Thin-film electronics based on all-2D van der Waals heterostructures

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ABSTRACT

Two-dimensional (2D) layered materials including metal, semiconductor, and insulator have received extensive attention in recent years. The weak van der Waals (vdW) interactions between 2D materials layers enable them to isolate monolayers and restack into artificial 2D vdW heterostructures in the desired sequence. These assembled all-2D vdW heterostructures are promising platforms for fabricating next-generation electronics as well as optoelectronics. In particular, the all-2D vdW heterostructure devices composed entirely of 2D layered material have received extensive attention due to their natural thickness, atomically sharp heterointerfaces, and excellent mechanical flexibility. Herein, we firstly introduce 2D vdW heterostructures and their preparation methods. Secondly, the recent progress of field-effect transistors (FETs) and photodetectors based on all-2D vdW heterostructures are summarized. Finally, we discuss some challenges of all-2D vdW heterostructure-based devices for practical applications and offer personal perspectives toward the future development of thin-film electronics.

1. Introduction

Since the successful exfoliation of single-layer graphene from graphite crystals in 2004 by Geim and Noveselov [1], graphene (Gr) has received extensive attention in thin-film transistors, optoelectronic devices, sensors, and energy storage and conversion [2], because of its ultra-high carrier mobility \( \sim 2 \times 10^5 \text{cm}^2\text{V}^{-1}\text{sec}^{-1} \) at 5 K) [3], thermal conductivity \( \sim 5300 \text{Wm}^{-1}\text{K}^{-1} \) [4], excellent mechanical strength (1.0 TPa) [5] and large specific surface area. The discovery of graphene further triggered the exploration and investigation of other 2D layered materials with diverse bandgaps and optoelectrical properties [2]. So far, 2D layered materials have grown up to a big family, including metals/semimetals (e.g. Gr, NbS\(_2\)), semiconductors (e.g. transition metal dichalcogenides (TMCDs), black phosphorus (BP) or III-VI layered materials) [6], and insulators (e.g. hexagonal boron nitride (h-BN)) [2,7]. These atomically 2D layered materials are promising as building blocks to construct novel electronics, given that traditional Si-based devices are approaching their physical dimension limitation [8].

2D layered materials are laterally composed of strong covalent bonds within the layers enabling in-plane stability. The weak van der Waals (vdW) interactions between layers allow us to isolate monolayer and restack them into artificial vdW heterostructures [5,7]. These 2D vdW heterostructures with various stack structures and tunable optoelectrical properties are ideal platforms for fabricating advanced thin-film electronics in the post-Moore era, open up a way for the design and realization of device functionalities [5,9]. For example, The BP/Gr heterostructure photodetector exhibited higher photoresponsivity and better long-term stability, compared with BP photodetector [10]. The carrier mobility of ReS\(_2\)/h-BN field-effect transistors (FETs) showed seven times larger than that of ReSe\(_2\) FETs on a SiO\(_2\) substrate [11].

It is worth mentioning that the performance of TMCDs based devices is greatly restricted by the large interface contact resistance between TMCDs and traditional metal electrodes, such as Pd, Al, Ti/Au, and Cr/Au [12]. Many strategies have been adopted to decrease contact resistance, such as using metal electrodes with suitable work functions, post-annealing treatment, or phase engineering [13–16]. Besides, using graphene electrode instead of traditional metal electrodes is an effective way to reduce interface contact resistance, and even form ohmic contacts at 2D channels/graphene interfaces by tuning graphene’s Fermi level through external gate voltages or elements doping [17]. It also further inspired the investigation of high-performance devices based on
all-2D heterostructures, which are completely made of 2D material for all components, including 2D semiconducting channel, 2D graphene electrode, and nor 2D h-BN dielectric layer.

Compared with traditional Si-based and none all-2D devices, all-2D heterostructure devices have some advantages. Firstly, all-2D devices have atomically sharp 2D-2D heterointerfaces, including channel/graphene electrode, channel/dielectric layer, or channel/encapsulation layer interfaces [18]. Secondly, lower contact resistance at channel/graphene electrode interfaces promotes efficient carrier transfer [15]. Thirdly, excellent mechanical flexibility as well as high optical transmittance, make them attractive in the field of flexible and wearable nanodevices [19]. Furthermore, the growing interest in all-2D devices is not limited to the discovery of new physical phenomenon in the laboratory, the final goal is to achieve industrial applications. Some researchers have summarized relevant progress of nanodevices based on 2D materials and 2D vdW heterostructures, but a mini-review focus on all-2D heterostructure devices is still lacking. Herein, starting from the introduction of 2D vdW heterostructures and their preparation methods, including mechanical transfer and chemical vapor deposition (CVD). And then, recent progress of FETs and photodetectors based on all-2D heterostructures was summarized. Finally, we put forward some challenges and personal opinions, for the preparation and practical application of all-2D heterostructure nanodevices.

2. The overview of 2D vdW heterostructures

Layered materials can be divided into metals, semiconductors, and insulators. Figure 1(a) displays several typical 2D structures of Gr, BP, TMCDs, and h-BN. Graphene has a typical hexagonal crystal structure and is zero bandgap material in theory. Graphene-based 2D vdW heterostructures have been investigated in thin-film electronics, such as Gr/TMCDs, Gr/TMCDs/h-BN, and BP/Gr/h-BN heterostructures [2,5,20]. BP is made up of phosphorus elements and has a direct bandgap ranging from 0.33 eV for the bulk crystal to 1.5 eV for the monolayer [10,21]. BP is unstable and sensitive to water and oxygen atmosphere, to prevent the oxidative degradation of BP-based devices, some encapsulation protective layers are usually employed, such as h-BN, Al2O3, and other hydrophobic polymers [22,23]. TMCDs with a chemical formula of MX2, where M represents the transition metal of group IV-VI, such as Mo, W, and Sn, and X stands for chalcogenide elements of S, Se, and Te [24]. TMCDs have a typical hexagonal structure, and each layer is composed of three layers stacked in the form of X-M-X [25]. h-BN has a similar structure to graphene, a hexagonal lattice structure of alternating B and N atoms, sp2 covalent bonds. h-BN has a large bandgap of ~ 6 eV and usually served as substrate, encapsulation layer, and top-gate dielectric layer in nanodevices [26]. For example, the field-effect mobility of h-BN/Gr/h-BN vertical device up to 140,000 cm²V⁻¹sec⁻¹ (4 K) and 100,000 cm²V⁻¹sec⁻¹ (room temperature), because h-BN efficiently suppresses carriers scattering [27]. Additionally, in an h-BN passivated Gr-MoS₂-Gr all-2D devices, a low subthreshold swing (SS) can be achieved [28].

2D layered materials have unique advantages for designing photodetectors. First, they interact strongly with light despite the atomic-level thickness, mainly grounded on the mechanism of photovoltaic effect, photoconductive effect, and photogating effect. Second, their band structure and absorption wavelength can be tuned by varying the number of layers. Third, surface-free dangling bonds enable the fabrication of 2D vdW heterostructures by combining various 2D materials or integrating them on any substrates [5], these 2D vdW heterostructures with reduced interfacial distortions benefit the transport of photogenerated carriers. Fourth, the intrinsic mechanical flexibility of 2D materials offering their potential application in flexible and wearable fields [18]. Photodetectors based on 2D vdW heterostructures also exhibited remarkable performances, such as ultrabroad-band photoresponse, ultrahigh photonresponsivity, ultrahigh external quantum efficiency, fast response speed of dozens of microseconds, and room-temperature operation.

The weak van-der-Waals interactions in 2D materials allow us to isolate monolayer and multilayer and restack them into vertical 2D vdW heterostructures, layer-by-layer stacking, just like Lego bricks [7], as shown in Figure 1(b). Besides, surface-free dangling bonds enable the fabrication of 2D vdW heterostructures with various structures and band alignments, without the limitation of lattice mismatch, simultaneously [29]. So far, most of the reported 2D vdW heterostructures were prepared by mechanical transfer and CVD, and their advantages and disadvantages will be described in the following content. Herein, we mainly emphasize the recent development and application in FETs and photodetectors based on all-2D heterostructure, where devices are built of the 2D semiconducting channel, graphene electrode, and h-BN dielectric/encapsulation layer, as presented in Figure 1(c).

3. Preparation of 2D vdW heterostructures

Up to now, most 2D vdW heterostructures are assembled in the laboratory using either mechanical transfer or CVD methods. The whole mechanical transfer process mainly consists of two steps, the first step is to
exfoliate 2D thin flakes from their bulk counterparts. The second step is to transfer and restack pre-fabricated nanoflakes into 2D vdW heterostructures with the assistance of a micromanipulation platform [29,31]. However, pre-fabricated 2D thin flakes suffer from random size, low yield, and time-consuming. And the crystal wrinkles, contaminant residues, bubbles, and uncontrollable stack orientation are unavoidable in manual transfer processes [31]. The CVD method, which is one kind of bottom-up strategy, can realize the direct growth of 2D vdW heterostructures with a large area, however, its growth dynamics and stacking angle remains challengeable [32]. In addition, the CVD technique can realize the preparation of vertical and lateral 2D vdW heterostructures, while the machinal transfer method only assembles vertical 2D vdW heterostructures [25]. The preparation of 2D vdW heterostructures with multilayer structures by the CVD technique is challenging at present, but the mechanical method is free from restrictions. In addition, solution-based printing methods were also briefly described for preparing all-2D vdW heterostructures devices [76–79].

### 3.1. Mechanical transfer

#### 3.1.1. Mechanical exfoliation

A mechanical force is applied to delaminate 3D bulk crystals into 2D thin flakes by eliminating the vdW interaction between layers, without destroying their in-plane structures, simultaneously [33]. Since the exfoliation of single-layer graphene (SLG) from highly oriented pyrolytic graphite (HOPG) with the assistance of Scotch tape [34], as illustrated in Figure 2(a). This Scotch tape-assisted method gradually becomes the most common strategy for exfoliating 2D flakes and remains still popular in the laboratory [35]. However, this method suffers from some disadvantages, such as time-consuming, unrepeatable process, exfoliated flakes with irregular shape and size (e.g. lateral size usually less than 100 μm) [36]. To produce high-quality 2D thin flakes, the substrate, tape adhesiveness, and/or operational temperature should be optimized [31]. For example, in 2015, Huang et al. developed a substrate enhanced method and produced graphene and other 2D thin flakes with a size of hundreds of micrometers [37]. Magda et al. proposed an
Au-assisted method and obtained MoS$_2$, WS$_2$, and Bi$_2$Te$_3$ monolayer with a size of up to centimeter-level [38]. The single-layer TMCDs flakes with near-unity yield were also obtained and their macroscopic dimensions were limited by the original size of bulk crystals [39].

### 3.1.2. Transfer and stacking

To form 2D vdW heterostructures for device applications, the pre-fabricated 2D flakes were transferred on the target substrate using polymer carriers [36]. In the following paragraphs, we mainly described the wet-transfer and dry-transfer processes. In 2010, Dean et al. utilized the polymethyl-methacrylate (PMMA) assisted wet-transfer technique to fabricate a Gr/h-BN heterostructure [40], as shown in Figure 2(b). Graphene was firstly placed on the surface of PMMA film with a water-soluble layer (i). The water-soluble layer was dissolved in deionized water and Gr/PMMA film floated on the water (ii). The Gr/PMMA film was transferred on a glass slide (iii), and then graphene was aligned on the h-BN substrate (iv). In the end, the PMMA carrier was dissolved in acetone solution and the Gr/h-BN heterostructure was further annealed in H$_2$/Ar mixed gas at 340°C for 3.5 h to reduce residual contaminants. The above method was called the wet-transfer process.

Compared to the wet-transfer method, Figure 2(c) displayed the fabrication process of all-2D h-BN/Gr-MoS$_2$-Gr FETs using a dry-transfer method [28]. Firstly, graphene was dry-transferred on Si/SiO$_2$ substrate (i) and was patterned to define the source/drain electrode (ii). The MoS$_2$ flake was transferred between graphene source/drain electrodes, as a semiconducting channel (iii). The h-BN was transferred on the surface of the MoS$_2$ channel (iv), the top graphene electrode was finally covered on the h-BN surface (v). In the dry-transfer process, the solution-related steps were simplified to make a clearer 2D interface. Such the dry-transfer method has been widely adopted for device fabrication. For example, Gr-WS$_2$-Gr photodetector arrays were done on a SiO$_2$/Si substrate using the dry-transfer method [41]. Moreover, to further avoid possible contaminant residues [42], Castellanos-Gomez et al. achieved the completely dry-transfer using viscoelastic stamps during the preparation process.
3.2. CVD growth

Compared to mechanical exfoliation, CVD is an efficient technique for scalable production of 2D materials and 2D vdW heterostructures with large areas [32], which could also avoid labor-intensive operations [44]. Generally, there are two approaches for synthesizing 2D vdW heterostructures: one way is to directly grow TMCDs heterostructures by one-step CVD method, another way is to sequentially grow 2D materials on top of other 2D materials.

Gong et al. used a one-step CVD method to realize the preparation of vertical and lateral heterostructures of WS₂/MoS₂ through controlling heating temperature [45]. A schematic of the synthesis process was displayed in Figure 3(a). The WS₂/MoS₂ vertical heterostructure was synthesized at 850°C (see Figure 3(b) and (c)), while the WS₂/MoS₂ lateral heterostructure was produced at 650°C (see Figure 3(d) and (e)). The nucleation and growth of WS₂ on the MoS₂ surface need adequate energy to overcome the nucleation barrier, compared with the growth of WS₂ on the edge of MoS₂. At the lower temperature of 650°C, the nucleation and growth of WS₂ on the MoS₂ surface were relatively tough and slow. On the other hand, sequential CVD growth is a more common method for preparing 2D vdW heterostructures. Liu et al. realized the fabrication of h-BN/Gr heterostructures via a two-step CVD process [46]. Firstly, graphene was grown on a Cu foil at 950°C, and then h-BN was in-situ deposited on the graphene/Cu substrate by a secondary CVD using ammonia borane (NH₃–BH₃) as a precursor. A large-area MoS₂/h-BN heterostructure was synthesized using the two-step CVD method [47]. Besides, Lin et al. successfully prepared MoS₂/WSe₂/Gr and WSe₂/MoS₂/Gr vertical heterostructures through adjusting CVD growth temperature [48]. Similarly, Li and co-workers realized the preparation of Gr-WSe₂-Gr lateral heterostructure via a two-step CVD process [49], as shown in Figure 3(f). Graphene was firstly deposited on a Cu foil by the first CVD process (i) and then it was transferred on a sapphire substrate (ii). Secondly, the transferred graphene was patterned after optical lithography and etching treatment (iii). After the secondary CVD growth of WSe₂, the Gr-WSe₂-Gr lateral heterostructure was finally produced (iv). The Gr-GaS-Gr and other lateral heterostructure devices were also synthesized by such the two-step CVD process [50].
Besides, solution-based printing methods have been employed for fabricating all-2D heterostructure single devices or arrays [76–79]. The preparation of stable and uniform nanosheet suspensions in organic solvents by liquid-phase exfoliation is a prerequisite, and then printing graphene electrodes, 2D nanosheet channels, and the dielectric layer to fabricate transistors or arrays. G. Kelly et al. realized the preparation of all-printed Gr/h-BN/Gr-WSe2-Gr thin-film transistors from nanosheet networks [76]. A Gr/h-BN/Gr-MoS2-Gr transistor on the paper was prepared by combining CVD and inkjet-printing techniques [77]. Similarly, Gr-MoS2-Gr photodetector was fabricated using the inkjet-printing technique [78]. Alzakia et al. realized the one-step preparation of Gr-MoS2-Gr and Gr-WS2-Gr photodetectors by electrohydrodynamic (EHD) printing [79].

4. All-2D vdW heterostructures-based field-effect transistors

The silicon-based complementary metal–oxide-semiconductor technique has been widely used in the modern integrated circuit industry, but its further application is limited by the constantly downsizing physical dimension, according to Moore’s law [24]. In Si-based transistors, low contact resistance can be achieved by introducing highly doped N++ or P++ regions. However, the high-concentration element doping method is not appropriate for TMCDs-based electronics, which might damage their lattice structures and introduce lots of defects, leading to unexpected Fermi-level pining at TMCDs/metal interfaces [14,18]. Actually, at 2D channel/metal interfaces, the contact resistance is still about 10–100 times higher than Si-based transistors [23,51]. Using graphene to replace traditional metal electrodes and then developing all-2D vdW heterostructures have already received attention due to its ultrahigh conductivity and tunable Fermi level [52]. Besides, graphene has excellent mechanical flexibility (failure strain of about 12% for SLG) and weak optical absorbance (transparency of 97.4% for SLG), which make it an ideal transparent electrode for flexible and wearable electronic devices [53]. In the following paragraphs, we will review the literature about all-2D vdW electronics with graphene contacts, which are classified into two groups, i.e. vertically and laterally graphene contacted all-2D FETs.

4.1. Vertically graphene contacted all-2D FETs

A typical example of a vertical all-2D FET (i.e. Gr/h-BN/Gr-MoS2-Gr heterostructure) is shown in Figure 4(a), in which that graphene flakes are taken as the top-gate and source/drain electrodes. h-BN and MoS2 are employed as the top-gate dielectric and semiconducting channel, respectively [28]. This FET showed a typical n-type behavior in the transfer curves (see Figure 4(b)). In 2014, Du et al. demonstrated that Gr-MoS2-Gr FETs can reach a high on/off current ratio and low contact resistance, compared with MoS2 FETs without graphene electrodes [54]. Lee et al. reported that a lower Schottky barrier at the graphene/MoS2 heterointerface in Gr-MoS2-Gr FET via tuning graphene’s Fermi level [55].

Given the linear energy dispersion relation near the Dirac point, graphene’s Fermi level can be adjusted by an external electric field, allowing the modulation of Schottky barrier height at 2D channel/graphene heterointerfaces. Other vertical all-2D FETs, such as h-BN/Gr-MoS2-Gr/h-BN [14], h-BN/Gr-WSe2-Gr/h-BN [56], and Gr-ReSe2-Gr [17], have also shown similar advantages of using graphene electrodes. In addition, Zhang’s group fabricated vertical all-2D vdW FETs with an ultra-short channel width of 4 nm [57]. Their experimental results displayed a nearly Ohmic contact and a negligible short-channel effect, providing a pioneering for preparing ultrasmall-scaled electronics based on all-2D heterostructures.

In all-2D vdW FETs, it has to be emphasized that h-BN was usually employed as the substrate, dielectric layer, and/or protective layer for electronics due to its surface-free dangling bonds, atomically flat, inert surface, and insulating behavior. As shown in Figure 4(c), h-BN served as a protective layer for device encapsulation to restrain the degradation of the semiconducting BP channel in h-BN/Gr-BP-Gr heterostructure and improve device stability. The h-BN encapsulated FET further showed similar transfer curves under air and vacuum conditions (see Figure 4(d)), indicating good passivation [21]. The result for the non-encapsulated FET was shown in its corresponding inset. Similarly, the h-BN/Gr-MoS2-Gr/h-BN FET achieved a record-high Hall mobility up to 34,000 cm²V⁻¹sec⁻¹ at low temperature, in which the MoS2 channel was fully encapsulated by h-BN and multi-terminal contacted by graphene electrode [58]. Duan et al. reported that the impurities scattering was significantly suppressed by the h-BN encapsulation layer in h-BN/Gr-MoS2-Gr/h-BN FETs, which showed field-effect mobility up to 1300 cm²V⁻¹sec⁻¹ at low temperature [14].

Apart from the implementation of unipolar electronics in all-2D vdW FETs, ambipolar FETs could be realized by using graphene contacts. Lee et al. prepared an h-BN/Gr-ReSe2-Gr/h-BN FET, whose corresponding schematic and optical image were revealed in Figure 4(e) and (f), respectively [52]. Ambipolar transfer characteristics can be successfully done owing to the tunable Fermi
level of the graphene and negligible and 30 meV Schottky barrier heights for the n- and p-channel regimes, respectively (see Figure 4(g) and (f)). When $V_{bg} < 0$ V (p-channel), the energy band bent upward, and the Fermi level near the valence band of ReSe$_2$ and graphene. Thus, the holes could easily inject from graphene into the ReSe$_2$ channel. When $V_{bg} > 0$ V (n-channel), the energy band bent downward, and the Fermi level near the conduction band of ReSe$_2$ and graphene, and electrons as the majority carriers were easily injected from graphene to ReSe$_2$. Such a demonstration shows the great potential of graphene contacts in all-2D vdW FETs. On the other hand, how to realize vertically integrated multilayer electronics as well as multiply on-current density via the layer-by-layer stacking process is also crucial for the practical applications of all-2D vdW heterostructures. Zhang’s research group firstly used an individual unit of the FLG/h-BN/Gr-MoS$_2$-Gr/h-BN/FLG FET (see Figure 4(i)) to assemble vertically integrated multilayer electronics (see Figure 4(j)) [9]. With increasing the number of the semiconducting MoS$_2$ channels, the multiplied on-current density, the integrated electronics showed a nearly ideal multiplication on on-current density (see Figure 4(k)), while their corresponding off-current density remained at the fA range (see Figure 4(l)).

Because all-2D vdW heterostructures possess both high mechanical flexibility and optical transmittance, in recent years, flexible and wearable electronics based on all-2D vdW heterostructures have also attracted great attention. For example, an ambipolar h-BN/Gr-WSe$_2$-Gr FET was made on a polyethylene terephthalate (PET) substrate [19], and its optical image was displayed in Figure 5(a). This flexible FET exhibited a high on/off current ratio up to $10^7$ and carrier mobility of 45 cm$^2$V$^{-1}$sec$^{-1}$, meanwhile, its electrical properties remained almost unchanged when applied in-plane mechanical strain of 2%, as shown in Figure 5(b). Besides, its corresponding transparency can keep about 88% in the whole spectral range, as shown in Figure 5(c). Yoon et al. reported a Gr-MoS$_2$-Gr FET on a PET substrate, which showed a typical $n$-type behavior, as illustrated in Figure 5(d) [53]. Slight variations in the electrical properties of mobility $\sim$ 6% and threshold voltage $\sim \pm 1.7$ V were observed, when bending up/down to 2.2 mm under
compressive and tensile tests (see Figure 5(e)). Furthermore, after $10^4$ bending cycles with a bending radius of 2.7 mm, the mobility decreased by around 30% and can be recovered after annealing treatment at 200°C for 2 h, as shown in Figure 5(f). In addition, large-area flexible all-2D FETs have been successfully prepared on PET [76] and paper substrate [77] by printing methods, but the transistor’s electrical performance was relatively lower.

4.2. Laterally graphene contacted all-2D FETs

Different from vertically graphene contacted all-2D FETs, laterally graphene contacted all-2D FETs are contacted with graphene electrodes through in-plane edge. The contact area is defined by the cross-sectional area of channels and graphene flakes. Such a structure of the edge-contacted graphene electrode allows more efficient carrier injection into the semiconducting channels, resulting in lower contact resistance. In 2016, Park et al. simultaneously synthesized a Gr-MoS$_2$-Gr FET with edge-graphene electrodes (1DG) and fabricated metal-MoS$_2$-metal FET with top-contact metal electrodes (2DM) [61]. The schematic illustration of both device structures and the corresponding transfer characteristics are shown in Figure 6(a) and (b), respectively. It can be found that the electrical performances of the lateral all-2D vdW FETs surpass that of the metal-MoS$_2$-metal FET with 2DM. Through the transmission-line method, the contact resistance ($R_C$) for the lateral Gr-MoS$_2$-Gr (black circles) and Gr-WS$_2$-Gr (blue triangles) FETs were further investigated, as shown in Figure 6(c). Owing to higher carrier injection in the edge-contact structure, lower contact resistances can be obtained, compared with different contact structures in Figure 6(d). It is noted that the 2DG denotes a vertical Gr-MoS$_2$-Gr FET with a variation of an overlap contact area. In 2017, Behranginia et al. employed seed-free consecutive CVD processes to synthesize high-quality lateral Gr-MoS$_2$-Gr heterostructures and comprehensively investigated their electrical properties [59]. The corresponding images of the lateral heterostructure and FET are shown in Figure 6(e) and (f), respectively. Through analyzing temperature-dependent transfer characteristics, the lateral Gr-MoS$_2$-Gr FETs revealed better device performances as well as lower Schottky barriers, which were attributed to the energy band rearrangement at the lateral graphene contact (see Figure 6(g) and (h)). In addition, Hong and co-workers realized a large-area growth of lateral Gr-MoS$_2$-Gr heterostructures through the two-step CVD process [60]. Their Gr-MoS$_2$-Gr FETs exhibited an on/off ratio up to $10^9$ and mobility of 8.5 cm$^2$V$^{-1}$sec$^{-1}$, both of which were higher than those of MoS$_2$ FETs.

Most researches about the lateral all-2D vdW FETs were n-type behaviors, while the investigation of p-type lateral FETs was relatively lacking. In 2017, Tang et al. demonstrated that p-type lateral Gr-WS$_2$-Gr heterostructures grown with the scalable CVD technique
5. All-2D vdW heterostructures-based photodetectors

Until now, traditional thin-film photodetectors still occupied the commercial market. Si and InGaAs-based photodetectors work at room temperature and respond to wavelengths ranging from visible to near-infrared regions. Besides, HgCdTe and GeSb/InAs-based photodetectors can work at 77 K and have a response ranging from mid-wavelength to long infrared wavelength [62,63]. Recently, novel photodetectors based on 2D materials and 2D vdW heterostructures have received extensive attention [64], which are divided into photconductor, phototransistor, and photodiode-based on the device structures. The key parameters include photoresponsivity ($R$), external quantum efficiency (EQE), the response time ($\tau$), and photodetectivity ($D^*$) [63,65]. In the early layered-materials-based photodetectors, the $R$ value was only a few mA/W [20]. In the follow-up researches, it is found that using graphene as device electrodes becomes an efficient way to improve optoelectronic performances by reducing Schottky barriers at 2D material/electrode interfaces as well as increasing device absorption area [66]. In the following paragraphs, we will provide a summary of the recent progress on all-2D vdW heterostructures-based photodetectors.
The basic structure of all-2D vdW heterostructures-based phototransistors is shown in Figure 7(a), which was made of a photosensitive 2D channel and two graphene electrodes. Such an ultimate photodetection performance strongly depends on the photosensitive 2D channel, graphene electrodes, external voltage, and interface contact between the photosensitive channel and graphene electrode. In 2013, Novoselov’s group realized the concept of an all-2D Gr-WS2-Gr photodetector on a SiO2/Si substrate, as shown in Figure 7(b). This device showed an R of 0.1 AW−1 and EQE of 30% [67]. Besides, the device current exhibited obvious changes under the condition of laser on and laser off and gate-voltage operations, as illustrated in Figure 7(c), which directly affect photocarrier transfer efficiency and then make an ultimate output photocurrent. Yu et al. also realized a similar device structure using the Gr-MoS2-Gr heterostructure, in which its R value up to 0.22 AW−1, corresponding to an EQE of 55% [68]. By shifting graphene’s Fermi level, Zhang et al. obtained tunable Schottky barriers at the graphene/MoTe2 interfaces, and then the Gr-MoTe2-Gr heterostructures can be taken as an ultrasensitive near-infrared photodetector [69]. The same operation of modulable graphene’s Fermi level was also utilized for Gr-InSe-Gr photodetector [70,71,81]. In 2015, Patanè et al. further compared the performance difference between Gr-InSe-Gr and Au-InSe-Au photodetectors, as shown in Figure 7(d), in which higher R values were obtained in the Gr-InSe-Gr heterostructures, attributed to the energy match between graphene’s electron affinity and the 2D channel’s work function [64]. Besides, Wei et al. found that the Gr-MoTe2-Gr contacted with asymmetric graphene based on p-type and n-type exhibited a higher performance than that of device contacted with symmetry graphene [71]. The integration of asymmetric graphene contacts produced a larger built-in electric field of about 100 kV/cm. Additionally, the contact interface between the photosensitive channel and graphene electrode also affects the device’s performance. Chen et al. found that CVD-grown Gr-WS2-Gr photodetector had a smaller van der Waals overlapping area between WS2 and graphene compared with mechanically transferred Gr-WS2-Gr device [72]. Directly CVD-grown WS2 on the edge of pre-patterned graphene to form in-plan epitaxial interfaces and obtained more uniform lateral heterostructures. The R of CVD-grown Gr-WS2-Gr photodetector was two orders of magnitude higher than the transferred device due to the lower Schottky barrier at CVD-grown WS2/graphene interface.

Because of the quantum confinement and the variation of interlayer traps when 2D material is thinned to a monolayer, the thickness of the photosensitive 2D channel is a factor to affects the performances of all-2D vdW heterostructure photodetectors. In 2018, Chang et al. investigated the Gr-SnSe2-Gr photodetectors with different thicknesses of the SnSe2 channel, and the schematic is shown in Figure 7(e) [73]. From their thickness-dependent results, a transition from the interface-dominated response for thin to bulklike response for thicker was found, showing the sensitivity of photodetectors using 2D materials on the different number of layers. The photocurrent and corresponding R values as a function of light power intensity are shown for different SnSe thicknesses in Figure 7(f) and (g), respectively.


dochangeswere incorporated into graphene electrodes sandwiched structures for photodetector [74]. The results indicate that photoresponse properties were strongly dependent

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**Table 1. Electrical properties of all-2D vdW heterostructure FETs.**

| Types          | FET structures (Preparation method) | Electrode; channel; dielectric layer | On/off ratio | Mobility (cm² V⁻¹ sec⁻¹) | Substrate | Year | Ref. |
|----------------|------------------------------------|-------------------------------------|--------------|---------------------------|-----------|------|------|
| Vertical       | Gr-MoS2-Gr(Transfer & CVD)         | FLG; MoS2; c-PVP                     | 10⁴          | 4.8                       | PET       | 2013 | [53] |
|                | h-BN/Gr-WSe2-Gr(Transfer & CVD)    | SLG; WSe2; h-BN                      | 10⁷          | 34/45(e⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻WebElement scrieshowed up to 0.22 AW⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻.chomp the energy match between graphene’s electron affinity and the 2D channel’s work function [64]. Besides, Wei et al. found that the Gr-MoTe2-Gr contacted with asymmetric graphene based on p-type and n-type exhibited a higher performance than that of device contacted with symmetry graphene [71]. The integration of asymmetric graphene contacts produced a larger built-in electric field of about 100 kV/cm. Additionally, the contact interface between the photosensitive channel and graphene electrode also affects the device’s performance. Chen et al. found that CVD-grown Gr-WS2-Gr photodetector had a smaller van der Waals overlapping area between WS2 and graphene compared with mechanically transferred Gr-WS2-Gr device [72]. Directly CVD-grown WS2 on the edge of pre-patterned graphene to form in-plan epitaxial interfaces and obtained more uniform lateral heterostructures. The R of CVD-grown Gr-WS2-Gr photodetector was two orders of magnitude higher than the transferred device due to the lower Schottky barrier at CVD-grown WS2/graphene interface.

Because of the quantum confinement and the variation of interlayer traps when 2D material is thinned to a monolayer, the thickness of the photosensitive 2D channel is a factor to affects the performances of all-2D vdW heterostructure photodetectors. In 2018, Chang et al. investigated the Gr-SnSe2-Gr photodetectors with different thicknesses of the SnSe2 channel, and the schematic is shown in Figure 7(e) [73]. From their thickness-dependent results, a transition from the interface-dominated response for thin to bulklike response for thicker was found, showing the sensitivity of photodetectors using 2D materials on the different number of layers. The photocurrent and corresponding R values as a function of light power intensity are shown for different SnSe thicknesses in Figure 7(f) and (g), respectively. Also, Gao et al. used the SnSe2 channel with different thicknesses was incorporated into graphene electrodes sandwiched structures for photodetector [74]. The results indicate that photoresponse properties were strongly dependent on the thickness of the photosensitive channel, graphene electrodes, external voltage, and interface contact between the photosensitive channel and graphene electrode.
on the channel thickness. The authors then claimed that multilayered 2D channels with graphene as contact electrodes have good optoelectrical properties for future electronics. Besides, to improve the stability of all-2D vdW photodetectors, the encapsulation of using h-BN was used. As shown in Figure 7(h), the photosensitive BP channel was encapsulated by h-BN in the h-BN/Gr-BP-Gr photodetector to avoid its direct exposure to air atmosphere [75]. This device exhibited a wavelength response range from visible to mid-infrared, corresponding to an EQE from 20% to 52% (see Figure 7(i)).

In the photodiode, p-n heterostructures can be used to prepare ultra-sensitive photodetectors, because the built-in electric field generated at heterointerface can separate photocarriers efficiently. Based on the photovoltaic effect, self-powered p-n heterojunction photodetectors without additional energy consumption were reported, such as MoS2/WSe2 [48], p-MoS2/n-InSe [83], WSe2/Bi2Te3 [84], etc. These above reported p-n heterojunctions were contacted with metal electrodes, a small amount of all-2D p-n heterojunction photodetectors contacted with graphene electrodes were also investigated. For example,
in 2014, Lee et al. synthesized a Gr-WSe2/MoS2-Gr p-n device, which exhibited a large photovoltaic EQE of 34% [85]. Tan et al. suggested that the R (up to $10^3$ A/W) of Gr-WSe2/MoS2-Gr heterobilayer increased by more than an order of magnitude compared with Gr-WS2-Gr and Gr-MoS2-Gr monolayer devices (see Figure 7(l)) [86]. It was attributed to strong Coulomb interaction at WS2/MoS2 layers. Their schematic diagram (see Figures 7(j)) and the optical image of Gr-WSe2/MoS2-Gr heterobilayer arrays (see Figures 7(k)) were also displayed. Furthermore, some flexible all-2D photodetectors fabricated on PET and polyimide (PI) substrate were also reported, even though their photodetection performance was inferior [78,79,82]. Table 2 summarized the device structures, preparation method, substrates, and performance of all-2D photodetectors that were fabricated by mechanical transfer, CVD, and solution-based printing methods.

### Table 2. Photodetection performance of all-2D heterostructure photodetectors.

| Structure (preparation method) | Substrate | R(A/W) (wavelength) | EQE (%) | Response time ($\tau_r$/ $\tau_d$) | $V_g/V_{ds}$ (v/v) | Year | Ref |
|-------------------------------|-----------|---------------------|---------|----------------------------------|-------------------|------|-----|
| Gr-MoS2-Gr/h-BN (Transfer & CVD) | SiO2/Si | 0.1 (633 nm) | 30 | NA | −40 / 0 | 2013 | [67] |
| Gr-MoS2-Gr | SiO2/Si | 0.22 (488 nm) | 55 | NA | NA | 2013 | [68] |
| Gr-WSe2/MoS2-Gr (Transfer) | SiO2/Si | NA (532 nm) | 35% | NA | 0 / 0 | 2014 | [85] |
| Gr-InSe-Gr (Transfer & CVD) | SiO2/Si | $\approx 10^3$ (633 nm) | NA | NA | 0 / 2 | 2015 | [64] |
| Gr-InSe-Gr | SiO2/Si | 60 (500 nm) | 14850 | 100 $\mu$s | 0 / 10 | 2015 | [70] |
| Gr-WSe2(1L)-Gr | SiO2/Si | 12.5 (532 nm) | 583933 | NA | 0 / 5 | 2016 | [41] |
| Gr-lnSe-Gr | SiO2/Si | 0.1 (1064 nm) | 12.9 | 24 $\mu$s / 46 $\mu$s | NA | 2017 | [69] |
| Gr-MoTe2-Gr | SiO2/Si | 60(500nm) | 4600 | NA | NA | 2018 | [73] |
| Gr-WS2(2L)-Gr | SiO2/Si | 1.3 × 10^3 (532 nm) | 3 × 10^4 | 38.2 ms / 32 ms | 80 / 0.5 | 2019 | [74] |
| Gr-WS2(1L)-Gr | SiO2/Si | 1.6 (750 nm) | 154 (1100 nm) | NA | 0 / 2 | 2019 | [80] |
| Gr-WS2-Gr/h-BN | SiO2/Si | 11.7 (270 nm) | NA | NA | $< 60$ ms | 2019 | [50] |
| Gr-MoS2-Gr/h-BN | SiO2/Si | 0.0276 (550 nm) | 0.0124 (1064 nm) | NA | 16.6 $\mu$s / 15 $\mu$s | 0 / 0 | 2019 | [81] |
| Gr-MoTe2-Gr | SiO2/Si | 1.6 (750 nm) | 154 (1100 nm) | NA | NA | 2020 | [81] |
| Gr-GaS-Gr (Transfer & CVD) | SiO2/Si | 19300 (470 nm) | NA | NA | NA | 2021 | [82] |
| Gr-MoS2-Gr (Transfer & CVD) | glass/PET | 0.128 (532 nm) | 2 × 10^6 | 100 $\mu$s | 0 / 4 | 2019 | [78] |
| Gr-MoS2-Gr (inkjet-printing) | glass/PI | 0.050 (515.6 nm) | NA | NA | NA | 2020 | [79] |
| Gr-MoS2-Gr (EHD printing) | glass | 1.5 × 10^-6 | 0.11 × 10^-4 | NA | NA | 2020 | [79] |

Notes: Transfer – Mechanical transfer, & – And, EHD – Electrohydrodynamic.

### 6. Conclusions and outlook

In this review, we first introduced the development of 2D vdW heterostructures and their preparation methods of mechanical transfer and CVD. The 2D vdW heterostructures with tunable optoelectrical properties, diverse band alignments, and atomically sharp heterointerfaces, which are ideal platforms for advanced nanodevices in the post-Moore era. Last, we summarized the recent progress of FETs and photodetectors based on all-2D vdW heterostructures. However, the practical application of all-2D vdW heterostructures-based thin-film electronics still has a long way to go. Until now, the preparation of all-2D vdW heterostructures is still in the laboratory stage. Once a reliable method was developed to achieve the mass production of 2D layered materials, relevant mechanical transfer platforms will soon be automated and industrialized, enabling the industrial application of 2D vdW heterostructures to become a reality.
On the other hand, most studies mainly focus on a single all-2D device level (i.e. device level) at present, however, integrating the individual all-2D device into an electronic system (i.e. system level) remains challengeable.

Last but not least, we further outlooked the research developments of 2D vdW heterostructures, not limited to all-2D heterostructures. Firstly, realizing the large-scale production of high-quality 2D vdW heterostructures with minimized crystal defects, controllable thickness, and stacking orientation have always been a great concern. Second, developing competitive metallic 2D materials to replace graphene and metal electrodes in thin-film electronics for lowering the interfacial contact resistance, such as Weyl semimetals and topological insulators (e.g. Bi2Te3). Third, device reliability and stability should be concerned and addressed, especially for air-sensitive 2D materials (e.g. BP and InSe). Besides, thermal expansion and contraction should not be overlooked, because mismatched thermal properties of different 2D thin flakes might lead to interfacial strain under various operating temperatures. Fourth, the band alignment engineering of 2D vdW heterostructures by introducing localized fields attracted attention, such as the floating gate field and ferroelectric polarization field. In a word, the 2D vdW heterostructure is a hot research field and also provides the possibility to shape the future of next-generation electronic and optoelectronic devices.

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