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Electrical properties of MoS_2/graphene heterostructure and pn junction diode

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Abstract

The two-dimensional nano-material heterojunction combination has recently become an important trend in component development. The graphene Fermi level can be precisely controlled using the oxygen adsorption. Therefore, graphene can be tuned from zero-gap to p-type semiconductor material using the amount of adsorbed oxygen. In addition, Molybdenum disulfide (MoS_2) films are currently the most potential semiconductor materials in two-dimensional (2D) transition metal dichalcogenides. In this study, we combine MoS_2 and graphene to produce a heterojunction with their unique properties and exhaustively study the interface and electrical properties of the device.

1. Introduction

In the past few years, two-dimensional (2D) transition metal dichalcogenides (TMDs) have shown intriguing physical properties and exciting prospects for a variety of applications. Molybdenum disulfide (MoS_2), a naturally occurring molybdenite, is one of the most stable layered metal dichalcogenides [1]. The band-gap could be controlled with different MoS_2 film thicknesses [2]. Due to the strong anisotropy of the layered structure, these materials have a specific physical and chemical 2D behavior. For example, crystals can be easily peeled along sheets (van der Waals planes), which make it possible to form a clean surface owing to the internal structure of crystals and has attracted interest for diverse applications.

Graphene, a monolayer of sp^2-hybridized carbon atoms arranged in hexagonal form which shows outstanding properties, such as high electrical conductivity, high carrier mobility, and optical transparency, resulting in being widely used in many research fields, including gas sensors, field emission displays, and graphene diodes [3]. In terms of material properties, oxygen functionalization is known to tune the band-gap of graphene. A gradual reduction in the graphene oxygen concentration also tailors the material band-gap to a different route [4].

In this work, we control the thickness of the few-layer MoS_2 films with different exfoliation times. We present Raman scattering studies of few-layer MoS_2 samples and find that these modes exhibit well-defined thickness dependence with two vibration modes shifting away from each other in frequency with increasing thickness [5]. This provides a convenient and precise diagnostic for determining the layer thickness of samples. Furthermore, we demonstrate semiconductor integration in the form of few-layer MoS_2 with graphene as a pn junction diode and measure the device current and voltage characteristics.

2. Experimental Procedure

Single MoS_2 crystal was grown by the chemical vapor transport (CVT) method, using Br_2 as a transport agent. Prior to crystal growth at quartz ampoule (20 cm length×22 mm outside diameter×17 mm inside diameter), containing Br_2 and the elements (Mo and S) was cooled with liquid nitrogen, evacuated to 10^{-6} torr and sealed. The ampoule was placed into a three-zone furnace with the charged pre-reacted for 24 h at 800°C with the growth zone at 950°C, to prevent product transport. After 240 h, the ampoule was then removed with wet tissues applied rapidly to the end away from the crystals to condense the Br_2 vapor. When the ampoule reached room temperature, it was opened and the crystals removed. Few-layer MoS_2 films were stripped from bulk MoS_2 single crystal using mechanical exfoliation method (Fig 1(a)). MoS_2 films were then transferred onto a stainless substrate to define the cathode and n-type material. In Fig 1(b) the real image of the few-layer MoS_2 films exhibited a square with a size of 2×3 mm^2.

After that, graphene was grown using thermal chemical vapor deposition (CVD). Cu foil was annealed at 1000°C for 60 min with Ar (200 sccm) and H_2 (4.5 sccm) mixture. CH_4 was then introduced into the quartz tube at a flow rate of 10 sccm for 10 min after annealing. Then, Cu foil was removed by a conventional chemical etching technique using $FeCl_3$ solution. Once etching was completed the graphene sheet was floated on top of the solution and could be transferred onto a few-layer MoS_2 films/cathode (the stainless electrode). We covered the stainless electrode under the graphene to define the anode as shown in Fig. 2. We adopted MoS_2 with different thicknesses as the pn junction of the n-type-side. Rectifying effects with a high on/off ratio were demonstrated in our proposed pn junction. The MoS_2 built-in potential barrier in the depletion region can be controlled by changing the layer thickness of MoS_2.

In this study, we fixed the thickness of the p-type material (graphene) with different thicknesses of n-type material (few-layer MoS_2 films) to fabricate the pn interface. In order to investigate the MoS_2 micro-structure, Raman spectroscopy was performed with Ar laser excitation at a wavelength of 514.5 nm. The heterojunction diode I-V characteristics were measured using a source meter (Keithley 2410) in ambient atmosphere at room temperature.

3. Results and Discussion

Few-layer MoS_2 film was exfoliated from MoS_2 bulk and transferred onto the cathode substrate. The Raman spectra of few-layer MoS_2 films show in Fig. 3(a). Two Raman modes, E^1_{2g} and A_{1g}, exhibited sensitive thickness dependence. The difference of Raman shift was decreased with the decreasing of MoS_2 thickness. The thickness of MoS_2 film was controlled with the different exfoliation times. The decreasing thickness of MoS_2 film could improve the built-in potential barrier (V_{bi}) and depletion region. In Fig. 3(b), the V_{bi} were shifted toward positive voltages from 0.8 to 3.1 V as the thickness of MoS_2 films decreased.

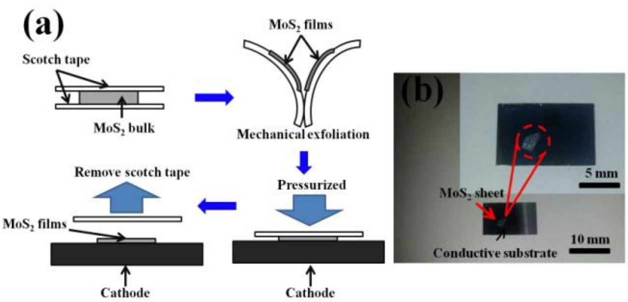

Fig. 1 (a) The appearance of the device including four stages of the fabrication procedure. (b) The real image of the sample.

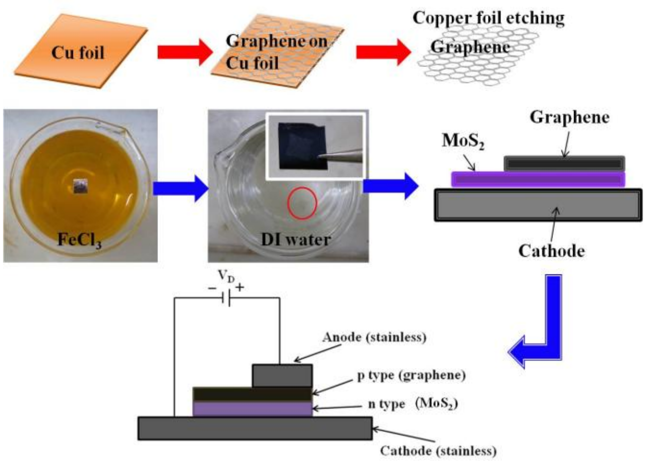

Fig. 2 The schematic diagram of the fabricated processfor pn junction diodes.

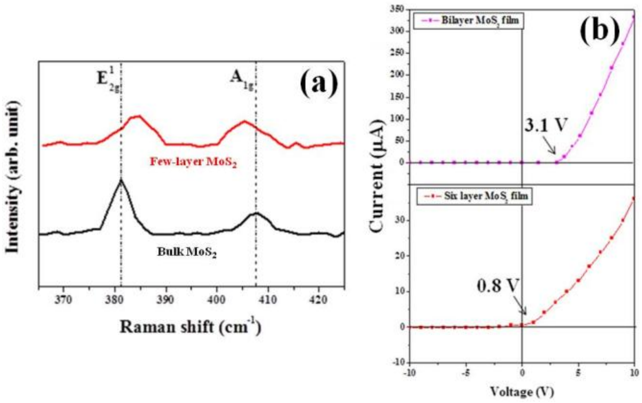

Fig. 3 (a) The Raman spectra of the few-layer MoS_2 film and Bulk MoS_2. (b) Typical I-V curves of the pn junction diodes fabricated with different thicknesses of the MoS_2 films.

4. Conclusions

We demonstrated a facile method for exfoliating high-quality MoS_2 films directly from bulks MoS_2 single crystals. The graphene sheets were successfully synthesized on Cu foils and transferred to few-layer MoS_2 films/cathode to characterize the electrical properties of the novel hybrid graphene/MoS_2 heterojunction. Through decreased the thicknesses of MoS_2 films, the indirect band-gap turned from 1.3 to 1.5-1.8 eV. Under controlling thicknesses of MoS_2 in pn junction diodes, the built-in potential barrier in the depletion region could be adjusted.

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