

# The preparation and the photoelectric characteristics of graphene/MoSe<sub>2</sub> heterojunction

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**Abstract.** The paper presents the preparation and optoelectronics characteristics of graphene/molybdenum selenide (MoSe<sub>2</sub>) heterojunction. MoSe<sub>2</sub> films were deposited on Si substrates using MoSe<sub>2</sub> powder as a raw material by a chemical vapour deposition (CVD) method, and then the graphene/MoSe<sub>2</sub> heterojunction was formed by growing a graphene layer on the MoSe<sub>2</sub> film using methane (CH<sub>4</sub>) as a raw material. The prepared MoSe<sub>2</sub> films consisted of many nanowires about 2 nm in diameter and 7 nm in length perpendicular to the surface observed by an Atomic force microscopy (AFM), while many small graphene pieces were formed and dispersed on MoSe<sub>2</sub> film surface. Additionally, we found that the growth of MoSe<sub>2</sub> film has a strong orientation growth in the (400) crystal plane, which is consistent with the MoSe<sub>2</sub> film composed of many parallel nanowires in the AFM picture. Moreover, we found that the graphene/MoSe<sub>2</sub> heterojunction has good absorption properties for visible light and has a significant photocurrent generation under illumination, indicating that the graphene/MoSe<sub>2</sub> heterojunction has excellent optical and electrical properties and has great potential for application in the field of optoelectronic devices.

## 1. Introduction

Graphene is a two-dimensional (2D) material formed by a single layer of carbon atoms with high electron mobility, light transmission, and high mechanical strength [1-4]. However, graphene is a zero bandgap semiconductor material, limiting its application in amplifiers and logic devices. Transition metal sulfide (TMDs) materials also have a layered structure. The layers are bonded with van der Waals forces and are easily stripped into single or multiple layers of two-dimensional materials. The single-layer TMDs are direct band gap materials and have excellent optical and electrical properties such as softness, transparency, high electron transmission speed, and high photoelectric conversion efficiency [5, 6]. It can be used to prepare flexible, transparent, high photoelectric conversion efficiency of optoelectronic devices.

Monolayered molybdenum selenide (MoSe<sub>2</sub>) is a direct band gap semiconductor with a band gap just between the wide bandgap semiconductor and the zero band gap of graphene, which can be used to fabricate high efficiency optoelectronic devices [7]. Studies have shown that a single layer of MoSe<sub>2</sub> has a strong photoluminescence effect at 1.55 eV, but the luminescence effect decreases and the luminescence peak shifts toward the long-wavelength with the number of layers increasing [8]. Compared with graphene photodetectors [9-11], the open-circuit voltage and response rate of a single-layer MoSe<sub>2</sub> photodetector [12] is even higher, and it has strong anti-fatigue properties and high stability with fast photoelectric response characteristics [13]. In addition, other TMDs two-dimensional materials such as MoSe<sub>2</sub> and graphene or MoS<sub>2</sub> are easily stacked vertically or laterally to form Van der Waals heterojunctions [14, 15]. Such heterojunctions have strong optical absorption, high photocurrent gain, wide response range, and ultra-fast (nanosecond) properties



[16-20]. It has caused people's research interest. Heterojunction transistors and solar cells prepared from 2D materials such as graphene and molybdenum sulfide have been widely reported, and relatively few studies have been conducted on MoSe<sub>2</sub>-based heterojunctions. In this work, graphene/MoSe<sub>2</sub> heterojunctions were prepared by chemical vapor deposition and the optoelectronic properties of the heterojunctions were studied. The related mechanism was discussed. The research in this area has a good guiding significance for the preparation of detectors, solar cells and other optoelectronic devices.

## 2. Experiment

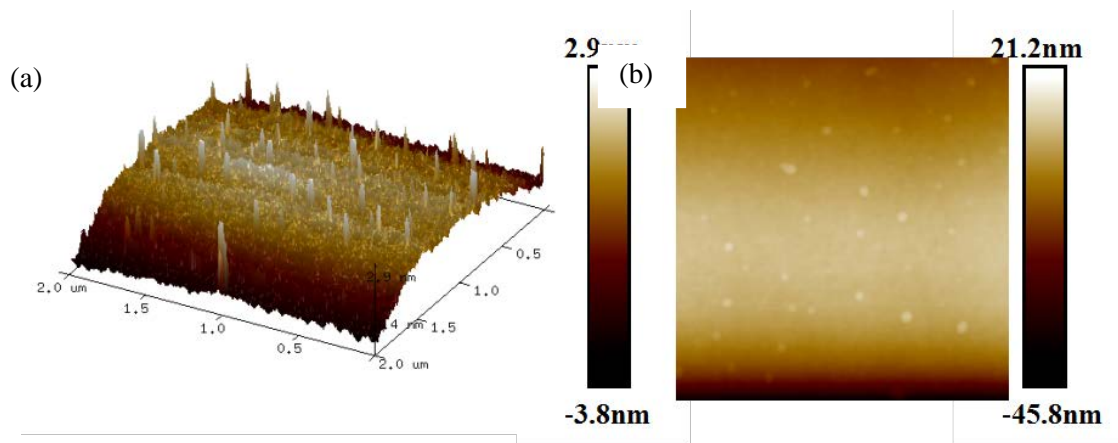
MoSe<sub>2</sub> and graphene films were deposited on p-Si (100) using a chemical vapor deposition method to form the heterojunction. The experimental system consists of five parts: temperature-controlled heating device, vacuum pumping system, air intake system, gas flow meter and water bath box. Before the experiment, the silicon substrate was immersed in dilute hydrofluoric acid to remove surface silica. Then ultrasonic cleaning with acetone, ethanol, deionized water followed by 10 minutes, and finally dried and arranged in a quartz tube. The Erlenmeyer flask containing the MoSe<sub>2</sub> solution was placed in a water bath and the water bath was kept at a constant temperature of 70°C. The reaction device (quartz tube) was pumped to a vacuum of about 10<sup>-2</sup> Pa. Having optimized the experimental conditions, we found that the ideal growth conditions for MoSe<sub>2</sub> film the reaction temperature was 700°C and the reaction time was 30 mins. When the reaction temperature was reached, Ar gas was introduced. MoSe<sub>2</sub> saturated vapor molecules were carried by Ar gas into the quartz tube. Nano-sized MoSe<sub>2</sub> thin film was formed by the MoSe<sub>2</sub> molecules adsorb and deposition on the substrate. After the samples were cooled, a few of the MoSe<sub>2</sub> samples were taken out to perform surface morphology and photoelectric characteristics test analysis.

The graphene thin film was subsequently formed on the MoSe<sub>2</sub> thin film to form a heterojunction. A few MoSe<sub>2</sub> film samples were still placed in a quartz tube, and the quartz tube is evacuated and heated. When the quartz tube is heated to 850°C, CH<sub>4</sub> and argon were introduced, and the ratio of methane to argon is 1:25. CH<sub>4</sub> was decomposed at high temperatures and carbon atoms are deposited on top of MoSe<sub>2</sub> to form graphene films. After 10 minutes of reaction, the experiment was finished.

The surface morphology and crystal structure of graphene and MoSe<sub>2</sub> thin film samples were observed by atomic force microscopy (AFM), X-ray diffractometry (XRD) and Raman spectroscopy. The electrical properties of the sample, such as film surface conductivity, electron mobility and photocurrent of graphene/MoSe<sub>2</sub> heterojunction were studied using the HMS-3000 Hall Effect Meter and I-V measurement. Finally, the light absorption characteristics of the samples were analyzed by UV-3600 spectrophotometer.

## 3. Results and Analysis

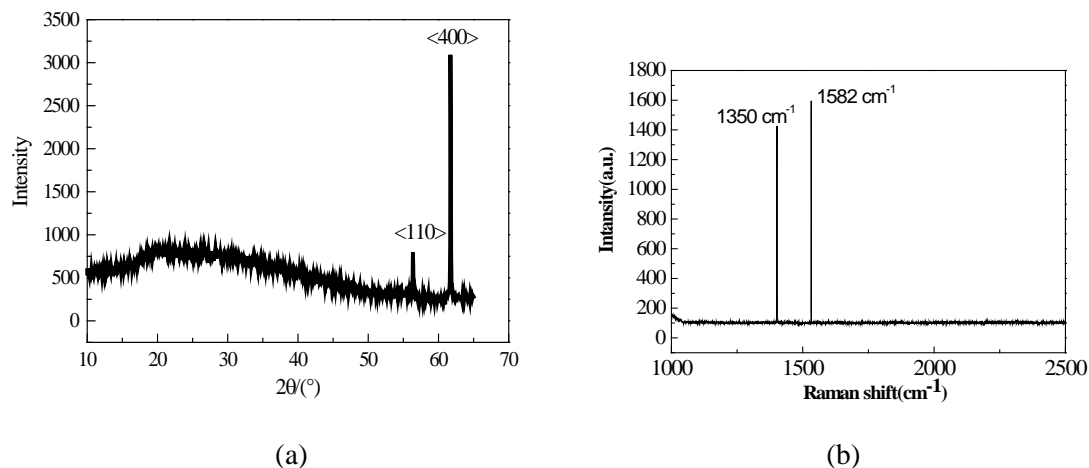
The surface morphology of MoSe<sub>2</sub> was observed by AFM. The two-dimensional morphology was shown in Figure 1(a). From Figure 1(a), it can be found that many vertical nanowires are distributed on the surface of MoSe<sub>2</sub> films grown by chemical vapor deposition. The nanowires have a diameter of about 2 nm, a length of about 7 nm, and a length to diameter ratio of about 3.5:1. The growth pattern of MoSe<sub>2</sub> showed a significant columnar growth [21]. Figure 1(b) shows the AFM morphology of the graphene film. The surface of the graphene film is very even and uniform. Many graphene sheets are distributed on the surface, indicating that the graphene was grown as an island with an average thickness of about 15 nm.



**Figure 1.** (a) The AFM picture of MoSe<sub>2</sub>. (b) The image of the graphene film.

The crystal structure of the prepared MoSe<sub>2</sub> thin film was characterized by X-ray diffraction. The results are shown in Figure 2(a). It can be seen that there are two obvious diffraction peaks at 57° and 62° from the MoSe<sub>2</sub> thin film, corresponding to the (110) and (400) crystal planes of MoSe<sub>2</sub>. The strength of the (400) crystal plane is much greater than that of the (110) crystal plane, indicating that MoSe<sub>2</sub> has a preferential growth orientation in the (400) crystal plane direction. In addition, these two diffraction peaks are linear, with a very narrow half-width, indicating that the MoSe<sub>2</sub> film is mainly crystalline. This is consistent with the uniform columnar growth of MoSe<sub>2</sub> observed in Figure 1.

The Raman spectrometer was used to characterize the structure of graphene. The results are shown in Figure 2(b). It can be seen that there are two strong Raman scattering vibration peaks at 1350 cm<sup>-1</sup> and 1582 cm<sup>-1</sup>, corresponding to the D and G peaks of graphene, respectively. The G peak is relatively strong, indicating that the grown graphene is multilayer graphene. Both the D peak and the G peak show a very narrow line shape, which indicates that the graphene growth film has good quality and less surface impurities or defects. The graphene 2D peak is located near 2700 cm<sup>-1</sup>. Compared with the G peak, the graphene has a weaker intensity, but the peak shape is sharp, indicating that the graphene layers are stacked neatly and the interlayer-layer interaction force is strong.

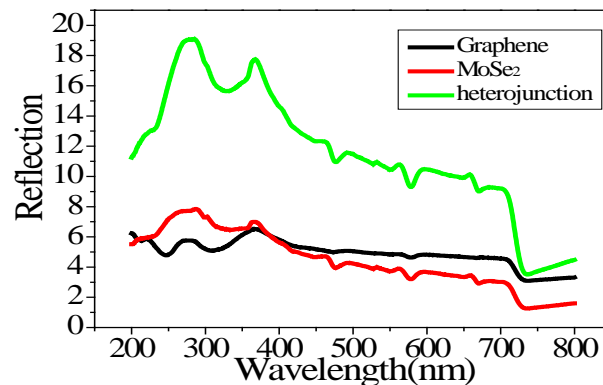


**Figure 2** (a) The XRD pattern of the MoSe<sub>2</sub> thin film. (b) The Raman spectrum of the graphene film.

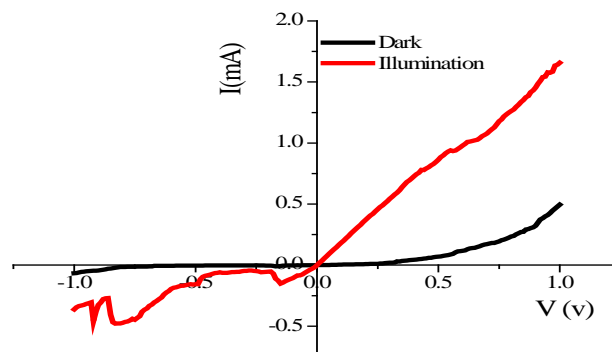
The reflection characteristics of graphene, MoSe<sub>2</sub> films and the heterojunctions were analyzed using UV-3600, as shown in Figure 3. It can be seen that the graphene films have the minimum reflections at 245, 310, and 725 nm; while MoSe<sub>2</sub> films have the minimum reflections at 330, 445, 574, 665, and 730 nm. The graphene/MoSe<sub>2</sub> heterojunction reflectance spectrum is similar to that of MoSe<sub>2</sub>,

with the minimum located at 315, 410, 570, 660, and 730 nm. In addition, the reflectivity of the heterojunction is significantly higher than that of the single graphene and MoSe<sub>2</sub> films, indicating that as the thickness of the deposited film increases, the reflectance of the heterojunction increases gradually and the light absorption decreases. Although the light absorption of the graphene/MoSe<sub>2</sub> heterojunction is lower than that of a single film, in general there is a strong absorption in the visible region, which can produce a significant photovoltaic effect. It can be used to make high-efficiency photovoltaic devices such as solar cells and photodetectors.

The I-V behavior of the heterojunction with and without illumination were performed by performing an I-V measurement on the sample, as shown in Figure 4. It can be seen that the heterojunction has a good I-V characteristics. When the light is irradiated, the current of the sample is significantly enhanced. It is shown that graphene/MoSe<sub>2</sub> has a remarkable photovoltaic effect under illumination. The open circuit voltage is 0.029V, and the short circuit current is  $1.41 \times 10^{-3}$  mA. The significant open-circuit voltage and short-circuit current indicate that its photovoltaic effect is significant, and can produce highly efficient solar cells and other devices.



**Figure 3.** The reflection spectrum of the graphene, MoSe<sub>2</sub> and the graphene/MoSe<sub>2</sub> heterojunction films



**Figure 4.** The I-V characteristic curves of the Graphene/MoSe<sub>2</sub> heterojunction

#### 4. Conclusions

High-quality MoSe<sub>2</sub> thin film was prepared on a p-Si substrate by CVD using MoSe<sub>2</sub> powder as a raw material. Afterwards, graphene was deposited on the MoSe<sub>2</sub> thin film to form a heterojunction using CH<sub>4</sub> as a raw material. The surface morphology, crystal structure, photoelectric properties of graphene films and MoSe<sub>2</sub> films, and the photoelectric properties of graphene/MoSe<sub>2</sub> heterojunctions were studied. The graphene/MoSe<sub>2</sub> heterojunction has a low surface reflectance, good light absorption

capability, significant photovoltaic effect, and has a good application prospect in the preparation of high-efficiency solar cells.

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