Enhanced Photosensitivity in Monolayer MoS$_2$ with PbS Quantum Dots

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Abstract Photocurrent enhancement has been investigated in monolayer (1L) MoS$_2$ with PbS quantum dots (QDs). A metal-semiconductor-metal (Au-1L MoS$_2$-Au) junction device is fabricated using a standard photolithography method. Considerably improved photo-electrical properties are obtained by coating PbS QDs on the Au-1L MoS$_2$-Au device. Time dependent photoconductivity and current-voltage characteristics are investigated. For the QDs-coated MoS$_2$ device, it is observed that the photocurrent is considerably enhanced and the decay life time becomes longer. We propose that carriers in QDs are excited and transferred to the MoS$_2$ channel under light illumination, improving the photocurrent of the 1L MoS$_2$ channel. Our experimental findings suggest that two-dimensional layered semiconductor materials combined with QDs could be used as building blocks for highly-sensitive optoelectronic detectors including radiation sensors.

Keywords: Photodetector, Molybdenum disulfide, PbS quantum dot

I. Introduction

Two-dimensional materials such as transition metal dichalcogenides (TMDCs) that consist of layered structures have received enormous attention [1,2]. In particular, TMDCs’ properties for the absorption and emission of photons can be exploited in solar cell arrays, wearable electronics, and transparent displays [3,4,5]. One prominent candidate material for optoelectronic devices is monolayer MoS$_2$, which offers a direct bandgap of 1.9 eV. Because of its unique optoelectronic properties and mechanical stability, it could be suitable for applications in optoelectronic devices [6,7]. However, the photocresponsivity and the sensitivity of monolayer MoS$_2$ are relatively low, and are poor compared with graphene-based photo-electronic devices [8].

For applications of high sensitive optoelectronics, emerging devices consisting of quantum dots (QDs) combined with graphene or atomically-thin MoS$_2$ have been demonstrated showing high gain and optical sensitivity [9,10]. In this study, we demonstrate a highly sensitive photodetector with 1L-MoS$_2$ coated with PbS QDs. We observe that the photocurrent and life time of a metal-1L MoS$_2$-metal device is improved by coating QDs on the 1L MoS$_2$.

II. Experiment

A monolayer MoS$_2$ film on SiO$_2$ was provided by the Professor Cha group at Oxford. The size of the MoS$_2$ flakes is on the order of several tens of micrometers. The MoS$_2$ flakes were grown by a conventional chemical vapor deposition method. Figure 1(a) shows an optical image of the MoS$_2$ flakes on the SiO$_2$ substrate. Fig. 1(b) shows the Raman spectrum confirming that the MoS$_2$ flake is a monolayer. The Raman spectrum exhibits two strong peaks: one is the in-plane $E_{1g}$ (384 cm$^{-1}$) and the other is the out-of-plane $A_{1g}$ (403 cm$^{-1}$). The difference $\Delta k$ between them is approximately 19 cm$^{-1}$, confirming that the MoS$_2$ flakes are predominantly one monolayer thick.

1. Device Fabrication

A conventional photo-lithography process was used to fabricate symmetric Au-1L MoS$_2$-Au junction devices on a SiO$_2$ substrate. The MoS$_2$/SiO$_2$/Si substrate was coated with a promoter and a photoresist (AZ 5214E) by using a spin coater (MIDAS, SPIN -1200D). Pre-coating was carried out at 1200 rpm for 10 seconds and the main coating was deposited at 5500 rpm for 1 minute. The uniformity of the photoresist layer is enhanced during this process. A hot plate was then used to perform soft-baking (pre-baking) for 50 seconds at 115°C. After the soft-baking process, UV light (wavelength: 350-450 nm, model: MIDAS, MDA-400M) was exposed for 7 seconds on the photoresist to make a device pattern, followed by a post-baking process at 115°C for 2 minutes. The exposure
process was then performed again for 10 seconds without the mask.

To make a desired pattern, the sample was immersed in a developer solution for 1 minute and cleaned in methanol and deionized (DI) water for 1 minute. After the desired pattern was formed on the MoS$_2$/SiO$_2$, metal deposition was carried out to make electrodes. Metal contacts were fabricated by thermal evaporation of Cr (20 nm) and Au (180 nm), respectively. Finally, acetone was used for lift off. The sample was then washed in DI water and dried using dry nitrogen gas. A schematic diagram of the fabrication process is shown in Fig. 2(a).

2. Method of PbS Quantum dots (QDs) coating

To coat PbS QDs on the MoS$_2$ channel layer sandwiched between the Au electrodes, a PbS QD solution was spin-coated onto the device at 3000 rpm for 15 seconds. Ligand solutions (tetrabutylammonium iodide) were applied for 30 seconds and then spun at 3000 rpm for 10 seconds. In order to remove impurity substance, the device was washed twice in methanol. To uniformly disperse the QDs on the surface, the spin-coating process was repeated 10 times.

3. Measurement of electrical properties

Figs. 2(a) and 2(b) show the fabrication method and an optical image of the Au-1L MoS$_2$-1L device, respectively. To measure the electrical properties (current-voltage characteristics), conventional 2-terminal transport

Figure 1. (a) Optical image and (b) Raman spectrum of MoS$_2$ flakes on a SiO$_2$/Si substrate. In the Raman spectrum, the frequency difference between the main peaks is 19.3 cm$^{-1}$, confirming the MoS$_2$ flake is one monolayer thick.

Figure 2. (a) Fabrication procedure and (b) optical image of Au-1L MoS$_2$-Au junction devices. (c) and (d) AFM images of 1L MoS$_2$ before and after coating the PbS quantum dots.
measurements were performed using a parameter analyzer (Keithley 4200-SCS). The bias-voltage ranged between -0.4 V and +0.4 V with an interval of 0.05 V. In order to measure the photocurrent characteristics of the device, a light controller (ILLUMINATOR KS-100H) was used in a dark box. The current-voltage characteristics of the device were measured at 0 Lux (light off), $1 \times 10^4$ Lux, $13 \times 10^4$ Lux.

**III. Results and Discussion**

Atomic force microscopy (AFM) images of MoS$_2$ with and without PbS QDs are presented in Figs. 2 (c) and (d). The morphology of the MoS$_2$ differs before and after coating the QDs.

Figure 3(a) shows the measured current-voltage characteristics of the Au-1L MoS$_2$-Au device without the QDs under different illumination intensities. As the intensity becomes stronger, the photo-current also increases. Figure 3(b) shows the time-dependent photo-response of the 1L-MoS$_2$ device without the QDs. The current was measured at a bias-voltage of 0.1 V with a laser intensity of $1 \times 10^4$ Lux. The laser pulse duration time was 15 seconds. When the laser was off, the photo-current decayed very rapidly.

The current-voltage characteristics of the QDs-coated 1L MoS$_2$ device measured under the same conditions are shown in Fig. 3(c). With the QDs, the photocurrent is considerably increased. This is presumably because electrons are transferred from the PbS QDs to the MoS$_2$ channel. Furthermore, the time-dependent photo-response of the Au-QDs-coated 1L MoS$_2$-Au device shows a considerably improved decay time, as shown in Fig. 3(d). These experimental findings suggest that hybridization of 2D layered semiconductors and quantum dots might be useful to improve the photo-electrical performance of the detectors and sensors.

**IV. Conclusions**

In order to investigate the effect of QDs on the optoelectrical properties of monolayer MoS$_2$ two different MoS$_2$ devices, Au-1L MoS$_2$-Au and Au-QD-coated 1L MoS$_2$-Au, were fabricated on SiO$_2$/Si substrates. We found that QDs improve the photo-current and the decay time in the 1L MoS$_2$, resulting in enhanced sensitivity. Our experimental findings suggest that 0D-2D hybrid devices are a promising device configuration for the next-generation optoelectronics devices for high-sensitivity detectors.

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