

Contents lists available at ScienceDirect

Physics Letters A





Tunable photoresponse with small drain voltage in few-layer graphene–WSe₂ heterostructures



Wei Luo^{a,b}, Shiqiao Qin^{a,c}, Mingsheng Long^b, Erfu Liu^b, Yajun Fu^b, Wei Zhou^b, Feng Miao^b, Sen Zhang^a, Renyan Zhang^{a,d}, Xue-Ao Zhang^{a,c,*}

^a College of Science, National University of Defense Technology, Changsha 410073, China

^b National Laboratory of Solid State Microstructures, School of Physics, Nanjing University, Nanjing 210093, China

^c State Key Laboratory of High Performance Computing, National University of Defense Technology, Changsha 410073, China

^d School of Physics and Astronomy, University of Manchester, Manchester M13 9PL, United Kingdom

ARTICLE INFO

Article history: Received 5 April 2016 Received in revised form 23 May 2016 Accepted 31 May 2016 Available online 2 June 2016 Communicated by R. Wu

Keywords: Graphene Transition-metal dichalcogenides Heterostructures Photoresponse Drain voltage

1. Introduction

Many two-dimensional (2D) materials with the formula MX_2 (M = Mo, W, Re, etc. and X = S, Se or Te) have proved to be excellent optoelectronic materials in previous researches [1–6]. Due to the doping effect in transition-metal dichalcogenides (TMDCs) with metal contacts [7–9] and the exotic electronic and optoelectronic properties of graphene [10,11], heterostructures based on TMDCs and graphene have been widely explored [12–14]. For example, few-layer WSe₂ transistor with graphene electrodes has shown ambipolar characteristics [14]. Thanks to the high valance band edge and small band gap [15,16], layered WSe₂ is a promising participator in photo electronic systems for its *p*-type properties. Recently, rectifying photovoltaic response was found in atomically thin *p*-*n* junctions within monolayer WSe₂ and its van der Waalsbonded hetero-interfaces [17–21], which demonstrate the potential of WSe₂ for novel optoelectronic applications.

As be proved, the photoresponsivity of heterostructures can be easily tuned by gate voltage, and that will be affected by drain voltage [22–24]. In this paper, we studied the photoresponse prop-

E-mail address: xazhang@nudt.edu.cn (X.-A. Zhang).

http://dx.doi.org/10.1016/j.physleta.2016.05.060 0375-9601/© 2016 Elsevier B.V. All rights reserved.

ABSTRACT

Two-dimensional layered heterostructures show great potential to develop optoelectronic systems. Here, we have investigated the photoresponse properties of two contact interfaces in few-layer graphene–WSe₂ heterostructures. The photoresponsivity of graphene–WSe₂ contact interface is about 2.67 mA/W, and the photoresponsivity of WSe₂-metal contact interface is about 0.2 mA/W. Photocurrent images show that the two contact interfaces behave differently under drain voltage from -0.5 V to 0.5 V. The photoresponsivity of one contact interface increases with the drain voltage, and that of the other decreases with the drain voltage. Experimental results and band diagram studies prove that the photoresponse properties of contact interfaces are tuned by small drain voltage. This study will be beneficial for understanding the effect of drain voltage on the heterostructures.

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erties of two contact interfaces in few-layer graphene–WSe₂ heterostructure, and obtained photocurrent images of the heterostructure under different drain voltage. Results show that the photoresponsivity of the contact interfaces could be tuned by small drain voltage.

2. Experiments

Device fabrication was started by using a polymer-free van der Waals adhesion technique to assemble layered WSe2 onto graphene flakes [25]. In detail, the WSe₂ and graphene flakes were mechanically exfoliated with scotch tape from bulk crystals of WSe₂ and Kish graphite, respectively. Then they are transferred onto SiO₂/Si (300 nm/0.5 mm) substrate with 95°C heating to improve flakes yield. Then a $\sim 1 \ \mu m$ thick layer of water-soluble polyvinyl acetate (PVA) on transparent elastomeric polydimethyl siloxane (PDMS) stamp was prepared. To pick up the WSe₂ flake, we attached the inverted PDMS stamp to a micromanipulator and aligned the PVA flake over a chosen exfoliated WSe₂ flake, brought the two flakes into contact, and then picked up the target WSe₂ flake via the van der Waals interaction. This pick-up process was done at a substrate temperature of 70°C. Finally, we transferred the WSe₂ on the PVA/PDMS stamp onto the graphene flake by melting the PVA at 90 °C, resulting in the van der Waals stack of WSe₂/graphene on the SiO₂/Si substrate. After dissolving the

^{*} Corresponding author at: State Key Laboratory of High Performance Computing, National University of Defense Technology, Changsha 410073, China.



Fig. 1. (a) Optical image of the fabricated device. The dark purple region is few-layer graphene, the blue area is few-layer WSe₂, insets show heights of the flakes. The scale bar is 10 μ m. (b) Raman spectra of the WSe₂ flake. (c) Raman spectra of the graphene flake, inset shows the FWHM of the 2D band of the graphene. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)



Fig. 2. (a) Back gate transfer characteristics of graphene–WSe₂ heterostructure. (b) Back gate transfer characteristics of WSe₂ transistor. (c) Back gate transfer characteristics of graphene transistor. The graphene–WSe₂ heterostructure and WSe₂ transistor are measured under $V_{ds} = 1$ V, the graphene FET are measured under $V_{ds} = 0.1$ V. Purple curves are obtained after device fabrication, green curves are extracted after anneal process, blue curves are obtained after air exposure. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

PVA in deionized water, we fabricated electrical contacts of Ag/Au (5 nm/45 nm) using conventional e-beam lithography and evaporation process.

In this paper, all the measurements were done in ambient atmosphere at room temperature. The thicknesses of the WSe₂ and graphene flakes were confirmed by atomic force microscopy (AFM, NT-MDT) and Raman spectra (Witec Alpha 300R). The electrical characterization was carried out using a Keithley 2636A digital Source-Meter. Optoelectronic properties of the heterostructures were measured in a Witec Alpha 300R with a 532 nm laser (spot size is about 700 nm through a \times 50 objective). Scanning photocurrent images are directly generated in the Witec Alpha 300R system with mapping of the heterostructures under focus laser illumination.

3. Results and discussion

Fig. 1(a) shows the optical image of the fabricated device. As shown in insets of Fig. 1(a), the thicknesses of the WSe₂ and graphene flakes determined by the AFM measurements are about 4.2 nm and 1.1 nm, respectively. The Raman spectrum from the WSe₂ and graphene flakes are presented in Figs. 1(b) and 1(c), respectively. The observed Raman-active modes of WSe₂ and graphene are consistent with the previously reported data [26,27]. It is proved that the materials are both few-layer flakes. The full

width at half maximum (FWHM) of 2D band of graphene shown in inset of Fig. 1(c) is about 57 cm⁻¹, indicating that the graphene is a three layer flake [26].

Next, we studied the electrical properties of the graphene-WSe₂ heterostructure. Results show that the graphene-WSe₂ heterostructure has *p*-type properties, which is different from that of fewlayer WSe₂ transistors with Ag contacts [7]. To elucidate this abnormal phenomenon, we studied the back gate transfer characteristics of the device under different conditions. Firstly, we measured its electrical properties after device fabrication. Then, we extracted its electrical properties after the device annealed in 300 °C Ar protected atmosphere for 1 h. Finally, we obtained its electrical properties after exposing the device in air for 12 h. Fig. 2(a) and Fig. 2(b) show the back gate transfer characteristics of graphene-WSe2 and WSe2 transistor, respectively. We found that the p-doped properties of WSe2-graphene heterostructure and WSe2 transistor are weakened by the anneal process. As the anneal process will remove the air molecule absorbed on the surface of flakes, we could know that the layered WSe₂ is *p*-doped by air molecule. Moreover, the *p*-type properties could be resumed after the exposing process. It is proved that air molecule will induce *p*-doped properties in layered WSe₂. Furthermore, *p*-doped properties of the graphene-WSe₂ heterostructure are also weakened by substituting the graphene with metal electrode. Since the

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