Low-Power, Large-Area and High-Performance CdSe Quantum Dots/Reduced Graphene Oxide Photodetectors

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ABSTRACT Graphene has unique and outstanding properties that make it a promising material for many applications. It has triggered considerable research in fields including solar cells, photodetectors, electrodes, and supercapacitors. Despite the favorable characteristics of devices using graphene have been widely explored, issues such as low absorbance, complex processing, and limited device size remain. Hence, we present large-area CdSe quantum dots (QDs)/reduced graphene oxide (rGO) films and corresponding photodetectors through a cost-effective and simple spin-coating method. As light turns on, CdSe QDs are excited and generate excess electron–hole pairs, leading to a significantly increased on/off current ratio of 2195 at a low bias voltage of $-1\, \text{V}$, compared to that of photodetectors without CdSe QDs. Decorating the rGO film with CdSe QDs enables the wavefunction modulation and enhances the light harvesting. Our proposed high-performance photodetector can be operated at a low voltage, which is beneficial for applications in various green and low-power consumption devices.

INDEX TERMS Large area, low power, photodetectors, reduced graphene oxide, CdSe quantum dots.

I. INTRODUCTION

Recently, graphene or other two-dimensional (2D) materials have attracted extensive interest owing to their outstanding properties, including high mobility, high transmittance, flexibility, and high conductivity [1]–[5]. Based on these merits, graphene has been employed in numerous applications, such as photodetectors [6]–[9], solar cells, supercapacitors [10]–[12], electrodes [13]–[15], and transistors [16]–[20]. However, monolayer graphene exhibits some limitations such as zero bandgap, weak light absorption and transfer-induced defects [21], [22]. To solve these problems, graphene composites have been developed to modify properties of graphene [23]–[26]. For example, it has been demonstrated that decorating graphene with metal nanoparticles can improve its performance in optoelectronic devices by the surface plasmon resonance effect [27]–[30]. However, the mass production of graphene-based devices is hindered by the complex fabrication process as well as the limited device size [31]–[33]. Moreover, the process of transferring graphene onto other substrates would damage the graphene film, resulting in the reduced performance. As a result, many researchers have attempted to develop various 2D materials beyond graphene [34] or low-cost reduced graphene oxide (rGO) films on a variety of substrates by a simple spin-coating method [35]–[38]. The decoration of rGO with metal nanoparticles [39]–[42], TiO$_2$ [43]–[45], and other 2D materials [46]–[49] has also been shown to further improve the characteristics of rGO. For example, rGO combined with Ag nanoparticles improves the performance of photodetectors owing to the surface plasmon resonance effect [41], [50].

Compared with traditional plasmonic metal materials, quantum dots (QDs) offer many advantages such as high extinction coefficients, narrower photoluminescence (PL)
spectra, brighter fluorescence, and ability to modify the semiconductor bandgap [51], [52]. Additionally, QDs can emit light at different wavelength by controlling the size, shape, or type of QDs [53]–[58]. This characteristic provides an effective way to tune the spectral response over a wide range for optoelectronic device applications. Therefore, the combination of rGO and QDs may potentially modify the light absorption property and enhance the carrier transport. However, little attention has been given to the development of large-area CdSe QDs/rGO films and the effect of wavefunction modulation on the performance of photodetectors.

In this study, we propose a simple fabrication method for large-area fluorescent CdSe QDs-tagged rGO photodetectors. CdSe QDs/rGO-based photodetectors are characterized under a low voltage bias to assess their performance for low-power consumption applications. As CdSe QDs/rGO films are under illumination, QDs are excited and the fluorescent phenomena is observed. This effect can modulate absorbance spectra, enhance the photocurrent and thus improve the on/off ratio of CdSe QDs/rGO photodetectors. These results demonstrate that CdSe QDs/rGO photodetectors have great potential not only for low-power consumption, but also open a possible strategy for broadband spectrum modulation in various devices.

II. EXPERIMENTS
Fig. 1(a) shows the fabrication process and the schematic structure of rGO and CdSe QDs/rGO photodetectors. First, Si substrates were rinsed by deionized (DI) water, cleaned in hydrochloric acid (HCl) and hydrofluoric acid (HF), and then rinsed again in DI water to remove native oxides. Subsequently, SiO₂ (thickness: 300 nm) was deposited on Si substrates by sputtering with a power of 20 W. The absorber layer was constructed by rGO and CdSe QDs/rGO films. The photograph of CdSe QDs solution is shown in Fig.1(b) and the diameter of the CdSe QDs is approximately 2–4 nm. For the fabrication of rGO, GO flakes were synthesized from graphite by a modified Hummer’s method. The GO flakes were then reduced to rGO; GO in DI water was sonicated in a flask, then hydrobromic acid was added to the GO colloids. The GO and hydrobromic acid mixture were refluxed in an oil bath at 110 °C. To create the CdSe QDs/rGO composite, CdSe QDs solution were added into rGO solution (CdSe QDs: rGO = 1:2). Then 80 µL rGO and CdSe QDs/rGO solutions were coated on 2 cm × 2 cm SiO₂/Si substrates by a spin coater at a spin rate of 3000 rpm for 30 s. Fig. 1(c) presents a large-area CdSe QDs/rGO film on a SiO₂/Si substrate; the width and length of this film are around 2 cm. Finally, 120 nm-thick silver electrodes were deposited on rGO and CdSe QDs/rGO films by sputtering. Fig. 1(d) shows the band energy diagram of the CdSe QDs/rGO/Ag structure. When CdSe QDs absorb incident light, photo-excited electrons transfer from the bottom of QDs conduction band to or above rGO levels. In addition, holes and electrons from electronic levels around rGO transfer to level below the top of QDs valence band.

III. RESULTS AND DISCUSSION
Fig. 2(a) presents an SEM image of the rGO film on the Si substrate. As can be seen from this figure, rGO flakes
are successfully coated on the substrate. A large-scale SEM image of rGO film is displayed in Fig. 2(b) which conforms that the large-scale rGO film is continuous and uniformly distributed. To investigate the roughness of the rGO film, an AFM image was taken as shown in Fig. 2(c). Based on the cross-sectional analysis of the AFM measured result, the roughness of the rGO film is about 5-40 nm. The diameter of CdSe QDs is 2-4 nm which can be evaluated by the TEM image as shown in Fig. 2(d). The large-scale TEM and AFM images are displayed in Fig. 2(e) and (f), respectively. They reveal that CdSe QDs are uniformly dispersed on the rGO film and nearly no overlapping between each QDs. Therefore, we may consider that CdSe QDs are one layered and the thickness is about 2-4 nm which is the diameter of CdSe QDs.

Raman spectroscopy is an effective and non-invasive tool for characterizing carbon-based materials. Here, we employ Raman spectroscopy with 633 nm excitation to determine the quality of rGO and CdSe QDs/rGO samples as shown in Fig. 3(a). It can be observed that both samples exhibit prominent G bands and D bands. For the rGO film, peaks of G band and D band are presented at 1347 and 1580 cm$^{-1}$, respectively. For the CdSe QDs/rGO film, peaks of G band and D band are displayed at 1336 and 1580 cm$^{-1}$, respectively. The D band is resulted from the breathing mode of the K-point phonons of A1g symmetry involving phonons near the K zone boundary, while the G band is ascribed to the first order scattering of phonons with E2g symmetry of the sp$^2$ carbon atoms [58]. The remarkable blue shift of D band in the CdSe QDs/rGO Raman spectrum could be attributed to the strong chemical interaction between CdSe QDs and rGO. The $I_D/I_G$ intensity ratio for rGO and CdSe QDs/rGO film is 1.03 and 1.04 respectively, suggesting a decrease in the average size of the sp$^2$ domains. To understand the energy transfer mechanism from CdSe QDs to the rGO film, the PL quenching effects were measured. The rGO film (acceptor) acts as an effective quencher of excited CdSe QDs (donor), that is to say, the rGO film can capture and store carriers from respectively.
QDs. As shown in Fig. 3(b), the PL spectrum of the CdSe QDs/rGO composite exhibits a strong peak at 540 nm when the sample is excited by a 325 nm light source. This PL peak is associated with a high rate of electron–hole pair recombination. The stronger the PL intensity, the higher the rate of recombination of photogenerated electron–hole pairs. The full width at half maximum (FWHM) of the CdSe QDs/rGO sample is about 50 nm, which is similar to the reported value for CdSe QDs [59]. In contrast, the rGO film without QDs has a flat PL curve, revealing that CdSe QDs on the rGO film bestows it with obvious fluorescence.

The spectral response of photodetectors is related to the light absorption and carrier transportation behavior. To study the light harvesting abilities of CdSe QDs and rGO films, the ultraviolet–visible (UV–vis) reflectance and absorbance spectra of Si, SiO\(_2\)/Si, rGO/SiO\(_2\)/Si, and CdSe QDs/rGO/SiO\(_2\)/Si samples were measured. The reflectance spectra in Fig. 4(a) reveal that when the rGO film is deposited, a slight decrease in the reflectance value is observed versus Si and Si/SiO\(_2\) alone. Additionally, the CdSe QDs/rGO composite film shows a modified and reduced reflectance spectrum. Fig. 4(b) displays the absorbance spectra of Si, SiO\(_2\)/Si, rGO/SiO\(_2\)/Si, and CdSe QDs/rGO/SiO\(_2\)/Si samples. Compare absorption curves between rGO/SiO\(_2\)/Si (green line) and CdSe QDs/rGO/SiO\(_2\)/Si (yellow line), the CdSe QDs/rGO/SiO\(_2\)/Si curve is shifted due to light interference in the visible range. Meanwhile, the absorption value of CdSe QDs/rGO/SiO\(_2\)/Si is significantly enhanced at the wavelength longer than 750 nm, demonstrating that CdSe QDs can effectively emit light and then the rGO film absorbs the emission in the given region. As a result, the improved absorption of CdSe QDs/rGO may be ascribed to excited carriers from CdSe QDs to the rGO film. Then the rGO film serves as the absorber layer and provides a transfer pathway of carriers.

In order to characterize the performance of rGO and CdSe QDs/rGO photodetectors, current–voltage (I–V) tests were performed using a 532 nm laser, a function signal generator, and a probe station. The I–V curves of photodetectors were measured by sweeping the voltage from −5 to 0 V under 0.14, 3.67, 7.33, 10.5, and 22.1 W cm\(^{-2}\) light density illumination at room temperature. The I–V measured results of rGO and CdSe QDs/rGO photodetectors are displayed in Fig. 5(a) and (b), respectively. It is found that both rGO and CdSe QDs/rGO photodetectors exhibit distinguishable currents in light and dark conditions, indicating that the rGO
film is well-distributed. In addition, the light current over dark current ratio ($I_{on}/I_{off}$) for the CdSe QDs/rGO photodetector (Fig. 5(b)) is improved compared to that of the rGO photodetector at a voltage of −1 V (2195 vs. 12). As light turns on, CdSe QDs are excited and electron-hole pairs are generated in the conduction band and the valence band. Electrons on the conduction band of CdSe QDs created by light excitation are accepted by rGO flakes. At the same time, holes in the valence band of a rGO film inject to the valence band of CdSe QDs. This will improve the separation of excited electrons and holes. Hence, the photocurrent of the CdSe QDs/rGO photodetector is enhanced compared to that of the rGO photodetector without excited carriers from QDs. Additionally, from the absorbance spectra, it can be estimated that rGO film absorbs only about 2~10 % of incident light. After the decoration of CdSe QDs, the absorption is significantly enhanced especially in the 400~500 nm and 700~1100 nm wavelength ranges. Therefore, the improved photocurrent of the CdSe QDs/rGO photodetector may be caused by the excited carrier from CdSe QDs.

Fig. 6(a) and (b) show the dependence of the absolute light current on the light intensity for rGO and CdSe QDs/rGO photodetectors operated at a bias of −5 V, respectively. As the light intensity is increased, more photons are absorbed in rGO and CdSe QDs/rGO layers, leading to an enhanced light current. As explained in PL and $I − V$ measured results, excited carriers transfer from CdSe QDs to the rGO film. Then carriers are captured in the rGO film and collected by Ag electrodes. When the power of incident light is increased, stored electrons in the rGO film is saturated, indicating that the electron transfer process will slow down, and the sensitivity will therefore be saturated. Moreover, to study the concentration of CdSe QDs on the performance of photodetectors, we also mixed CdSe QDs and rGO solutions with different ratio (CdSe QDs: rGO = 1:1). Then the mixed solution was coated on SiO$_2$/Si substrates by a spin coater with the same spin rate of 3000 rpm for 30 seconds. CdSe QDs: rGO = 1:2 sample exhibits higher photocurrent then that of CdSe QDs: rGO = 1:1 sample as light intensity larger than 7.33 W/cm$^2$. This is because CdSe QDs with proper density display a fluorescence property and CdSe QDs also don’t obstacle the absorption of rGO. Therefore, by controlling the density and the distribution of CdSe QDs and rGO, the photocurrent could be obviously improved under low and high light intensity illumination.

We measured the external quantum efficiency (EQE) of rGO photodetectors with and without CdSe QDs to explore the photoresponse of photodetectors. These photodetectors were operated at a bias of −5 V under different wavelength illumination ranging from 300 to 1100 nm. As shown in Fig. 7, the EQE values of the CdSe QDs/rGO photodetector are enhanced from 2% to 50% and 8% to 20% at wavelength around 450 and 900 nm, respectively, compared to those of the rGO photodetector. The enhancement may come from the light harvesting effect by CdSe QDs. Additionally, as CdSe QDs absorb the incident light, they create excess carriers at some wavelength regions and then carriers transfer to rGO, giving rise to an improved EQE response. From the EQE characteristics of rGO and CdSe QDs/rGO photodetectors, it is also proved that both rGO and CdSe QDs/rGO photodetectors have photoresponse in wide wavelength ranges, revealing that these photodetectors can operate under UV, visible and near infrared illumination.

Fig. 8(a) and (b) display the absolute light current–time response of rGO and CdSe QDs/rGO photodetectors, respectively. Both photodetectors are measured in the dark and under illumination with a laser light source of 532 nm at a bias of −5 V. As the laser source is turned on, the light current increases sharply owing to the effective carrier generation and transport. Moreover, rGO and CdSe QDs/rGO photodetectors exhibit reproducible light current over several on/off cycles. The response time of a photodetector is estimated as the time required to change the photocurrent intensity from 10% to 90%. Based on a single-cycle on/off curve of
FIGURE 7. EQE characteristics of rGO and CdSe QDs/rGO photodetectors. The photoresponse of the CdSe QDs/rGO sample is enhanced due to effective wavefunction modulation and light harvesting.

FIGURE 8. Light current–time response of (a) rGO photodetector and (b) CdSe QDs/rGO photodetectors measured in the dark and under illumination using a laser source of 532 nm at a fixed bias of −5 V. Photocurrent as a function of time for the calculation of response time of (c) rGO and (d) CdSe QDs/rGO photodetectors.

FIGURE 9. Detectivity as a function of voltage of rGO and CdSe QDs/rGO photodetectors operated from 0 to −5 V under 0.14 W/cm² illumination.

IV. CONCLUSION

In conclusion, a large-area CdSe QDs/rGO film was created by a simplified fabrication process, and the corresponding photodetector was developed. The $I - V$ curve of the CdSe QDs/rGO photodetector demonstrates that the photodetector has a superior $I_{on}/I_{off}$ ratio of 2195 even at a low bias of −1 V. In addition, the light current-light intensity and the light current–time response reveal that the photodetector possesses good reproducibility and reliable performance. Decorating the rGO film with CdSe QDs improves characteristics of the CdSe QDs/rGO photodetector owing to the enhanced light harvesting and the effective wavefunction engineering. In the future, we will further construct rGO photodetectors on flexible substrates which may open a new way for wearable, high-performance and low-power consumption graphene-based products.

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