The progress of fabricating the 2D materials and heterostructure devices

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Abstract. Contemporarily, the superior performance of low-dimensional heterostructure devices has attracted extensive attention of scientists. In order to offer a clearer understanding of low-dimensional materials and heterostructure devices, this paper introduces the basic concepts of low-dimensional materials and heterostructure fabrication and arranges the devices constructed by two-dimensional heterostructure materials. Primarily, the background information of state-of-art low-dimensional materials is demonstrated. Moreover, starting from the preparation of low dimensional materials, the mainstream methods of fabricating the two-dimensional materials are discussed. Then, the fabrication methods of two-dimensional material heterostructure and the general classification of two-dimensional material heterostructure devices are summarized. Last but not least, the full text is summarized and prospected. This paper aims to provide a more specific reference and guidance for the development of two-dimensional heterostructure devices in the future.

Keywords: Two dimensional materials, heterojunctions, exfoliation, chemical vapor deposition, semiconductor devices.

1. Introduction

The low-dimensional layer of matter has always been a field that scholars want to explore, which has drawn countless attention. After the discovery of graphene in 2004, it set off an upsurge in the research of low-dimensional materials [1]. As the low-dimensional materials have special structures, two-dimensional semiconductor materials are widely considered to possess broad application prospects in nanoelectronics, circuit integration and other fields due to their unique chemical and physical performances different from traditional semiconductor materials [2, 3]. In addition, since there are no dangling bonds on the surface of two-dimensional semiconductor materials, various two-dimensional semiconductor materials can be stacked to form van der Waals heterostructures, resulting in a series of brand-new physical and chemical phenomena.

Firstly, obtaining high performance and high-quality two-dimensional semiconductor materials is an important basis for the preparation of heterostructures and related devices. At present, there are two main methods to prepare two-dimensional semiconductor materials, one is top-down method (e.g., mechanical exfoliation), the other one is bottom-up chemical synthesis method. So far, the main application of two-dimensional semiconductor materials is implemented in the fields of ultra-thin optoelectronic devices, integrated electronic devices and wearable flexible devices [4-7].

In this paper, the current progress of low-dimensional heterostructures devices is summarized. At the first part, different preparation methods of the preparation of two-dimensional materials are organized. In the second part, the methods of constructing two-dimensional material heterojunction are summarized. In the third part, it sorts off the latest applications of heterojunction structure in devices. Afterwards, the limitations of current work are reviewed and the future is prospected. The aim is to provide more clear ideas for other scholars and to guide the development of two-dimensional heterojunction materials and devices in the future.

2. Overview

Since the birth of the world's first electronic computer in the United States in 1946, the way to obtain high performance in electronic devices while saving the volume of equipment has become the most critical and vital problem. In 1952, Geoffrey from the British Radar Research Institute. William Arnold Dummer was the first person to really put forward the concept of a "solid state circuit", or integrated circuit. Since then, the development of integrated circuit, which is the cornerstone of the development of modern information society, has become one of the main forces to promote technology in the information age. Moore's Law, formulated in 1965 by Gordon Moore of Intel, predicted that the integration degree of integrated circuit transistors would double every 18 months. Until May 2021, IBM announced that it had successfully developed the 2nm process, and showed the media the wafers and their microscopic pictures using the 2nm process. The aims and objectives for the development of IC are mainly low power consumption, intelligent, miniaturization etc.

Conventional and common materials have been difficult to fully meet the requirements of deep development. Two-dimensional materials have the characteristics of small size and easy integration [8]. The two-dimensional materials family includes superconductors, metals, semiconductors, insulators and other kinds of materials. For example, layered AuTe2Se4/3 with superconductivity, monolayer NbTe2 with metallic properties, MoS2 and WS2 with semiconductor properties, etc. [9]. In two-dimensional materials, atoms are combined together by covalent bonds, and the atoms between layers are stacked on the top of each other by tiny van der Waals forces. The quantum confinement effect caused by ultra-thin thickness just provides excellent confinement conditions for carriers, hence it is considered as an ideal and excellent material for the construction of a new generation of electronic devices [10]. Unfortunately, it cannot fully meet the current demand for high-performance and multifunctional electronic devices due to the single physical and chemical properties of a single component two-dimensional material. However, the heterostructures formed by the stack of two or more two-dimensional materials show better performance than a single material in various fields [11-14]. Therefore, the study of two-dimensional heterojunction devices plays a crucial role in the further development of information technology.

3. Manufacturing process of two-dimensional materials

In order to realize the wide application of two-dimensional material devices, it is essential to solve the difficulty of large-scale fabrication for high-quality two-dimensional nanomaterials. Nowadays, the mainstream fabrication methods of two-dimensional nanomaterials can be divided into two categories: "top-down" and "bottom-up". The "Top-down" refers to exfoliate the existing materials directly from the block materials based on physical methods supplemented by chemical methods. The "bottom-up" approach is to fabricate two-dimensional nanomaterials by chemical synthesis from the perspective of particle size.

3.1. Top-down fabrication method

3.1.1. Mechanical exfoliation

The first material fabricated by mechanical exfoliation is graphene, which is still applicable to other two-dimensional materials (e.g., TMDs and black phosphorus) [1, 15]. In detail, the procedure of the fabrication can be described as follows (sketched in Fig. 1). Primarily, the two-dimensional material block is placed on a special tape. Subsequently, one needs to stick repeatedly and exfoliate the block material. Afterwards, it is needed to transfer the sample on the special tape to the target substrate and stand still for a period of time. Then, the tape should be slowly removed to ensure that the sample material is transferred to the target substrate. Finally, the optical microscope is used to observe and obtain the required thickness of the material. The material prepared by mechanical exfoliation has high stability and does not destroy the intrinsic structure of the sample ascribed to the high purity. In addition, there is no step to destroy the structure of the sample, i.e., the obtained samples have high quality and few defects. Although the method is simple to operate, its efficiency is low and it is unmanageable to control the thickness, size and shape of the exfoliation. The mechanical exfoliation is mostly operated in the laboratory.

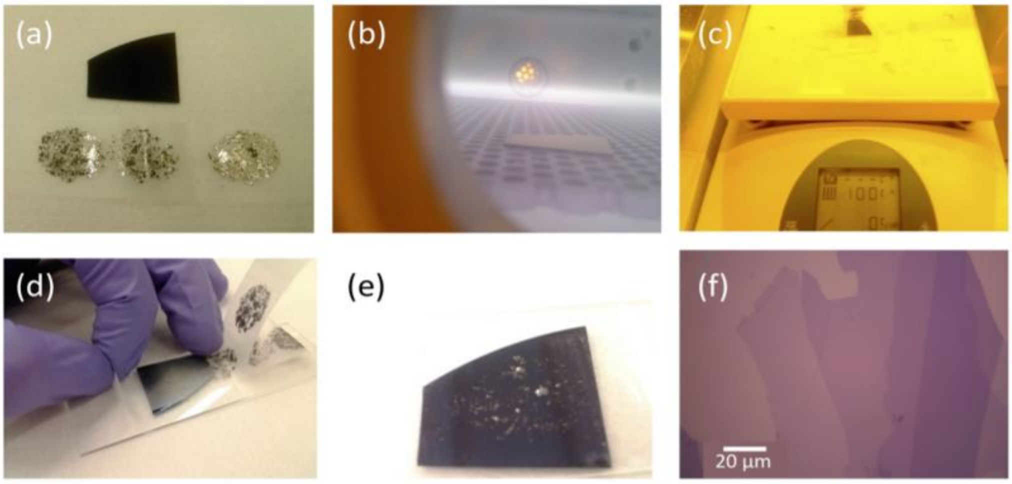

Figure 1. The sketch of the improved exfoliation process of layered crystals (graphene) [15]. (a) Image of SiO_2/Si substrate and tape with graphite sheet. (b) SiO_2/Si substrate with oxygen plasma cleaning. (c) Contact between the substrate surface and the tape with graphite, heating the tape on the hot plate in air at 100°C for 2 minutes. (d) The step for removing the substrate from the hot plate and strip the tape. (e) Image of substrate after stripping the graphene. (f) Optical micrograph of graphene flakes in (e). The thickness of the flake varies gradually from 1 to 4 layers.

3.1.2. Liquid phase exfoliation

Liquid phase exfoliation is also an important method for fabricating two-dimensional materials. One of them is liquid phase ultrasound-assisted exfoliation, which was proposed by Coleman in 2011 and had been improved and developed for many times [16, 17]. The specific method is to put two-dimensional material blocks into the exfoliation solution, and use ultrasound and centrifuge to exfoliate the material, thus obtain two-dimensional materials in lower layers. Although the cost of this method is cheap and two-dimensional materials can be prepared on a large scale, the number of sample layers obtained is difficult to control as the mechanical exfoliation. Therefore, it cannot be widely used in precision electronic equipment at present.

Another liquid phase exfoliation method that can fabricate two-dimensional materials is lithium ion embedding assisted exfoliation method [16]. A small volume of lithium ions is embedded into two-dimensional material blocks by electrochemical method to weaken the interlayer interaction force, and then the sample material with few layers can be easily obtained by external force. The disadvantage is that this method will affect the intrinsic properties of the material, which will reduce the performance of the device, i.e., requires to be further improved. However, it possesses the advantages including low production cost, high efficiency and large-scale preparation.

3.2. Bottom-up fabrication method

3.2.1. Hydrothermal/solution thermal synthesis method

Hydrothermal/solution thermal synthesis method is a way to obtain different types, appearances and crystallinities of sample materials by controlling the reaction parameters, e.g., pressure, temperature, time and precursor type in a closed space under high temperature and pressure. This method cannot accurately control parameters in industrial large-scale preparation and is susceptible to multiple factors, making it difficult to obtain excellent performance and quality materials [18].

3.2.2. Chemical vapor deposition

Chemical vapor deposition (CVD) is often used for the fabrication of transition metal dichalcogenide, and it is also one of the highly anticipated industrial large-scale preparation methods of two-dimensional materials with controllable size [19]. The principle is to sublimate the reaction materials at high temperature, and then use one or more carrier gases to chemically react in the air flow, and finally form deposits on the substrate surface. The preparation technique of this method is simple, extensible and reproducible, and has developed rapidly in recent years. To sum up, CVD gives consideration to both the quality and efficiency of the fabricating the low-dimensional materials. However, the application of CVD in the growth of wafer-level single crystal transition metal dichalcogenide still needs further researches.

4. Mainstream fabrication process of heterostructures

The way to construct two-dimensional material heterostructures accurately and efficiently is one of the difficulties and emphases in current work. Meanwhile, addressing this issue is also the basis of large-scale fabrication of two-dimensional heterostructures. Up to now, the construction of two-dimensional heterostructures can be divided into two categories, one is the mechanical transfer method which directly splices two-dimensional materials through mechanical exfoliation in physical means, and the other is the application of induced epitaxial deposition.

4.1. Mechanical transfer method

Mechanical transfer is the simplest method to construct two-dimensional heterojunction with high flexibility. Physical transfer technology is mainly used to transfer one or more two-dimensional materials to another substrate with two-dimensional materials to realize stacking of different materials and construct two-dimensional heterojunctions [19]. The heterojunction constructed by mechanical exfoliation transfer method has high quality and superior performance. However, on account of the low efficiency, limited size and sophisticated steps, it is difficult to achieve large-scale industrial fabrication. Besides, it is commonly used in the laboratory as an important step to explore physical properties.

4.2. Induced epitaxial growth method

Mechanical exfoliation transfer is relatively simple but difficult to construct large area heterojunction due to its low efficiency, hence it is more suitable for laboratory discovery. Therefore, explore a method for massive fabrication of heterostructures is a crucial task. Currently, researchers have found that low-dimensional heterojunctions can be directly generated through chemical synthesis [20, 21]. The mechanism is illustrated in Fig. 2. Compared with mechanical transfer method, it is more efficient and can achieve large area heterojunction growth, which is the most likely to form an industrial fabrication method. Nevertheless, the control of parameters is highly required, which is greatly affected by numerous external parameters, i.e., further research is demanded in process exploration.

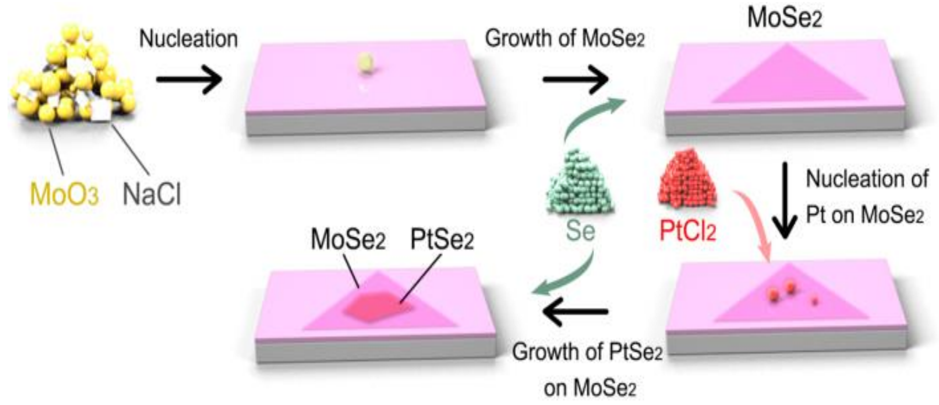

Figure 2. The growth mechanism of PtSe2/MoSe2 heterostructures [20].

5. Two dimensional heterostructure devices

With the deeper development of the related research, the two-dimensional heterostructure devices have attracted a large number of scholars because of their outstanding performances. Recently, heterostructure devices are mainly fell into three categories, namely electronic devices, optoelectronic devices and spintronic devices.

5.1. Electronic Devices

Electronic devices are not only one of the indispensable components of integrated circuits, but also cornerstone of the development of information age. Integrated circuits use semiconductor manufacturing process, on a small piece of single crystal silicon to make many transistors, resistors, capacitors and other components. The channel thickness of traditional silicon-based Field effect transistor (FET) must be less than one third of the channel length in order to avoid short channel effect, and the channel process of traditional transistor is approaching the theoretical limit [22, 23]. Two-dimensional semiconductor materials, as the thinnest state-of-art semiconductor materials, can relatively pass smoothly because charge on its smooth surface is not affected by surface states and other factors. Moreover, the semiconductor properties and band gap of the materials itself make the switching ratio and mobility of low-dimensional material devices very superior. In addition, the heterojunction constructed by a variety of low-dimensional materials has been proved to have better performance than a single low-dimensional material. To be specific, the charge-carrier injection of asymmetric heterojunction electronic devices constructed by Cheng et al. can switch between tunnelling and thermal activation under negative and positive bias respectively, and the current switching ratio can reach 6×10^8 with rectification ratio surpassing 10^8 [24].

5.2. Optoelectronic devices

Optoelectronic devices refer to the devices that adopt photoelectric effect to realize the optoelectronic signal conversion or storage. Common optoelectronic devices contain LED, photosensitive resistance, etc. Compared with the optoelectronic devices constructed by a single low-dimensional semiconductor material, the optoelectronic devices constructed by low-dimensional heterostructure have unique electron transport performance and optoelectronic characteristics [23]. When two semiconductor nanomaterials with different work functions contact to form a heterojunction, the different Fermi levels will lead to the redistribution of carriers in the heterojunction, resulting in many novel physical properties. Scholars have made unremitting investigations on two-dimensional heterojunction optoelectronic devices and obtained various rich achievements. For example, the CsPbBr3/graphene heterojunction optoelectronic device established by Jin et al. has realized the integration of the persistent response of positive (ultraviolet light) and negative (visible light) photoconductive materials in the same device [25].

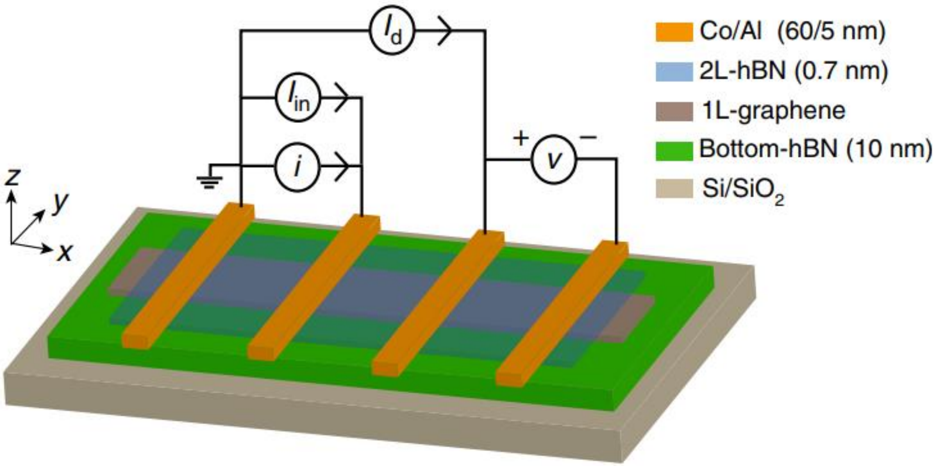

Figure 3. Illustration of spin device with ferromagnetic/hBN/graphene/hBN heterostructure [29]

5.3. Spintronic devices

Spin is the inherit feature of particle. As for electrons, in addition to charge, the spin angular momentum is $\pm h/4\pi$. With the participation of the external magnetic field, the electron is not only affected by Lorentz force, but also coupling between the internal magnetic moment and the external field. By introducing spin properties into semiconductor devices and using electron charge and spin as information carriers, a new generation of devices, called electronic spin devices, is developed. For example, graphene, a two-dimensional material, can conduct the spin with electrons effectively, and has a high application prospect in spintronic devices [26-28]. The ferromagnet/bilayer-HBN/graphene/hBN heteromechanism established by Gorram et al. has a spin-injection polarization as high as 50% (135%) at a positive bias of +0.6V and a reverse polarization as high as -70% (-60%) at a reverse bias of -0.4V [29]. The typical structure is depicted in Fig. 3.

6. Limitations and prospects

In the process of preparing two-dimensional materials, the mechanical exfoliation method can be used to obtain a few layers or a single layer of low-dimensional materials for the fabrication of various devices. Nevertheless, the thickness of the low-dimensional materials obtained by this method is different, and this process is accidental and uncontrollable. Samples obtained by the same or similar exfoliation techniques are not identical. In addition, this method is slow, complicated and inefficient. Therefore, chemical synthesis is the best solution for two-dimensional material fabrication. However, large-scale industrialized preparation of two-dimensional materials is not mature. For example, the diameter of tubular furnace chamber has not found a universal solution, and the quality of the synthesized low-dimensional nanomaterials is poor compared with mechanical exfoliation. Therefore, one should propose novel approaches to obtain high quality samples while giving consideration to the synthesis efficiency.

Nevertheless, as for the construction of low-dimensional heterojunction, it is difficult to obtain both high quality and large area heterostructures by mechanical transfer method. Meanwhile, the heterostructures prepared by chemical induction generation method have the contamination problem caused by impurity residues. Owing to the low efficiency of mechanical transfer method and other problems, it is still necessary to explore a method to guarantee the quality of large-scale synthesis on the basis of large-scale construction of heterostructures in the future.

As a matter of fact, there is still a long way to fumble and explore for the devices built by heterostructures. At present, it is mainly limited by processing equipment and technology (e.g., the introduction of a variety of residual impurities during the lithography), resulting in a negative impact on device performance. Further developments of processing technology for low-dimensional devices and inspection equipment for checking devices are urgent issues [30].

7. Conclusion

In summary, this paper mainly introduces the components and applications of two-dimensional material and heterostructures. Specifically, a variety of main fabrication methods are listed starting from the preparation of low-dimensional nanomaterials, followed by narrating the mainstream way to build low-dimensional heterostructures, and finally introduced the classification of low-dimensional heterojunction devices. Two-dimensional heterojunction devices have brought new vitality and power to the development of devices in the post Moore era. However, it is still necessary to make breakthroughs in all aspects, e.g., the measure to strengthen the manufacturing technology and level of low-dimensional devices, the path to achieve large-scale two-dimensional materials fabrication and construction of heterostructures. Overall, these results summarize the development of the state-of-art two-dimensional materials and devices, which offer clear insights for scientific discovery and technological breakthrough in certain field.

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