



Graphene films as transparent electrodes for photovoltaic devices based on cadmium sulfide thin films



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ARTICLE INFO

Keywords:

Graphene
CVD process
Raman spectroscopy
Optical spectroscopy
Optical constants
Photoconductivity

ABSTRACT

Graphene films were synthesized and characterized to demonstrate their potential applications as the transparent electrodes in photovoltaic devices. Graphene films were prepared by chemical vapor deposition of graphene on copper substrates followed by transferring them onto the desired substrates. The quality and uniformity of graphene transferred onto different substrates were characterized by Raman spectroscopy and UV–VIS–NIR reflectance as well as transmission spectroscopy. The results showed that the films synthesized contained 1–3 layers of graphene with the sheet resistance and optical transparency in the ranges of 50–350 Ω/sq and 85–95%, respectively. Thin films of cadmium sulfide (CdS) were deposited on different substrates by chemical bath deposition method to study the optoelectronic behavior at the interface of graphene/CdS. The results showed that transient response time and the magnitude of photocurrent were enhanced by the graphene electrodes. The order of photocurrent merit was found to be CdS/graphene /Glass > CdS/ITO/glass, indicating that the CdS/graphene stack is to be the most efficient electron transport matrix for the separation of photo excited carriers in the CdS films.

1. Introduction

Graphene, with its carbon atoms arranged in honeycomb lattice [1,2], has proven to be a useful electronic material after its first discovery in 2004 [3] because of its minimum thickness of 0.32 Å [4,5] and unique physical and electronic properties [1–3]. Its ambipolar transistor behavior [6] high intrinsic carrier mobility of 5000 to 20,000 $cm^2/V.s$ for flat-bed transistors [7] and its tenfold improvement in the mobility exceeding 200,000 $cm^2/V.s$ for suspended graphene films [8] have drawn worldwide attention in semi-conductor industry. In addition to high carrier mobility, graphene based thin film transistors (TFTs) also exhibit large critical current densities ($\sim 2 \times 10^8$ A cm^2) [9], high saturation velocity ($\sim 5 \times 10^7$ cm/s) and very high cutoff frequency up to 100 GHz [10]. Recently, thermally reduced graphene oxide and CNT based devices have been reported to have a strong absorption behavior in the near infrared (NIR) region [11,12]. This unusual absorption behavior may provide a pathway to use graphene as an optoelectronic material for infrared remote sensors [13] for the utilization of NIR light in solar-cells and for the successful manifestation of ultrafast optical devices [14–17]. In addition to its superiority over other electronic materials, graphene has also been investigated as a potential candidate for transparent electrodes in photovoltaic devices [18–21] due to its high transparency ($\sim 97.7\%$ for monolayer) [22] and

relatively low sheet resistance (\sim few hundred Ω/sq) [21,23]. The characteristic feature of graphene is that even with more than 97% of transmission, it absorbs rest of the light ($\sim 2.3\%$ for monolayer) in the visible spectrum without any significant reflection ($< 0.1\%$) [1,3]. Among many different transparent electrodes available in industrial scale, indium tin oxide (ITO) glasses are commonly used in solar cells [20,22,24,25]. Generally, commercially available ITO glasses show the optical transparency $\sim 90\%$, reflection $\sim 6\%$ in visible region (550 nm), sheet resistance ($< 100 \Omega/sq$) and infinite scalability [25]. Since defect-free graphene transmits more than 95% of visible light with negligible reflection, it can replace the existing transparent electrode materials, such as ITO and CNTs in optoelectronic devices. Until now, very few researchers have demonstrated the use of graphene as a transparent electrode in optoelectronic devices. X. Wang et al first demonstrated the performance of transparent graphene as electrode in organic solar cell [18]. Q. Su and et al constructed the heterojunction solar cell with PEDOT: PSS and ZnO as the buffer layer with conductive graphene oxide (GO) and reduced graphene oxide (RGO) films [20]. Deposition of graphene by chemical vapor deposition (CVD) has been investigated to produce large-area graphene layers. It has been reported that a more conductive CVD graphene film with sheet resistance $\sim 230 \Omega/sq$ and a transparency of $\sim 72\%$ (at around 550 nm) showed the performance comparable to ITO based device [23]. In this work, we have synthesized

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a large area and uniform graphene on a copper foil by CVD method and transferred them onto a different substrates followed by etching of copper substrate. These graphene films have a optical transparency in the range of 85–95%, negligible reflection ($< 0.4\%$) and low sheet resistance in the range of $300 \pm 25 \Omega/sq$.

Nanostructured semiconductor films based on cadmium compounds (CdS, CdSe, CdTe) have been reported very effective for their use in thin-film solar cells with the efficiencies up to $\sim 20\%$ [26–28]. Recent reports have shown that the semiconductor quantum dots based on cadmium compounds can improve photovoltaic efficiency by generating multiple charge carriers under photo-excitation [29–31]. However, the major impediment in cadmium based semiconductors is the fast recombination of charge carriers generated by photo-excitation process [26–31]. In order to enhance the photovoltaic conversion process, it is essential to effectively retard the recombination of electron-hole pairs [32]. One way to minimize recombination process is that the coating of an efficient electron-transport material, such as conductive polymer films, carbon nanotubes (CNTs), graphene [29,32–34]. Graphene is superior to other materials as an electron-transport matrix due to its high electrical conductivity and optical transparency ($\sim 95\%$). Although graphene/CdS nano-composites synthesized by chemical method have been used by many researchers for photocatalysis [32,35–37] and optoelectronic devices [38], very few researchers have reported the use of large area graphene as electron transport matrix with cadmium based semiconductor films [38,39]. In this work, CVD graphene films were investigated as the transparent electrodes for CdS layers with excellent electron transfer matrix.

2. Experimental details

2.1. Synthesis, transfer and fabrication of graphene based devices

Graphene films were grown on a copper foil (Alfa Aesar, 99.9995%, $25 \mu\text{m}$ thick) by a CVD method at a temperature of 1100°C . After graphene films were deposited on a Cu substrate, it was transferred to desired substrate (glass, Au or CdS) by etching of Cu films with aqueous 1.0 M FeCl_3 at room temperature. The details about the synthesis and the transfer procedure of graphene films have been discussed elsewhere [5,40–44]. In order to fabricate the devices, a thin layer of gold ($\sim 100 \text{ nm}$) was coated on the surface of the transferred graphene film by a vacuum evaporation method. The devices were patterned by photolithography process followed by the etching of gold with aqueous KI/I_2 solution. Photolithography began with the substrate that was previously cleaned with Isopropyl alcohol (IPA) followed by rinsing with the acetone. The substrates were then coated with a photoresist layer and exposed to a focused beam of UV light through the mask. The exposed areas of the photoresist (PR) were dissolved in a developer solution to create the PR patterns. The CdS thin films were deposited on the gold patterns by chemical bath deposition (CBD) method described in Section 2.3. A similar technique was used to deposit CdS thin films on the surface of graphene films. Finally, the PR were removed by rinsing with acetone.

2.2. Deposition of CdS thin films

Polycrystalline CdS thin films were deposited on different substrates by a CBD method. The reagents used in CBD include 0.2 mmol of $\text{Cd}(\text{NO}_3)_2$, 0.4 mmol thiourea (H_2NCSNH_2), 0.9 mmol of NH_4NO_3 , and NH_4OH (15 ml). These chemicals were dissolved 120 ml of DI water. The beaker containing the final solution was kept in a hot water bath to maintain the temperature. All the depositions were carried out at an optimal growth temperature of 70°C . The cleaned substrates were tied up with a Teflon tape and dipped in the above solution. The deposition was carried out for 15 min to make sure that the CdS films are pinhole free. After deposition, the CdS films were washed with DI water to remove the extra solvents and CdS particles on the films and dried

under nitrogen ambient at 120°C .

2.3. Materials and measurements

The quality of the graphene films was examined by Raman spectroscopy, scanning electron microscopy (SEM), and optical spectroscopy. The SEM images were obtained using a scanning electron microscope (Carl Zeiss SMT AG Supra 35VP FE) operating at an accelerating voltage of 15.0 kV and using an in-lens ion annular secondary electron detector. The Raman spectra of graphene were obtained at the room temperature with a Raman spectrometer (Renishaw-Invia) in back scattering geometry with the laser excitation of 632.8 nm at a power level of 1.7 mW . The morphology and the structure of CdS films were also determined by the X-ray diffraction (XRD, D/MAX2500) with a $\text{Cu K}\alpha$ radiation. The optical transmittance and reflectance spectra of graphene films, CdS films, ITO and glass substrates were measured with a double beam UV–VIS–NIR spectrophotometer (Shimadzu). Electrical measurements on graphene/CdS films were performed by connecting devices with a HP-4145B semiconductor parameter analyzer and a high impedance multimeter (Keithly, Model-1200) via electrical feed-through on a test chamber. A laser beam of 520 nm (2 W) was used as the IR light source.

3. Results and discussion

3.1. Structural properties

Fig. 1(a) shows typical Raman spectra of few layer graphene. As reported by many researchers about quality graphene synthesized by CVD process, the Raman peaks appeared corresponding to the D band ($\sim 1330 \text{ cm}^{-1}$), G band ($\sim 1580 \text{ cm}^{-1}$) and 2D band ($\sim 2660 \text{ cm}^{-1}$) [45] for films produced in this work. From the typical spectra shown in Fig. 1(a) for few-layer graphene, the following conclusions can be drawn:

1. The spectra consist of D band, indicating the presence of some defects in the crystal. The intensity of D band relative to G band (I_D/I_G) was found to increase with the increase in number of layers in graphene. Although the intensity ratio increased significantly (~ 0.16 to ~ 0.45), the change in FWHM was not significant ($\sim 30 \text{ cm}^{-1}$ to $\sim 35 \text{ cm}^{-1}$) in going from monolayer to bi- and tri-layer graphene. However, the broadening of D band increased significantly after three layers. The intensity of D band appeared in the Raman spectra for monolayer graphene is either comparable or relatively less than the intensities reported by others [5,45,46]. However, its unavoidable to have defects in graphene synthesized on copper substrate through a CVD process. These defects could be associated with the grain boundaries in the copper foils where graphene was initially made. This band also indicates the vacancies and strained hexagonal and non-hexagonal distortions in the film that may have arisen during transfer process [47].
2. The intensity of G band did not change from monolayer to few layer graphene. The intensity and FWHM of 2D band relative to G band (I_{2D}/I_G) changed significantly from monolayer to few layer graphene. The intensity of 2D band was twice as strong as G band for monolayer graphene, whereas it was comparable with the intensity of G band for tri-layer graphene and decreased for further increase in number of layers. On the other hand, FWHM for 2D band increased slowly for graphene layers up to tri-layer and increased significantly afterwards. The broadening of 2D band corresponds to the increase in number of layers. All D, G and 2D bands were shifted towards higher Raman shifts with increasing the number of layers.

The uniformity of the transparent conducting layer is one of the important characteristics in designing optoelectronic devices. To investigate the uniformity of the layer, a $2 \text{ by } 2 \text{ cm}^2$ area of graphene

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