



Full Length Article

Enhanced photoresponse characteristics of transistors using CVD-grown MoS₂/WS₂ heterostructures

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ABSTRACT

Semiconductor heterostructures based on transition metal dichalcogenides provide a broad platform to research two-dimensional nanomaterials and design atomically thin devices for fundamental and applied interests. The MoS₂/WS₂ heterostructure was prepared on SiO₂/Si substrate by chemical vapor deposition (CVD) in our research. And the optical properties of the heterostructure was characterized by Raman and photoluminescence (PL) spectroscopy. The similar 2 orders of magnitude decrease of PL intensity in MoS₂/WS₂ heterostructures was tested, which is attribute to the electrical and optical modulation effects are connected with the interfacial charge transfer between MoS₂ and WS₂ films. Using MoS₂/WS₂ heterostructure as channel material of the phototransistor, we demonstrated over 50 folds enhanced photoresponsivity of multilayer MoS₂ field-effect transistor. The results indicate that the MoS₂/WS₂ films can be a promising heterostructure material to enhance the photoresponse characteristics of MoS₂-based phototransistors.

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1. Introduction

In recent years, two-dimensional (2D) semiconducting transition metal dichalcogenides (TMDs) materials, including MX₂ (M = Mo, W; X = S, Se), have attracted much attention owing to their unique structure as well as remarkable physical and chemical properties [1–4]. As a member of the TMD family, in contrast to graphene with no band gap, MoS₂ is a semiconductor which has the controllable band gap from 1.8 eV (direct-gap) to 1.2 eV (indirect-gap) depending on the number of layers [5–7]. Due to its semiconducting properties, MoS₂ has been used in electronic devices and circuits based on field effect transistors (FETs) [5,8–13]. MoS₂ has also been considered a promising candidate for emerging optoelectronic devices [3,14–19]. Compared to bulk MoS₂ materials which was widely concerned as a high-grade solid lubricant and catalyst, layered MoS₂ films have potential applications in many fields such as optics, electrics, electronics. The layered MoS₂ films can also be used in manufacture of high responsivity photodetectors, led, photodiodes, solar cells, electroluminescent devices, etc [20]. Similar to MoS₂, other TMDs films

are all layered semiconductor materials with the definite band-gap. Recently, the chemical vapor deposition (CVD) method has been utilized to synthesize MoS₂ films [5,21–24], however, the synthesis of large-scale, high-quality, single-layer MoS₂ films has always been a significant challenge for practical device development [5].

For decades, heterostructures consist of two different materials have become the basis for developing the new type functional materials [25]. However, the functionality of conventional semiconductor heterostructures is often limited by inefficient charge transfer across interfaces due to the interfacial imperfection caused by lattice mismatch [26]. Therefore, the choice of the constituent material is limited to the kind of materials those having the similar lattice structure. The recently developed 2D layered materials, such as graphene [27], boron nitride (BN) [28] and TMDs [29], have opened up a new path for the fabrication of Van Der Waals heterostructures. These Van Der Waals heterostructures exhibit excellent mechanical flexibility and their preparation method is consistent with the current thin-film technology. Up to present, a majority of researches for Van Der Waals heterojunction have been focused on graphene and multilayer TMDs materials. Vertical tunneling transistors based on multilayer MoS₂ or WS₂ as the tunneling barrier between graphene layers, acquired high switching

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ratios [30,31]. This heterostructure exhibits promising applications in optoelectronics due to the novel optical properties of TMDs material and exceptional transport performance of graphene can be concentrated together [32,33].

In contrast to semi-metallic semiconductor structures, the semiconductor heterostructure formed by different TMDs materials are similar to the conventional semiconductor heterostructure. When applied to the active layer of a photoelectric device, it can overcome the limitation of a single TMD layer. For instance, the light absorption of single TMD film based photovoltaic devices is relatively low. Although the monolayer TMDs material can absorb more than 10% of incident light, such high absorption coefficients cannot be scaled up by increasing thickness of the films because of the band gap changes from direct to indirect. Nevertheless, the monolayer TMD film in the heterostructure maintains its own direct band gap owing to the weak interlayer coupling. Therefore, the absorption capacity can be enhanced simply by stacking layers. Compared to the monolayer TMD films, the multilayer TMD materials has not been extensively researched for application in devices. However, in previous studies, Li et al. reported the observation of thickness reduction induced crossover of electrical contact at Au/MoS₂ interfaces. [34] They found that the interfacial potential barrier can be finely tailored from 0.3 to 0.6 eV by merely varying the thickness of MoS₂. The contact resistivity of devices slightly decreases with the reducing of MoS₂ thickness, which mainly governed by the quantum confinement effect. [34] In addition, the density of states of multilayer MoS₂ is three times that of monolayer MoS₂, which will lead to considerably high drive currents in the ballistic limit. And the multiple conducting channels can be created by field effect in multilayer MoS₂ for boosting the current drive of FETs [35]. Moreover, the Hall mobilities increased with density, reaching 375 cm²/(V·s) for the bilayer, in contrast to 250 cm²/(V·s) for the monolayer [36]. The multilayer TMD films also have the advantages such as more stable structure and easier to prepare than the monolayer TMD films. Therefore, in our researches, we not only prepared the large-area multilayer MoS₂/WS₂ heterostructures, but also studied optical properties of the MoS₂/WS₂ heterostructure. Then we fabricated the multilayer MoS₂/WS₂ heterostructures based FET and shown it offers a compelling case for applications in enhancing the photoresponse characteristics of TMD based transistors.

2. Experimental details

We synthesized the MoS₂ films using a chemical vapor deposition system (CVD, KTL 1700-1400). The reaction process is mainly based on the instability of MoO₃ at 650 °C, which can be easily volatilized into the gaseous state. Then a thin layer of MoS₂ film is deposited on the substrate after the reaction of gaseous MoO₃ with sulfur vapor. The detailed preparation process is that, firstly, the SiO₂ (270 nm)/Si substrates of 2 cm × 2 cm in size were prepared and cleaned in trichloroethylene, acetone, ethanol, and deionized (DI) water for 10 min each. Then, we put 0.05 g MoO₃ powder (99.5%, Aldrich) and the substrate on two quartz boats and inserted them into the high temperature zone of furnace. Additionally, the sulfur powder (99.98%, Aldrich) was put on another quartz boat and placed at the low temperature zone of furnace. The two temperature zones of furnace were heated to 650 °C and 160 °C, respectively. This temperature was continued for 60 min in an atmosphere of Ar (20 sccm) in the furnace. Then, the furnace was allowed to cool naturally in the Ar atmosphere. Adopt this method, MoS₂ films were synthesized on the SiO₂/Si substrate. The WS₂ and MoS₂/WS₂ films were prepared by the same way. The schematic illustration of the CVD-grown MoS₂, WS₂ and MoS₂/WS₂ system was shown in Fig. 1a and b. The MoO₃ and

WO₃ powder were used to provide the source of molybdenum and tungsten. And the temperature programming process of XO₃ (X = Mo, W) and S precursors was shown in Fig. 1c.

In order to grow large size, uniform and continuous triangular MoS₂/WS₂ film, considering the sublimation temperature of MoO₃ and WO₃ are different, the control variable method in the matched group experiments of CVD-grown MoS₂/WS₂ films was used to research the influence of different reaction conditions on the morphology of the MoS₂/WS₂ films. The experimental conditions of the matched group was shown in Table 1. The effect of different reaction temperature on the experimental results was studied in the group 1–3 with the amount of reactant was unchanged. And the influence of the amount of reactants on the experimental results was discussed in the group 4–8 with the reaction temperature was unchanged. During the whole experiments of the matched group, these conditions consist of the amount of S powder (0.2 g), the heating time (1 h) and the flow rate of shielding gas (20 sccm) were all invariant.

3. Results and discussion

3.1. Morphological analysis

The morphology of CVD-grown TMD (MoS₂ and WS₂) films were analyzed by the Scanning electron microscopy (SEM, JSM-6010LA). The results show that the morphology of CVD-grown TMD films are both triangular. The size of the triangles is 25 μm (MoS₂ film) and 10 μm (WS₂ film), respectively, as shown in Fig. 2a and f. In particular, the hexagonal morphology of TMD films was observed at the edge of the substrate, as shown in Fig. 2b. In order to research the morphological evolution of the CVD-grown TMD materials, the morphology of MoS₂ films with different reaction time (20 min, 40 min and 60 min) was compared, as shown in Fig. 2c, d, e. We discovered that the morphology of the regular hexagonal MoS₂ gradually became irregular and finally grew into the triangular shape with the increased of the reaction time. The schematic diagram of shape evolution for the CVD-grown MoS₂ films was shown in Fig. 2g [1].

The effects of different conditions on the morphology of the MoS₂/WS₂ film were investigated by the matched group. Fig. 3a–h correspond to group 1–8 of the contrast tests shown in Table 1. The obvious triangular structure can be observed with the reaction temperature of 750 °C and 800 °C, as shown in Fig. 3a, b. And the evident large-size continuous triangle was discovered with the temperature of 800 °C. The morphology of the sample would be irregular as the temperature continues to rise, as shown in Fig. 3c. We can also observe that the size of the triangle decreased along with the amount of reactants increased from Fig. 3d–h. The pattern of triangle disappeared with the amount of oxide exceed 0.1 g (Fig. 3f). And the morphology of the sample was completely irregular with the amount of oxide increased to 0.15 g. Thus, we concluded that the best reaction conditions of CVD-grown high-quality and triangular-shaped MoS₂/WS₂ films were the reaction temperature of 800 °C with the dosage of MoO₃ and WO₃ were both 0.05 g.

Subsequently, we discovered the different morphologies of different regions in the same MoS₂/WS₂ sample. The sample prepared with the conditions of matched group 4 was separated into two regions, as shown in Fig. 4a and b. The region A shown the large area continuity overlapping triangular type of the MoS₂/WS₂ heterostructure, as shown in Fig. 4c and d, the sizes of the two triangular films were 10 μm (WS₂) and 50 μm (MoS₂), respectively. The morphology of samples in region B was tiled quadrilateral nanosheets, the size of the nanosheets was 15 μm–4 μm, as shown in Fig. 4e. Owing to the samples of region B were really thick, the

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