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Direct growth of nanocrystalline graphene/graphite all carbon transparent electrode for graphene glass and photodetectors

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ABSTRACT

Nanocrystalline graphene/graphite hybrids all carbon transparent electrodes were fabricated from photoresist with a photolithography method by the carbonization and graphitization of the aromatic molecules in the photoresist. The sheet resistance and transmittance of the hybrids transparent electrodes can be easily optimized by modulating the photoresist grid size and gridline width. The optimized hybrids were fabricated on quartz for graphene glass that exhibits excellent properties for heating device. They were also engaged on Si with a thin oxide layer by a catalyst-free and direct growth method for high-performance Schottky junction photodetectors that exhibit remarkable properties on photodetection with the photovoltage responsivity exceeding up to 10^5 V/W under the incident light power of 0.05 μW, which make it suitable as weak-light-signal detectors. This study should be helpful on the various applications of catalyst-free, cost-effective, transparent, electrically and thermally conductive graphene.

1. Introduction

Graphene is composed of one-atom-thick hexagonal network of carbon atoms packed with a two-dimensional (2D) honeycomb lattice, which is deemed as one of the most attractive candidate materials for next-generation optical and electrical devices due to its unique properties [1–8]. It has been proved that the sheet resistance is as low as 30 Ω/□ and the transmittance is around 97.7% for the monolayer graphene, which is superior to the current commercial indium tin oxides (ITO) transparent electrodes [9–11]. Furthermore, in comparison with ITO, graphene has high mechanical strength and excellent flexibility [12]. These extraordinary properties make graphene suitable for future stretchable transparent electrodes applications in solar cells [13–16], field effect transistors (FETs) [17–19], photodetectors [20,21], light-emitting diodes (LEDs) [22,23], and touch screens [24,25]. As we all know, in order to realize the application of graphene in real nanoelectronics, the transparence and conductivity are critical aspects [12,26]. Several approaches have been applied to improve the conductance of graphene transparent electrodes like doping graphene and introducing metal nanowires onto graphene films [11,27,28]. However, the instability of the graphene-dopant system and the high cost of metal nanowires may limit the application of these methods. Thus, for realistic applications, high-performance devices using large-area high quality graphene transparent electrodes with simple and cost-effective synthesis methods are highly desirable.

Nanographene/graphite has been successfully fabricated from photoresist through the carbonization and graphitization of the aromatic molecules in the photoresist at high temperature [29–31]. In this work, we developed a catalyst-free tunable method to directly grow large-scale nanocrystalline graphene/graphite all carbon hybrids patterns on arbitrary substrates by using

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photoresist. We demonstrated that the sheet resistance and transmittance of the nanographene/graphite all carbon transparent electrodes can be easily adjusted by modulating the photoresist grid size and gridline width, whose optimized performance is comparable to that of the reported carbon nanotube and Ag nanowire based transparent/conductive materials [32–34]. The performance (sheet resistance 2 kΩ/□ and transmittance 85%) of our optimized sample should be good enough for certain electronic devices because the similar transparent electrodes obtained by chemical exfoliation graphene were applied in solar cells [35]. Notably, we can directly grow nanographene/graphite all carbon hybrids on quartz substrates. Thus, graphene glasses for heating devices were easily obtained. At the same time, nanographene/graphite hybrids were successfully applied for transparent electrodes on silicon to form graphene-silicon heterojunction photovoltaic configurations, which are attracting great attention owing to their simple fabrication and extraordinary photoelectric properties [28,36–44]. Because the source material for nanographene/graphite all carbon transparent electrodes is photoresist and the fabrication process is mainly based in the current-used photolithography, we provide simple, controllable, environmental friendly and low-cost synthesis method to develop nanographene/graphite all carbon transparent electrodes for high performance graphene glass and functional optoelectronic device applications in the future.

2. Experimental

2.1. Fabrication of nanographene/graphite on quartz substrates for transparent electrodes

Nanographene/graphite transparent electrodes were fabricated from the photoresist (purchasing from the Suzhou Ruihong Electronic Chemicals Co. Ltd.). As shown in Fig. 1, firstly, 1.5-μm-thick photoresist film was spin-coated on the quartz substrate and the film was baked on the hot-plate (110 °C) for 3 min. Secondly, different grid size and gridline width of photoresist grids were patterned on the quartz substrates with the standard photolithography method. Thirdly, the samples were then heated for 10 min in a vacuum horizontal quartz tube at 1000 °C under the protection of 100 standard cubic centimeters per minute (sccm) 5% H_2/Ar gas flow.

2.2. Fabrication of nanographene/graphite grids structures on SiO_2/Si with thin oxide layer for high-performance Schottky junction photodetectors

The silicon used here is n-type with a 300-nm-thick thermal layer, which has a resistivity of 1–10 Ω cm. Nanographene/Graphite Grids Structures on SiO_2/Si structures for photodetectors were fabricated as schematically shown in Fig. 2: (1) 1.5-μm-thick photoresist film was spin-coated on SiO_2/Si substrates and an area of 3 mm × 3 mm of photoresist grids are patterned with the standard photolithography, the grid size is 200 μm × 200 μm, and the gridline width is 20 μm; (2) The as-obtained photoresist structures on SiO_2/Si was wet-etched by BOE (NH_4F: 10 wt% HF: H_2O = 8 g: 2 ml: 12 ml). After the patterned silicon exposed thoroughly, they were then rinsed with deionized (DI) water for three times; (3) The photoresist film on the SiO_2/Si structure was absolutely removed with acetone and rinsed with DI water repeatedly; (4) The dried SiO_2/Si structure was treated in air gas flow at 800 °C for 20 min to get a thin oxide film on the exposed silicon window; (5) 1.5-μm-thick photoresist structure was aligned on the square window; (6) The samples were then heated for 10 min in a vacuum horizontal quartz tube at 1000 °C under the protection of 100 sccm 5% H_2/Ar gas flow. In order to avoid the contact between the nanographene and the side silicon, the central part was protected, while the nanographene around the edge was etched by oxygen plasma. In the end, the aimed device comes into being.

2.3. Characterizations

X-ray photoelectron spectroscopy (XPS) was performed with a VG Scientific ESCALab 220i-XL electron spectrometer. The transmission electron microscope (TEM) images were taken by a Tecnai F20 field-emission TEM. The transmittance of the nanographene/graphite hybrids transparent electrodes was measured with a Hitachi U-3900H UV-VIS spectrophotometer, and the sheet resistance was measured by a four-probe setup with a ST-2258A source meter. The water contact angle (CA) of the nanographene/graphite quartz glass was obtained by using the Optical Contact Angle Measuring Device (OCA15EC, Dataphysics, Germany). The electrical and photoelectrical properties of the produced photodetector were characterized with a Keithley 4200-SCS semiconductor analyzer at room-temperature in atmosphere. The light source was a light-emitting diode (LED) lamp whose power can be modulated. We used an optical power meter of SGN-1 to measure the power of the light and calculated the incident power on the configuration according to their area.

3. Results and discussion

In order to improve the conductance of graphene transparent electrodes, nanographene/graphite all carbon transparent electrodes have been successfully fabricated. The growth process is schematically shown in Fig. 1a and details of the transparent electrodes fabrication process are described in the experimental section. Firstly, 1.5-μm-thick photoresist film was spin-coated on the

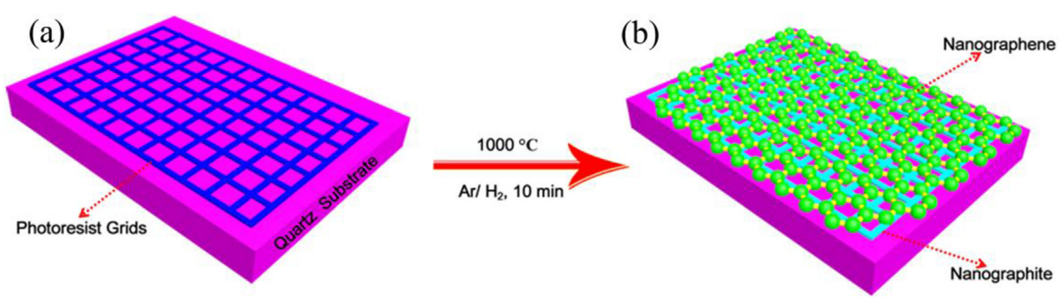

Fig. 1. - Schematic illustration of the fabrication nanographene/graphite grids structures on quartz substrates. (a) Photoresist grids are patterned on the quartz substrates with the standard photolithography method. (b) After high-temperature treatment, the photoresist grids is transformed into nanographite with nanographene formed on the around parts. (A colour version of this figure can be viewed online.)