Academic Journal of Science and Technology
ISSN: 2771-3032 | Vol. 22, No. 1, 2026

Graphene-TMDs Van der Waals Heterojunction Photodetectors: From Mechanisms and Fabrication to Performance and Applications

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Abstract: Photodetectors, as the core devices for photoelectric signal conversion, are widely used in key fields such as optical communication, imaging, and sensing. The rise of two-dimensional materials provides a new idea for solving the material bottleneck of traditional photodetectors. Graphene, with its ultra-high carrier mobility and broad spectral response advantages, has become an ideal platform for constructing high-speed photodetectors. However, its zero-bandgap property leads to problems such as low photoresponsivity and high dark current. Transition metal dichalcogenides (TMDs) have a suitable direct bandgap and strong light-matter interaction, but their carrier mobility is relatively low. By constructing van der Waals heterojunctions of graphene and TMDs, the advantages of both can be fully utilized to achieve efficient separation and transport of photogenerated carriers, significantly improving the photodetection performance.

Keywords: Graphene; Transition metal dichalcogenides; Heterojunction; Photodetector; Van der Waals heterojunction.

1. Introduction

1.1. Research Background, Purpose, and Significance

Photodetectors are semiconductor devices for photoelectric conversion. After the light signal enters the photoelectric semiconductor material, photogenerated carriers are generated. Under the action of a bias voltage, light signals of different powers can be converted into corresponding photocurrent signals to achieve photoelectric conversion, and they have a wide range of applications in various industries. In recent years, two-dimensional materials have provided new ideas for solving the material bottleneck of photodetectors due to their unique electronic and optical properties. Graphene, as a two-dimensional material, has ultra-high carrier mobility, broad spectral response ability, and flexibility. However, the zero-bandgap characteristic results in low photoresponsivity and high dark current of graphene, making it difficult to meet the application requirements of the new generation of optoelectronic devices and greatly limiting its application in high-performance photodetection. Transition metal dichalcogenides (TMDs), such as MoS_2, WS_2, $MoSe_2$, and WSe_2, have a suitable direct bandgap, can absorb photons in the visible to near-infrared region, and have strong light-matter interactions. However, the carrier mobility of TMDs is 10 - 100 $cm^2 V^{-1} s^{-1}$,resulting in a slow response speed and low carrier transport efficiency of photodetectors[3]. To combine the advantages of graphene and TMDs and make up for their respective limitations, researchers have proposed the strategy of constructing heterojunctions of graphene and TMDs. Such heterojunctions are usually combined by van der Waals forces, without considering the problem of lattice matching, and can combine two-dimensional materials with different characteristics to realize band engineering and optimization of photoelectric properties[1]. The graphene/TMDs heterojunction can effectively promote the separation and transport of photogenerated carriers, broaden the spectral response range, and improve the photoresponsivity, detectivity, and response speed[2,3]. In recent years, research groups worldwide have made a series of breakthroughs in the directions of controllable preparation of heterojunctions, precise interface regulation, and device structure optimization, promoting the development of such devices towards practicality and integration.

1.2. Research Status at Home and Abroad and the Framework of This Paper

Research on two-dimensional material heterojunction photodetectors has formed a mature research field worldwide. Graphene-TMDs van der Waals heterostructures stand out as a key research hotspot due to their complementary properties.

Globally, research efforts fall into two major directions. Some studies focus on fundamental mechanisms, including heterojunction stacking, charge transfer and infrared photodetection performance[4]. Other works prioritize engineering applications, concentrating on interface modulation, scalable fabrication and device integration to advance practical implementation[5]. Most existing researches only discuss individual technical segments, rather than the full research chain.

Published reviews also have obvious limitations. Yang et al. (2022) elaborated the optoelectronic properties of TMDs heterostructures, but ignored the synergistic effect of graphene and relevant fabrication and application issues[6]. Ahn et al. (2025) summarized advances in diverse 2D heterostructure photodetectors, yet failed to conduct a full-chain analysis targeting graphene-TMDs systems from mechanism to practical use. Up to now, a systematic review covering mechanisms, fabrication, performance regulation and applications of graphene/TMDs heterojunction photodetectors is still absent[7].

This paper consists of four parts. The introduction presents the research background, significance and overall structure. The second part illustrates material fundamentals, fabrication methods and performance regulation strategies. The third part

discusses current challenges and future prospects. The final part concludes the whole work, summarizes existing achievements and limitations, and proposes future research directions.

2. Material Systems and Device Technologies

2.1. Material Basis and Band Engineering of Graphene and Transition Metal Dichalcogenide Heterojunctions

2.1.1. Photoelectric Properties of Graphene and Its Role in Heterojunctions

Graphene is a honeycomb two-dimensional crystal formed by single-layer carbon atoms through sp^2 hybridization. It has a uniform intrinsic light absorption coefficient, exhibits a broadband absorption of approximately 2.3% across the visible to near-infrared spectrum. Its spectral response range covers the ultraviolet, visible, near-infrared to terahertz bands, making it an ideal candidate material for broadband photodetectors. At the same time, graphene has ultra-high carrier mobility and excellent conductivity, which can quickly transport photogenerated carriers and provide strong support for the research and development of high-speed response devices. The zero-bandgap characteristic of graphene can be effectively compensated by constructing a heterojunction with TMDs. In the heterojunction, graphene can play multiple roles: First, as a transparent conductive electrode, it can replace traditional metal electrodes, reduce contact resistance, and enhance light transmittance. For example, the graphene/$PdSe_2$/Ge heterojunction can effectively improve the performance of the device in the deep ultraviolet to mid-infrared band. Second, as a carrier transport channel, in the WS_2 quantum dot/RGO(Reduced Graphene Oxide) heterojunction, the high mobility is used to quickly extract photogenerated carriers generated in the TMDs layer, suppress recombination, and improve detection efficiency. Third, it can assist in charge separation through the built-in electric field at the interface, form a band bending with TMDs, and drive the directional transfer of photogenerated carriers. For example, in the graphene/MoS_2 heterojunction, electrons are quickly transferred from MoS_2 to graphene, significantly improving the photoresponsivity[4,10].

2.1.2. Photoelectric Properties of TMDs and Their Selection

TMDs have a layered graphene-like structure with the general formula MX_2, where M is a transition metal element and X is a chalcogen element. Most single-layer samples have a direct bandgap, and the bandgap can be precisely controlled through element composition and the number of layers. The bandgap of single-layer MoS_2 is about 1.8 eV, WS_2 is about 2.0 eV, $MoSe_2$ is about 1.5 eV, and WSe_2 is about 1.6 eV. This suitable and adjustable direct bandgap enables TMDs to produce strong light absorption and photoluminescence in the visible to near-infrared region, making them ideal light absorption layer materials for constructing high-sensitivity photodetectors[2,7]. In addition, some TMDs have a crystal anisotropic structure, showing significant polarization sensitivity.

When constructing a graphene/TMDs heterojunction, the selection of TMDs needs to comprehensively consider factors such as bandgap size, light absorption range, carrier mobility, and band matching with graphene. For visible light detection, MoS_2 or WS_2 with a bandgap in the visible light range can be selected; for near-infrared detection, $MoSe_2$, WSe_2, or $MoTe_2$ with a smaller bandgap are suitable; for polarization-sensitive detection, anisotropic TMDs such as $PdSe_2$ and $ReSe_2$ are ideal choices[2,6].

2.1.3. Band Alignment and Carrier Separation Mechanism of Heterojunctions

The interface band alignment of graphene/TMDs heterojunctions is categorized into three configurations: Type-I (straddling type), Type-II (staggered type), and Type-III (broken-gap type) [1,4]. Type-II heterojunctions are most widely adopted for photodetectors. In this structure, the conduction band minimum and valence band maximum reside in two different materials, which enables spatial separation of photogenerated electrons and holes and effectively suppresses carrier recombination. This working principle corresponds to the photovoltaic effect, which features fast response speed yet relatively low photoconductive gain. Taking the graphene/WSe_2 heterojunction as a typical example, photoexcited electrons from WSe_2 rapidly transfer to the graphene layer, while holes are confined within WSe_2. The interfacial hole injection time is as short as 0.35 ps, demonstrating the outstanding rapid carrier separation capability of the photovoltaic effect[6,8].

Apart from the photovoltaic effect, graphene/TMDs heterojunctions also exhibit the photogating effect, which originates from distinct physical mechanisms and presents opposite performance characteristics. In this case, photogenerated carriers in the TMDs layer are captured by intrinsic defects or interfacial states, forming localized charge accumulation regions. Such trapped charges continuously modulate the electrical conductivity of graphene and thus produce remarkable photocurrent gain. Different from the photovoltaic effect, the trapping and de-trapping processes of carriers inevitably slow down the device response, while delivering ultra-high photoresponsivity and gain. For instance, in WS_2/WSe_2 nanodot composite photodetectors, WSe_2 nanodots serve as efficient hole traps. Under light illumination, the accumulated holes allow cyclic utilization of electrons in the WS_2 channel, achieving a superior photoresponsivity of 8.4×10^2 A/W and a high gain of 2.0×10^3[8].

Overall, the photovoltaic effect and photogating effect show an obvious trade-off between response speed and photoelectric gain: the photovoltaic effect is suitable for scenarios requiring ultrafast detection, whereas the photogating effect is more applicable to weak-light detection where high gain is prioritized over response rate. Furthermore, the Schottky barrier formed between graphene and TMDs can be flexibly tuned via gate voltage regulation or chemical doping, so as to further optimize carrier injection efficiency [3,5,8].

2.1.4. Stacking Structure and Interface Regulation

Common heterojunction stacking structures are divided into vertical heterojunctions and lateral heterojunctions. The layers are only combined by van der Waals forces, and the stacking order affects the band structure by regulating the interlayer interaction. Interface defects, doping, and impurity adsorption can all cause non-radiative recombination of carriers, which is the core reason for performance degradation. Through means such as AlO_x interface passivation and defect engineering, interface recombination can be suppressed, and the response range of the WS_2/Ge heterojunction can be extended from 200 nm (deep ultraviolet) to 4.6 μm (mid-

infrared).

2.2. Heterojunction Preparation Methods and Types

2.2.1. Vapor Deposition Method

Chemical vapor deposition (CVD) is the main method for preparing large-area, high-quality graphene and TMDs thin films. The CVD growth of graphene usually uses metal foils such as Cu or Ni as substrates, and carbon source gases such as methane are introduced at high temperatures for decomposition and deposition. TMDs use metal oxides or metal halides as metal sources and sulfur powder, selenium powder, etc. as chalcogen sources to deposit TMDs thin films on the substrate. By directly synthesizing WS_2 nanodisks on graphene through a layer-by-layer, transfer-free CVD method, defects and contamination introduced during the transfer process can be effectively avoided, and the quality of the heterojunction interface can be improved. Physical vapor deposition (PVD) methods such as pulsed laser deposition (PLD) are suitable for the preparation of complex heterojunctions and can achieve the deposition of material combinations that are difficult to grow by CVD[3,6].

2.2.2. Liquid-Phase Exfoliation and Solution Processing Methods

Liquid-phase exfoliation (LPE) is a low-cost, large-scale method for preparing two-dimensional material nanosheets. Through means such as ultrasound, shear force, or electrochemistry, bulk two-dimensional materials are exfoliated into single-layer or few-layer nanosheet dispersions in solvents. This method is simple to operate and low in cost, and is suitable for preparing heterojunction dispersions for solution-processed devices. The photoresponsivity of the MoS_2/graphene vertical heterojunction photodetector prepared by modified LPE is about 7 times higher than that of the traditional solution method[4,5].

2.2.3. Transfer Stacking Technology

Transfer stacking technology is the core method for constructing van der Waals heterojunctions, which allows two-dimensional materials prepared by different methods to be precisely stacked together to form various complex heterostructures. A typical transfer process includes mechanically exfoliating two-dimensional material flakes from bulk crystals, picking them up with transparent tape or elastic polymers, and finally precisely aligning and transferring them to the target substrate or another layer of two-dimensional material[2,3]. The core difficulty of transfer stacking technology lies in achieving precise transfer without damage and contamination, and avoiding the introduction of bubbles, wrinkles, and impurities during the stacking process. Despite the challenges, this technology is still an important means for preparing various novel graphene/TMDs heterojunctions and exploring their physical properties and device applications.

2.2.4. Construction of Hybrid-Dimensional Heterojunctions

In addition to the van der Waals heterojunctions formed between two-dimensional materials, graphene/TMDs can also be combined with materials such as 0D quantum dots, 1D nanowires, and 3D bulk semiconductors to form hybrid-dimensional heterojunctions. This structure can integrate the advantages of materials of different dimensions and further expand the performance and functions of devices. For example, the WS_2 quantum dot/1D reduced graphene oxide flexible fabric forms an ultraviolet photodetector with good flexibility and wearability; the combination of graphene/TMDs/3D semiconductor materials (Si, Ge) can achieve self-powered broadband detection[4,8].

2.3. Device Performance Parameters and Regulation Strategies

2.3.1. Key Performance Parameters and Their Characterization

The key parameters for evaluating the performance of graphene/TMDs heterojunction photodetectors include photoresponsivity (R), specific detectivity (D*), response speed (rise time τ_r and fall time τ_d), dark current (I_{dark}), spectral response range, and polarization sensitivity. Photoresponsivity R is defined as the ratio of photocurrent to incident light power, $R = I_{ph}/P_{in}$, with the unit A/W. Graphene/TMDs heterojunctions can usually achieve a higher R value than single materials through efficient charge separation and transport and the photogating gain mechanism. Specific detectivity $D^* = R\sqrt{(A/2qI_{dark})}$ (A is the device area, q is the elementary charge, and Idark is the dark current), which characterizes the weak light detection ability and requires both high R and low I_{dark}. The response speed determines the device's ability to follow changes in the light signal. The high carrier mobility of graphene helps to achieve a fast response, but the carrier trapping and release processes in TMDs may slow down the response speed[2,3].

2.3.2. Strategies for Improving Photoresponsivity and Detectivity

Improving photoresponsivity and detectivity is one of the core goals of research. The main strategies include enhancing light absorption, promoting carrier separation and transport, and reducing dark current and noise. Light absorption can be enhanced through localized surface plasmon resonance (LSPR). For example, in the WS_2 nanodisk/graphene heterojunction, the LSPR effect generated by the WS_2 nanodisks increases the photoresponsivity by 7 times compared with the continuous WS_2 layer structure. Introducing optical microcavities or photonic crystal structures and combining TMDs with perovskites or quantum dots can also enhance the light absorption intensity. Promoting carrier separation and transport mainly depends on optimizing the band structure and interface properties of the heterojunction. Constructing a Type-II heterojunction can use the built-in electric field to separate photogenerated electrons and holes; introducing graphene as a transparent electrode and carrier transport layer can reduce contact resistance and accelerate carrier extraction; interface engineering (such as the AlO_x passivation layer) can reduce interface defect states and suppress carrier recombination. Reducing dark current and noise is crucial for improving D*[3,5,10]. Working in the self-powered mode (zero bias) is the most direct way to reduce dark current, which is usually achieved by constructing a p-n heterojunction using the photovoltaic effect.

2.3.3. Strategies for Optimizing Response Speed

The response speed is affected by the processes of carrier generation, separation, transport and recombination. Graphene's high carrier mobility facilitates rapid carrier transport, and shortening the device channel length can markedly reduce carrier transit time. Defect states at heterojunction interfaces promote non-radiative

recombination and prolong the response time. This issue can be effectively mitigated through interface passivation and defect engineering [11].

Although the photogating effect can improve photoresponsivity, it usually leads to a slower response speed. To address this limitation, specially designed heterojunction structures can be adopted. The built-in electric field within such structures helps accelerate the release of trapped carriers, and material combinations with ultrafast exciton dynamics can also be utilized. Furthermore, optimizing electrode design and miniaturizing device dimensions can lower parasitic parameters and extend the cutoff frequency of devices [7,9].

2.3.4. Regulation Mechanism and Applications of Polarization Sensitivity

High-performance polarization-sensitive photodetectors can be fabricated by combining graphene with anisotropic transition metal dichalcogenides (TMDs) such as $PdSe_2$ and $ReSe_2$. Graphene/$PdSe_2$/Ge heterojunctions achieve broadband polarization-sensitive photodetection covering the deep ultraviolet to mid-infrared range. Their maximum polarization sensitivity reaches 112.2, one of the highest values ever reported among two-dimensional material-based photodetectors[4]. In contrast, most similar devices deliver polarization sensitivity lower than 80, and typical anisotropic TMDs photodetectors only achieve 30 to 75. These devices are promising for applications including military reconnaissance and remote sensing imaging [2,7,12].

3. Core Challenges and Future Development Directions

3.1. Major Existing Challenges

3.1.1. Difficulties in Precise Control of Interfacial Quality

Wrinkles, impurities and interfacial bubbles introduced during the transfer process create abundant defect states, which exacerbate carrier non-radiative recombination and degrade the consistency of device performance. To date, no industrial breakthrough has been achieved in the controllable fabrication of atomically clean

interfaces [5,10].

3.1.2. Bottlenecks in Scalable Fabrication

Mechanical exfoliation and precise transfer techniques used in laboratory settings are only suitable for small-batch samples. When adopting CVD to synthesize large-area uniform graphene/TMD heterostructures, the process suffers from poor stability and low production yield, which fails to meet industrialization requirements [6].

3.1.3. Performance Trade-off Dilemma

The photogating effect can drastically boost photoresponsivity, but it slows down carrier trapping and release, which in turn sacrifices response speed. It is extremely difficult to achieve high photoresponsivity and ultrafast response simultaneously [3,4].

3.1.4. Poor Device Stability and Integration Compatibility

TMDs are susceptible to oxidation and degradation under ambient air and humidity, resulting in short device service lifetime. Moreover, the issues related to thermal budget and interfacial compatibility between two-dimensional material devices and conventional silicon-based CMOS processes remain unsolved, restricting on-chip integration [9].

3.1.5. Mismatch Between Cost and Industrialization

High-quality two-dimensional materials and high-precision transfer equipment incur high costs. Although solution processing is low-cost, the performance of its derived devices is far inferior to that of samples prepared by vapor deposition. Thus, it is challenging to strike a balance between cost and device performance [6].

3.2. Future Development Prospects

3.2.1. Precise Regulation of Atomically Clean Interfaces

Develop transfer-free direct CVD growth, atomic layer deposition (ALD) passivation and in-situ doping technologies to effectively suppress interfacial defects and construct ultra-clean heterojunction interfaces [5,10].

3.2.2. Low-Cost Scalable Fabrication Technologies

Explore large-scale manufacturing approaches including roll-to-roll CVD, inkjet printing and solution spin-coating. These techniques enable the continuous production of large-area, high-uniformity heterojunction films and advance the industrial implementation of such devices [6].

3.2.3. Breakthroughs in Performance Trade-offs

Design graded energy bands and multi-stage carrier transport channels, and employ material systems with ultrafast exciton dynamics. In this way, high photoresponsivity, high detectivity and ultrafast response can be realized concurrently [3,4,10].

3.2.4. Multifunctional Integration and Flexible Devices

Integrate multiple functions such as self-powered operation, polarization sensitivity, flexible wearability and photoelectric sensing. Develop lightweight integrated devices targeting the Internet of Things and biomedical applications [7,13].

3.2.5. Silicon-Based Heterogeneous Integration

Realize monolithic integration of graphene/TMD heterojunctions with silicon and germanium-based optoelectronic chips. The proposed structures are compatible with existing semiconductor processes, and can be extended to high-end applications such as optical communication chips and infrared imaging arrays [9].

3.2.6. Exploration of New Material Systems

Combine two-dimensional materials with emerging materials including perovskites, quantum dots and topological insulators, so as to expand the detection wavebands to the deep ultraviolet and terahertz regions. In addition, develop cutting-edge devices such as single-photon detectors and room-temperature infrared photodetectors [5,8,14].

4. Conclusions

4.1. Main Research Conclusions

This paper systematically reviews the research progress of photodetectors based on graphene/TMD van der Waals heterostructures. The key conclusions are drawn as follows. Van der Waals heterostructures constructed by graphene and TMDs can effectively compensate for the inherent drawbacks of single-component materials. Type-II band alignment acts as the core mechanism for achieving efficient carrier separation. Interface modulation, defect engineering and fabrication process optimization are critical strategies to improve device performance. Such photodetectors exhibit unique advantages in broadband detection, high-speed response and polarization sensitivity, and are capable of meeting the photodetection demands of diverse application scenarios.

4.2. Research Innovations and Limitations

Innovations: This paper comprehensively summarizes the material properties, band structure mechanisms, fabrication methods and performance regulation strategies of graphene/TMD heterostructures. It incorporates both overseas studies on fundamental mechanisms and domestic research on practical applications, forming a full-chain literature review. This work also clarifies the core routes for performance improvement and major industrialization bottlenecks of such heterostructures, offering a systematic reference for subsequent research.

Limitations: As a literature review, this study does not involve experimental verification. In-depth analysis regarding the quantitative regulation of interfacial defects in heterostructures, technical details of scalable production processes, and device stability under extreme environments remains to be further supplemented. In addition, this review does not cover cutting-edge sub-directions such as terahertz detection and single-photon detection.

4.3. Future Research Directions

Subsequent studies should focus on core bottlenecks and prioritize the following key directions:

1.Establish a quantitative atomic-level interface characterization and regulation system, clarify defect-performance correlations, and ensure the reproducibility of high-performance devices.

2.Optimize low-cost scalable processes, develop standardized parameters and industrial-grade evaluation criteria, address film uniformity and stability issues, and promote engineering transformation.

3.Strengthen interdisciplinary collaboration, advance "material-device-circuit-system" co-design, break through silicon-based process compatibility barriers, and accelerate high-end application deployment.

4.Explore the integration of emerging material systems, focus on solving fundamental issues such as photoelectric mechanisms of multi-component composites and long-term stability of flexible devices, and expand research boundaries.

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