, e–g) are reprinted with permission from Ref. [111]. Copyright 2018, American Chemical Society. (c, d) are reprinted with permission from Ref. [110]. Copyright 2019, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

the property of anisotropy which Si-based detectors do not possess. However, for better performances of the BP/Si detector, the stability will be the focus to be studied.

OTHER LOW-DIMENSIONAL HETEROSTRUCTURE-BASED DEVICES

Apart from Si-based heterostructure-based photodetectors, ger-

manium (Ge) heterostructure is also employed to fabricate IR photodetectors. Ge has distinct properties, such as high light absorption, low cost and easy fabrication process, and narrower bandgap (0.66 eV) [114]. Graphene/Ge heterostructure detectors have unique characteristics. Zeng *et al.* [62] fabricated an SLG/Ge Schottky heterojunction for the IR light sensing. The graphene/Ge photodetectors were sensitive to IR light with the

responsivity of 51.8 mA W^{-1} , detectivity of 1.38×10^{10} Jones, and on/off ratio of 2×10^4 . It is worth noting that the photovoltaic IR detector exhibited a peak sensitivity at 1400 nm, corresponding to the intrinsic absorption of Ge, which means a good spectral selectivity. In addition, the SLG/Ge detectors exhibited good reproducibility and long-term stability. In fact, in Huang's report [11], the graphene/HfO₂/Ge and graphene/HfO₂/a-MoS₂ photodetectors were also studied, and the graphene/HfO₂/Ge photodetectors exhibited high responsivity of 9.03 A W^{-1} (at 0.1 V bias), fast response speed of $\tau_{\text{rise}} = 11 \mu\text{s}$, $\tau_{\text{decay}} = 38 \mu\text{s}$, which are superior to previous graphene/Ge devices. And the graphene/HfO₂/a-MoS₂ photodetectors showed sensitivities to 473–2712 nm light with responsivity of 5.36 A W^{-1} and detectivity of 1.57×10^9 Jones. These excellent performances suggested the graphene-based devices may have great potential for future IR detection application. Mahyavanshi *et al.* [115] demonstrated the MoS₂/Ge heterostructure photodetectors. At different bias voltage, the detector showed distinct photoresponsivity for UV, visible, and NIR light. And the response to UV light is a significant breakthrough for MoS₂-based photodetectors. On the other hand, some scientists have tried to directly fabricate the heterostructure of MoS₂ thin film and substrate. MoTe₂/Ge heterostructure-based devices are also exploited for NIR detection. Chen *et al.* [116] transferred MoTe₂ nanosheet on an undoped Ge substrate with 30-nm-thick Al₂O₃ as an isolating layer. The as-made photodetector exhibited excellent performances with very high responsivity of $12,460 \text{ A W}^{-1}$ and specific detectivity of 3.3×10^{12} Jones. However, the response speed is relatively low with a specific time of 5 ms, and the reverse current at dark is large due to the low barrier between MoTe₂ and Ge. So a GeO₂ was introduced between the MoTe₂ and Ge [117], and the reverse current was reduced by more than four orders of magnitude at a cost of reduced responsivity and detectivity (specific value of 15.6 A W^{-1} and 4.86×10^{11} Jones, respectively).

Gallium nitride (GaN) has also become a hot material to be used with MoS₂ to develop hybrid photodetectors due to their

low lattice mismatch. Zhuo *et al.* [118] made a PtSe₂/GaN photodetector which was *in-situ* fabricated by synthesis of vertically standing low-dimensional PtSe₂ film on n-GaN substrate. The PtSe₂/GaN heterojunction device has excellent photo-response properties under illumination by DUV light. This photodetector had a high responsivity of 193 mA W^{-1} , an ultrahigh specific detectivity of 3.8×10^{14} Jones, $I_{\text{on}}/I_{\text{off}}$ ratio of $\sim 10^8$, as well as short rise and decay times of 45/102 μs . They also fabricated the self-powered few-layered MoS₂/GaN p-n heterojunction photodetector which could work in DUV [119]. In order to develop the MoS₂/GaN p-n heterojunction photodetector, the MoS₂ thin films were transferred onto a GaN substrate where the Au electrode was deposited on the MoS₂ film and the Ni/Au electrode was deposited on the GaN *via* the EBE method. The MoS₂/GaN photodetector exhibited a current $I_{\text{on}}/I_{\text{off}}$ ratio of 10^5 at an illumination intensity of $2.4 \mu\text{W cm}^{-2}$ at zero bias voltage, indicating the photodetectors could work in the UV region without ultra power. The MoS₂/GaN photodetectors exhibited photoresponsivity of 187 mA W^{-1} , detectivity of 2.34×10^{13} Jones, fast response speeds of 46.4 μs at 0 V bias. Recently, Li *et al.* [13] tried to fabricate monolayer MoS₂/GaN-based devices and introduced tensile strain by depositing ultrathin Al₂O₃ layer (3 nm) to improve photovoltaic performances. As a result, the monolayer MoS₂/GaN/Al₂O₃ heterostructure-based photodetector achieved higher responsivity of 24.6 A W^{-1} , and their G and EQE are as high as 520% and 8381%, respectively. On the other hand, strain-adjusting of the bandgap of MoS₂ using designed pattern substrate is also an effective way to improve the performances of MoS₂/GaN-based photodetector. Liu *et al.* [120] designed orthohexagonal GaN arrays on a sapphire substrate, and then transferred MoS₂ on the pattern GaN substrate (PGS) with the help of polymethyl methacrylate; at the same time, they transferred MoS₂ on the flat GaN substrate (FGS) for comparison (Fig. 10). Under the same conditions, the MoS₂/PGS-based photodetector showed higher photocurrent and higher responsivity; the NEP decreased to $3.88 \times 10^{-13} \text{ W Hz}^{-1/2}$, and D^* increased to 5.6×10^8 Jones when

Figure 10 (a) Scanning electron microscopy (SEM) image of the PGS, where the inset shows the size of the patterned array; (b) schematic cross-sectional view of the PGS; switching characteristics and rise/fall times of (c, d) MoS₂/FGS and (h, i) MoS₂/PGS photodetectors under 460 nm laser illumination at 3 V; (e, f) Raman spectra of the MoS₂ film (e) before and after transfer and (f) at the top and bottom of the patterned array; (g) core-level X-ray photoelectron spectroscopy (XPS) spectrum of MoS₂/PGS. Reprinted with permission from Ref. [120]. Copyright2021, American Chemical Society.

compared with the MoS₂/FGS-based devices.

Moreover, the heterostructures of low-dimensional materials and other conventional substrates also have different performances. Nasr *et al.* [121] successfully combined MoS₂ and glass to fabricate a low-power photodetector. They first synthesized atomically thin MoS₂ on transparent glass substrate at a relatively low temperature *via* metal organic CVD (MOCVD), then grew high-*k* (dielectric constant) and ultra-thin dielectric Al₂O₃ layer as the top-gate dielectric to accomplish the low-power phototransistors. The as-made MoS₂-based photodetectors can be excited by low-intensity light source instead of usual laser excitation. It is worth noting that the energy consumption of the MoS₂-based photodetector is as low as about 100 pico Joules, which shows promise to realize the energy-efficient low-dimensional glasstronic technology. Xu *et al.* [122] successfully fabricated a MoS₂/GaAs heterostructure as a self-driven photodetector. And the MoS₂/GaAs heterostructure-based photodetector showed excellent optoelectronic performances with responsivity of 321 mA W⁻¹ and detectivity of 3.5 × 10¹³ Jones, respectively. To further improve the performances of the detector, they introduced an h-BN thin film to constitute a MoS₂/h-BN/GaAs sandwich structure which could effectively suppress the dark current. And the responsivity was increased to 419 mA W⁻¹ as well as the detectivity was increased to 1.9 × 10¹⁴ Jones.

SUMMARY AND OUTLOOK

Optoelectronic devices based on heterostructures of 2D materials have exhibited novel phenomena and achieved many high performances. It can be said that we have achieved great success in the research of 2D material/conventional substrate heterostructures. We herein reviewed the recent progress in the performances of cross-dimensional material-based photonic and optoelectronic devices, mainly focused on several typical 2D materials, including graphene, TMDCs, and BP. In recent years, tremendous advances have been made in the design of cross-dimensional heterostructures, such as larger spectrum range, faster speed, and better optoelectronic performances. For example, the spectral range of MoS₂/Ge is broadened to IR region. The patterned GaN helps a lot to improve the performances of MoS₂-based photodetectors. However, the promising and the one which has attracted the most attention, is the combination of 2D material and silicon. Some kinds of Si-based cross-dimensional heterojunction devices can break the limitations of the original low-dimensional materials and obtain higher responsivities and detectivities. In addition, the Si-based low-dimensional heterostructure devices are more likely to show high speed characteristics, providing a certain possibility for future communication circuits. Most important of all, silicon is the conventional material widely used in integrated circuits for that the cost of silicon is relatively cheap and the related technology is the most mature. Therefore, the Si substrate-based cross-dimensional heterostructure devices have the most potential to be embedded in modern large-scale integrated circuits as well as with a commercial potential. On the other hand, the low-dimensional materials can make up for some shortcomings of silicon materials, enhancing the light absorption for silicon even providing an opportunity for the smaller and smaller channel length in future. Herein, in our opinion, the future focus of cross-dimensional heterostructure is Si-based heterostructure. And there are some possible directions just for

reference: (1) find more kinds of low-dimensional materials to probe different possibilities of the Si-based heterostructure. Out of a sea of low-dimensional materials, there are always better low-dimensional materials that are more suitable for silicon-based heterostructure systems to be found and synthesized. (2) Dig deep in the common materials. The band structures of these heterostructures can be adjusted by the methods of introducing defects of low-dimensional materials, such as adjusting doping concentrations of Si substrate, and changing the interface states between the heterostructure. (3) Utilize the modern technologies, especially for these large-area 2D materials. The mature technologies would help the 2D/3D heterostructures form photodetection arrays, and even provide a potential compatible to large-scale integrated circuits.

Despite the novel phenomena shown and many high performances gained in the cross-dimensional heterostructures, there are still some problems and challenges. There are some fields requiring additional improvement: (1) processability and practical application. Most of reported studies are manipulated by manual system, which causes a hindrance for the industrialization and mass production. (2) Key parameters. Most of present devices can only enhance one or two aspects of the key parameters. Some cross-dimensional structure-based devices even have exhibited worse performances than pristine independent low-dimensional material-based devices. (3) Small synthesis area, except for several kinds of low-dimensional materials. Most low-dimensional materials are obtained with small scale, so it is difficult to obtain their arrays, hindering fast imaging that can be used in practical applications. Herein, more efforts need to be cast to improve full aspects of the devices. It is a long-term process for all stuff involved, not only for the researchers of low-dimensional materials to get high-quality crystalline and probe the possibilities of new cross-dimensional heterostructures, but also for the practitioners of conventional technologies to perfect various processes of the optoelectronic devices, as well as for the theoretical physicists to provide a broader platform to design more band structures with demand, even for the governor of related department, the ordinary people to pay more attention to low-dimensional materials to support the development of related industries. There are many significant challenges in producing low-cost, large-scale, high-performance photodetector devices for commercial applications, and therefore factors such as optimization of the photoresponse, reproducibility, and long-term environmental stability of photodetector devices used for photodetectors need much more attention.

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Jingshu Zhou received his BSc degree in microelectronic engineering from Nankai University in 2013. Currently, he is a PhD student under the supervision of Prof. Zhongming Wei at the Institute of Semiconductors, Chinese Academy of Sciences (IOS-CAS). His research interests focus on the preparation and photoelectric properties of 2D materials.

Xiangkai Zhao received his BSc degree in optoelectronic information science and engineering from Jilin University in 2003. Currently, he is working as an engineer at IOS-CAS. His research interests focus on optoelectronic detection and optoelectronic system.

Zhongming Wei received his BS degree from Wuhan University (China) in 2005, and PhD degree from the Institute of Chemistry, CAS (ICCAS) in 2010 under the supervision of Prof. Daoben Zhu and Prof. Wei Xu. From August 2010 to January 2015, he worked as a postdoctoral fellow and then assistant professor in Prof. Thomas Bjørnholm's group at the University of Copenhagen, Denmark. Currently, he is working as a Professor at IOS-CAS. His research interests include low-dimensional semiconductors and their optoelectronic devices.

二维/三维硅基异质结光电应用研究进展

周劲澍^{1,2}, 辛凯耀¹, 赵向凯^{3*}, 李冬梅³, 魏钟鸣^{1*}, 夏建白¹

摘要 以硅为基础的半导体技术在过去几十年随着摩尔定律取得了很大进展,但是它也正接近自己的物理极限。近年来,二维材料因其独特的结构和优越的性能,被认为是突破传统硅基光电器件限制的契机。本文针对二维材料探测器与三维硅基系统混合集成的研究现状,简述了几种用于光电探测器的二维材料的基本特性。随后,总结了基于二维材料的硅光子集成光电探测器的研究进展,并对其器件结构和主要性能进行了展述。其后,又补充介绍了结合其他传统材料为衬底的二维器件。最后,我们对二维/三维混合硅基异质结构的发展前景进行了展望。