Graphene Two-Dimensional Heterostructure and Its Application in High-Efficiency Optoelectronic Devices

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Abstract: Graphene is a highly promising material for future optoelectronic devices due to its outstanding electronic and optical characteristics. Its integration into advanced systems offers solutions to the stability challenges faced by traditional solar cells. This article begins by outlining the chemical vapor deposition methods used to synthesize high-quality graphene and discusses strategies for optimizing its interface engineering within devices. These developments are shown to improve the separation and transport of photogenerated carriers in two-dimensional heterostructures, thereby boosting photoelectric conversion efficiency. Moreover, graphene's outstanding conductivity and high optical transparency. make it ideal for applications as transparent electrodes, carrier transport layers, and enhancements to photoactive materials. By systematically examining these roles, the article highlights graphene's transformative potential in optoelectronics while also addressing the technical challenges and future application prospects. Overall, graphene has the potential to play a central role in the evolution of optoelectronic technologies, with ongoing research paving the way for practical and scalable implementations.

1 INTRODUCTION

As a groundbreaking two-dimensional material, graphene has drawn considerable interest in optoelectronics due to its remarkable electronic, optical, and mechanical properties. Its ultra-high carrier mobility, wide spectral absorption range, ultra-fast light response speed, and excellent mechanical flexibility make it a key candidate material for new optoelectronic devices. In recent years, the swift advancement of low-dimensional materials science has prompted researchers to engineer heterostructures by integrating graphene with other two-dimensional materials, which not only regulates the interface electronic states, but also significantly improves the separation and transmission efficiency of photogenerated carriers, providing new research ideas to enable high-performance optoelectronic device design

Within the domain of photovoltaic devices and solar cells, the unique advantages of graphene provide unprecedented opportunities for the improvement of photoelectric conversion efficiency. Its high conductivity and optical transparency make it an ideal alternative material for transparent electrodes, which can effectively replace the traditional indium tin oxide (ITO). In addition, the excellent charge transport ability of graphene helps to reduce carrier recombination loss and improve charge collection efficiency. More importantly, the tunability of graphene energy band structure and its functionalization characteristics enable it to achieve a good match with a variety of light-absorbing materials, to optimize the interface contact, and solve the problem of low photoelectric conversion efficiency of traditional materials.

Graphene and two-dimensional heterostructures show great potential in the field of optoelectronic devices, especially in improving response speed, sensitivity, and stability. Despite these advances, practical applications face significant challenges, especially in the large-scale fabrication of high-

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quality interfaces, lack of graphene band gap, difficulty in accurate control of energy band alignment and interface charge transport, interlayer coupling, and Mohr potential regulation. In addition, the balance between high performance and stability, flexibility, and integrability in device engineering is also a technical problem. Therefore, promoting high-quality two-dimensional material synthesis, accurate design and regulation of heterostructures, and development of new device structures are urgent problems to be solved.

Herein, we offer a detailed review of the recent research trends in graphene in new optoelectronic devices, focusing on its application in two-dimensional heterostructures and efficient optoelectronic devices. By analysing the core role of graphene and its performance in different optoelectronic devices, this review seeks to offer theoretical suggestions for the development of optoelectronic technology in the future, and prospects the future direction of graphene in the optoelectronic field

2 PREPARATION AND PHOTOELECTRIC PROPERTIES OF GRAPHENE

2.1 Structure and Properties of Graphene

Graphene is a highly conductive material that is composed of carbon atoms arranged in a hexagonal lattice structure. A carbon atom is composed of a unique set of electron arrangements and is structured within a hexagonal lattice framework. As depicted in the provided illustration, a single molecular crystal structure is observed (Muthuvinayagam et al., 2023). In the 2023 study, it was determined that each carbon atom within graphene forms sp^2 hybridized orbitals (as depicted in Figure 1). The structural integrity of the monolayer graphene is maintained by the strong intermolecular forces known as van der Waals interactions.

As depicted in Figure 2, the graphene structure formed from carbon-carbon covalent bonds demonstrates a unique lattice arrangement. In the cell's internal structure, the carbon atoms A and B are positioned such that they are not aligned directly with one another, thereby creating two separate triangular lattice-like two-dimensional frameworks.

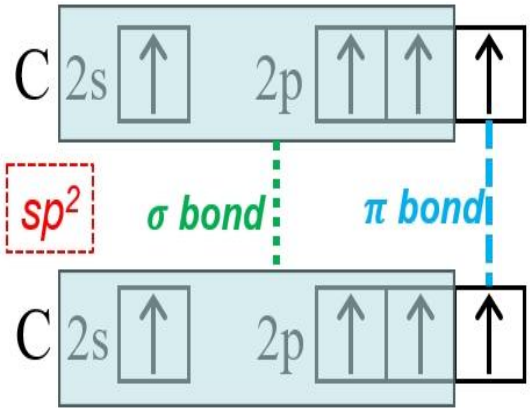

Figure 1: Carbon hybrid track (Original)

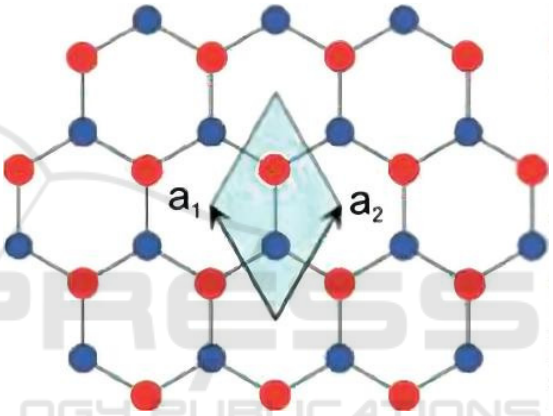

Figure 2: Graphene layer network (Biro, Nemes-Incze, and Lambin, 2012)

Note: the light blue highlighted area represents the cell, which is represented by vectors a1 and a2. It contains two atoms from sublattice a (blue) and b (red)

Graphene has a unique electronic structure and electrical properties. By applying gate voltage and other means, it is possible to induce the conductivity of electrons or holes in graphene (where holes represent a loss of electrons with positive charge), which is like the conductive phenomenon in Semiconductors (Katznelson, 2007). However, due to its special electronic structure, unlike semiconductors, graphene has no insulating state, which gives it higher conductivity and makes not sensitive to changes in the external environment.

Due to the extremely thin monolayer structure of graphene, it is almost transparent, so it can absorb more incident light in a wide band range (Bonaccorso et al. 2010). Due to these inherent characteristics, graphene has a significantly lower electronic

transition energy threshold, which can efficiently use incident photons and generate photocurrent, making graphene have significant advantages in the application of high-performance optoelectronic devices.

2.2 Preparation of Graphene by CVD

To obtain graphene with high conductivity, chemical vapor deposition (CVD) is a preferred method. The principle is achieved by cracking carbon sources (such as methane and ethylene) (Gong et al. 2021). The growth mechanism can be divided into two types, which depend on the solubility of the metal substrate in carbon. When the solubility is high, the carburization-decarburization mechanism can be used (at high temperature, carbon atoms penetrate the metal substrate and then diffuse to the metal surface through rapid cooling, thus forming a graphene layer). On the contrary, the surface growth mechanism can be used (at high temperature, carbon atoms are pyrolyzed from a gaseous carbon source and adsorbed on the metal surface, and finally form a continuous graphene film) (Losurdo et al. 2011).

2.3 Photoelectric Conversion Efficiency of Graphene

Graphene plays an important role in batteries, especially in energy storage, ion loading, and transport. Graphene can not only be used as an active material and catalyst in lithium battery slurry but also plays an important role in organic solar cells (OSCs).

The electrodes of OSCs need to be highly transparent and conductive. Therefore, transparent conductive electrodes need to achieve the best balance between transparency (T) and conductivity (σ) (Li et al. 2008). This balance can be achieved by the following formula:

$$T = (1 + [M_0/2Rs][X_0/X_{2d}])^2 \quad (1)$$

Of which, M_0 is free space impedance, X_0 and X_{2d} represent optical conductivity and two-dimensional conductivity, respectively. The resistance Rs is proportional to the number of layers (n), which can be figured up by the following formula: $Rs = (nX_{2d})^{-1}$ (Sun et al., 2015).

According to this formula, graphene can be modified and doped to optimize its performance. When the n is around 4, a better balance between transparency and conductivity can be achieved (Mahmoudi, Wang, and Hahn, 2018). This makes graphene more advantageous than other materials in the market.

3 HETEROSTRUCTURE OF GRAPHENE AND OTHER MATERIALS

3.1 Concept of Two-Dimensional Structure and Two-Dimensional Heterostructure of Graphene

3.1.1 Two-Dimensional Structure

Graphene is a type of two-dimensional layered material (2DLMs), where each layer consists of a two-dimensional lattice formed by covalently bonded carbon atoms. It is worth noting that the neighbouring layers are physically linked through relatively weak intermolecular forces known as van der Waals interactions, which play a critical role in determining their structural integrity and overall behaviour (Yi et al. 2019). These intrinsic properties form the cornerstone upon which the surface behaviour and heterostructure formation are built.

The charge carriers within graphene operate with an intrinsic massless nature and exhibit extraordinary mobility, which significantly enhances their transport properties. Quantum Hall Effect (QHE) has recently demonstrated remarkable potential to be observed at temperatures that surpass conventional limits, with recent experimental results indicating feasibility even near room temperature.

By utilizing the framework of density functional theory (DFT), recent research by a team of scientists has demonstrated that when water droplets are physically adsorbed onto the surface of graphene, they significantly alter its electrical characteristics (Taherian et al., 2013). The findings indicate that graphene exhibits a notable level of hydrophobic properties, which suggests it effectively repels water. The hydrogen bonds within water molecules in liquid droplets are significantly stronger than the energy required to adsorb these molecules onto a single-layer graphene surface. As a result, its influence on the electrical characteristics of graphene is minimal. The recent findings demonstrate that graphene's unique properties make it an exceptional candidate for protecting different substrate surfaces while simultaneously maintaining their inherent wettability characteristics. The surface characteristics of these materials play a pivotal role in the development of graphene-based composites and optoelectronic devices, as highlighted by the research (Dai, Wang, and Wu, 2016).

3.1.2 Introduction of Heterostructure

In optoelectronic devices, graphene alone cannot achieve all functions, but by forming a

heterostructure with other materials, it can achieve functions that a single material cannot achieve. In 2010, Dean and other scientists proved that hexagonal boron nitride (hBN) can be used as an ideal two-dimensional substrate material for graphene, because hBN has excellent electrical insulation, excellent thermal conductivity and excellent lubrication performance (Wang et al. 2014).

The vertical stacking of graphene and hBN can be achieved by van der Waals forces. However, if the transverse connection structure is to be formed, the manufacturing process will be more complex. To obtain shape-controllable graphene hexagonal boron nitride (G-hBN) heterostructures, a method has been proposed, as shown in Figure 3.

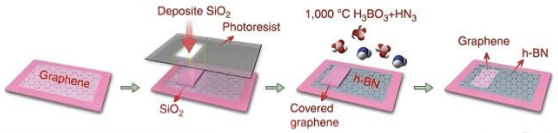

Figure 3: Shape-controlled transverse g-hBN heterostructure fabrication: the conversion process of spatial control (Li et al., 2009)

Graphene hBN heterostructure can be obtained by covering the required graphene region with silica, using boric acid as a boron source and ammonia as a nitrogen source to convert G-hBN at high temperature. Because graphene has high carrier mobility, it can quickly absorb the generated photoelectrons and control the conductive polarity of the storage unit in the device, which makes it of great significance in the application of photodetectors and solar cells (Wang et al., 2014).

3.2 Graphene Heterostructure

Although the zero band gap semiconductor properties make graphene have ideal photoelectric properties, its optical absorption is low, only about 2.3%. To overcome this limitation, it has become a widely used strategy to form a heterostructure between quantum dots (QDs), a material with excellent optical absorption properties, and graphene (Gan et al. 2012).

As shown in Figure 4, when the quantum dot is hybridized with neutral graphene, electrons are transferred from the quantum dot to graphene, thereby generating a built-in electric field at the graphene/quantum dot interface (Song et al. 2015). Under illumination, quantum dots absorb light and generate electron-hole pairs, which are separated at the graphene/quantum dot interface. The built-in electric field transfers holes to graphene, while retaining electrons in quantum dots.

The strong and tunable light absorption characteristics in the quantum dot layer can effectively generate and transfer charges to graphene. Thanks to the high charge mobility of graphene, these 1V rw111charges can be circulated in graphene many times, which significantly improves the photoelectric conversion efficiency of devices (Konstantatos et al. 2012).

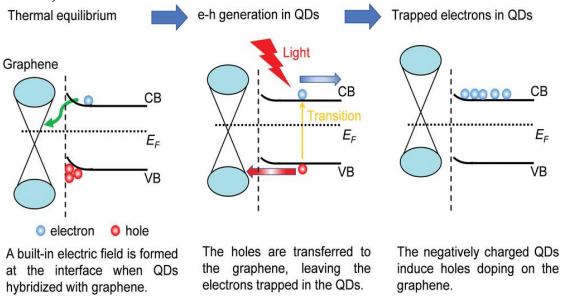

Figure 4: Energy level diagram of graphene/quantum dot interface (Konstantatos et al., 2012)

As shown in Figure 4, quantum dots (QDs) can enhance the scattering and absorption of incident light, so they can effectively improve the response current and responsivity of photodetectors.

Additionally, the graphene/quantum dot heterostructure can significantly improve the performance of the visible light detector. Graphene-based photodetectors can be divided into three types according to their response spectral range: ultraviolet photodetectors, visible photodetectors, and infrared photodetectors. Graphene/quantum dot heterostructures have unique advantages, which can combine the strong light absorption ability and quantum effect of quantum dots with the high charge mobility of graphene. This combination plays an important role in achieving high response and detection. The performance of photodetectors and organic solar cells (OSC) can be further optimized by treating the surface of quantum dots, effectively doping quantum dots, and adjusting the size of quantum dots.

4 APPLICATIONS OF GRAPHENE IN THE OPTOELECTRONIC FIELD

4.1 Graphene as a Transparent Electrode

Indium tin oxide (ITO) remains the predominant transparent electrode employed in photovoltaic

devices due to its favourable photoelectric characteristics, notwithstanding challenges such as limited indium availability, mechanical fragility, and chemical instability. (Lagerwall & Scalia, 2012). Graphene has mechanical flexibility, rich resources, and excellent photoelectric performance, which perfectly makes up for the shortcomings of ITO, improves the durability and stability of devices, and reduces the cost.

In 2020, Koo et al. (2022) Introduced a novel form of transparent electrode integrating polyimide and CVD graphene, which has excellent conductivity and mechanical stability, and achieved high efficiency (15.2% PCE) close to traditional rigid devices in flexible organic solar cells. In 2024, Bourahla et al. Optimized graphene/ITO composite electrode by introducing silver nanowires (Bourahla et al., 2024), which significantly improved the charge transfer ability, reduced the resistance, and maintained high transmittance, showing good device adaptability. Fang et al. Introduced silver-doped graphene electrode structure into micro light-emitting diodes (Fang et al., 2025), which effectively improved the radiation efficiency and device performance. These studies further verified the potential of graphene in a new generation of flexible and high-performance optoelectronic devices.

In summary, graphene has made major advancements in transparent electrode materials in recent years. By combining with PI, Ag NW, and ITO, researchers have effectively overcome the shortcomings of traditional ITO electrodes in mechanical stability, chemical durability, and resource sustainability. Graphene electrode shows excellent photoelectric performance and application potential in the fields of flexible optoelectronic devices, heterojunction solar cells, and μ led. In the future, further optimizing the preparation process of graphene electrodes and improving their interface engineering and charge transfer characteristics will provide strong support to facilitate the fabrication of high-performance and reliable next-generation optoelectronic devices

4.2 Role of Graphene in Charge Carrier Transport

Graphene plays an important role in boosting the performance of solar cells by leveraging its ultra-high carrier mobility, two-dimensional structure, and excellent electrical properties. As an electrode or transport layer, it can significantly accelerate charge transmission, reduce resistance loss, and improve current density. By constructing a heterojunction with semiconductor materials, it can effectively enhance charge separation efficiency and inhibit composite loss. The out-of-plane π bonds establish weak yet essential interactions between graphene layers through the overlap of π electrons oriented perpendicular to the graphene plane. These interactions facilitate the delocalization of π electrons, forming a cross-layer delocalization network or electron array, which significantly enhances the conductivity of electrons. The highly delocalized π - π conjugate network in graphene not only gives the material excellent conductivity but also provides an ideal transmission path for charge concentration, which further improves the charge collection efficiency (Yi et al., 2020). Pure graphene exhibits a zero-energy band structure. Upon doping, its energy band will open. Graphene optimizes its energy level matching through interface engineering and chemical doping, to reduce the charge transmission barrier, and reduce the composite loss caused by temperature rise by using its excellent thermal conductivity (Jiang et al., 2021).

Heteroatom doping has been demonstrated to be the most effective method to control the band gap engineering, optical phenomenon management, and structural characteristics adjustment in graphene, opening new possibilities for nano-optoelectronic applications, especially in energy-related devices (Sheng et al., 2011). These characteristics enable graphene to show significant efficiency improvement potential in perovskite solar cells, organic solar cells, dye-sensitized solar cells, and other devices. By further optimizing the interface modification, doping process, and material composite strategy of graphene, graphene is expected to assume a more vital role in the development of efficient and stable next-generation solar cells.

4.3 Improvement Strategies for Graphene/Light-Absorbing Layer Heterostructures

Graphene heterostructures have various types, including van der Waals heterostructures, p-n junctions, and hetero band docking structures, showing high electron mobility, adjustable band gap, and excellent thermal conductivity, which have a wide range of applications.

Niu et al. Proposed a new type of heterostructure, which uses the multilayer stacking mode of BN/graphene monolayer/BN and BN/WSe α/ α/graphene monolayer/WSe α/bn. Among them, WSE α, as a typical transition metal disulfide, has excellent electrical and optical properties, and can

effectively shield the superlattice effect, which is often affected by periodic potential disturbance caused by lattice mismatch or interlayer torsion angle. Through the encapsulation of WSE ɑ, the interaction between electrons in graphene is significantly enhanced, thus stimulating a stronger electron correlation effect. The study further reveals that the non-centrosymmetric stacking of boron nitride (BN) layers leads to sliding ferroelectricity in the heterostructure. This is a spontaneous polarization behaviour caused by the relative slip between atomic layers, which is reversible and directionally controllable. In single-layer graphene devices, this sliding ferroelectricity makes the charge polarization switching more robust, and also provides greater flexibility for the selection of material systems, offering a novel design approach for the construction of stable and efficient graphene optoelectronic devices.

Naderi and Ahmad (2023) significantly improved the thermal stability and photoelectric performance of the device by introducing a graphene oxide (RGO) layer into ZnO-based solar cells. As a graphene-derived material with good conductivity and high chemical stability, RGO plays an effective protective role in the device structure. Electrophoretic deposition technology is used in this study, which is a process of uniformly depositing charged particles on the substrate surface by applying an external electric field. RGO layer is introduced upon the surface of ZnO nanorods, which improves the adaptability of the device to high high-temperature environment. Experiments show that RGO can fill the oxygen vacancy defects commonly existing in ZnO, which usually reduce the optical stability of the material. After the introduction of RGO, the device showed a slower decline in efficiency at high temperatures, which further verified the unique advantages of graphene materials in improving the durability of optoelectronic devices.

In general, graphene/light absorbing layer heterostructure shows great potential in improving the optical absorption, carrier transport, and stability of optoelectronic devices. Researchers have significantly improved the efficiency of solar cells, photocatalytic systems, and photodetectors incorporating graphene through innovative design of heterostructures, interface engineering optimization, and material doping. In the future, further exploring the synergy mechanism of graphene in multifunctional heterojunction structures and optimizing its integration strategy with other 2D materials will provide a broader research space for the construction of efficient and stable optoelectronic devices.

5 CONCLUSION

This paper reviews the research progress of two-dimensional heterogeneous optoelectronic devices based on graphene. Graphene synthesis methods, preparation costs, and large-scale manufacturing challenges are the research focus. CVD is suitable for industrialization, but the process is complex, and the cost is high. The redox process has a low cost and high yield, but RGO has defects. High-quality graphene can be prepared by the epitaxial growth method, but the cost is high and depends on the SiC substrate. The performance optimization and stability of graphene in optoelectronic devices are key. Although graphene is conductive, transparent, and flexible, its stability is affected by the environment. The optimization strategy includes surface functionalization, doping technology, and packaging process. These improvements contribute to the application of graphene in photovoltaic cells, photodetectors, LEDs, etc.

The research directions of graphene optoelectronic devices include using artificial intelligence to accelerate material screening and performance prediction, developing ultra-fast optoelectronic devices based on graphene, and improving optoelectronic performance through three-dimensional structure design. To promote the industrialization of graphene materials, it is necessary to develop low-cost, high-efficiency, and environment-friendly synthesis processes, and optimize the uniformity, stability, and controllability of materials. Combined with multidisciplinary means, the application of graphene in intelligent optoelectronic systems, flexible wearable devices, and ultrafast optoelectronic devices will bring new breakthroughs in optoelectronic technology.

AUTHORS CONTRIBUTION

All the authors contributed equally and their names were listed in alphabetical order.

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