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Enhancement of broadband ultraviolet visible photodetection by boron nitride nanoparticles in bulk graphene oxide layer

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ABSTRACT

A proposed boron nitride/graphene oxide (BN/GO) composite photodetector (PD) was prepared using a drop cast method for photodetection in the ultraviolet (UV) and visible light ranges. Morphology of the GO, distribution of BN nanoparticles and composition of B, N, C and O elements were confirmed by field emission scanning electron microscope (FESEM) analysis and energy dispersive x-ray (EDX) spectroscopy results. Raman scattering indicates high frequency intrinsic E_{2g} vibrations at 1349 cm^{-1} and low intense peaks at 803 cm^{-1} from hexagonal BN (h-BN) structures in the bulk GO layer, while an overlap in the D and G bands at 1349 and 1581 cm^{-1} respectively indicates the presence of the GO layer. Excellent photoconduction of 380 and 405 nm UV sources and a 650 nm red laser source showed that the measured photocurrent is dependent on power regardless of the signal wavelength. Variations in sensitivity allow the device to be operated at selected direct current (DC) bias voltages, while high frequency modulation at 1000 Hz showed a profound rise and fall time about 13.6 and 245.6 μs respectively. Characteristics as self-powered PD was observed, has an enhancement about 3450% at its zero bias voltage.

1. Introduction

Recently, photodetection technology has seen significant and rapid advancement due to the discovery of new functional materials such as gallium nitride (GaN), aluminium nitride (AlN), boron nitride (BN) [1,2], graphene oxide (GO) [3], reduced graphene oxide (rGO) [4], zinc oxide (ZnO) [5] and black phosphorus [6]. These materials play an important role in the development of optoelectronic applications, encompassing areas such as defence, sensors, health and climate change. In particular, nitride semiconducting materials have shown significant potential for the development of new photoconductors. Of the many nitrides studied, BN in particular has the advantage of synthesizing nanoscale structures with various morphologies that are suitable for deep ultraviolet (UV) optoelectronic applications [7]. Likewise, hexagonal BN (h-BN) and cubic-BN (c-BN) which have similar structures to that of graphite and diamond respectively have bandgaps greater than 5, high thermal conductivity, high hardness and stable electrical resistivity [8]. The bandgap of c-BN at 6.3 eV [9] is higher than that of diamond, suggesting that c-BN materials would be an ideal structure for deep solar blind photodiodes (PDs) [10,11]. Furthermore, the width of wide bandgap of BN based materials can be modulated to reach as low as 2 eV or less during synthetization [12]. Li et al. [13] used the plasma-enhanced chemical vapor deposition (PECVD) to adjust the bandgap of c-BN to 4.78 eV by the introduction of sulphur elements as dopants. However, the difficulty in synthesizing high quality c-BN remained a primary challenge. On the other hand, Soltani et al. [10] used a c-BN substrate as a semiconductor to fabricate a deep UV metal/semiconductor/metal configured PD using electron beam deposition, with the formed interdigitated circular electrodes enabling the high homogeneity of electric fields between the electrodes and realizing a peak responsivity of 180 nm. Following that, Jiang et al. [14] and Dahal et al. [14,15] suggested that h-BN films would have the ability to meet the development requirements of deep UV photonic devices compared to c-BN structures. Thus, a variety of h-BN nanostructures in nanosheets [16], nanotubes [17] and nanorods [18] have been synthesized. In this manner, the BN nanosheet, which is an isoelectric analogy of graphene becomes of significant interest owing to the many similar physical properties and structural characteristics they share [19].

However, PDs that are powered externally face a number of issues including high maintenance cost, limited lifetime and environmental issues [20]. Therefore, there is a need for a self-powered, flexible and wearable devices that can operate without any external power supply. These self-powered PDs are attracting significant attention as they have key advantages such as low cost, energy saving operation, fast response

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time, high photoresponsivity and good photosensitive linearity over a wide light intensity range [21]. Self-powered PDs can be realized through the functionalization of two dimensional (2D) materials such as graphene by exploiting its zero bandgap [22]. Chemically tunable graphene was reported to be able to facilitate the fabrication of environmental sensitive polarizers for light, humidity and sound measurement [23]. Both GO and graphene have direct bandgaps between 0 and 1 eV which could allow for tunability of bandgap, such as the bandgap of graphene monoxide at about 0.952 eV [24]. Velasco-Soto et al. [25] demonstrated a method for controlling the bandgap in GO layers using an environmental friendly reagent like fructose to tune the bandgap from 2.7 to 1.15 eV [26], and this ability is an important characteristic in enabling the absorption of infrared (IR) photons for photodetection. Miao et al. [27] also noted that the efficiency of graphene based Schottky junction devices can be optimized by tuning the work function as compared to indium tin oxide (ITO) Si junction device.

In this regard, GO is a highly potential candidate for the development of self-powered PDs as it possesses carboxyl and hydroxyl functional groups [28] that form multifunctional structures with 2D materials such as BN and ZnO [29,30]. A multifunctional GO - BN structure could maximize the structural and thermal properties of GO while preserving its optical properties. Simulation studies by Shiue et al. [31] on graphene-BN PDs coupled to the optical mode of a silicon (Si) waveguide exhibited maximum responsivity of 0.36 AW^{-1} and high speed operation with a 3 dB cut off at 42 GHz. Chemical vapor deposition (CVD) [32] and pulse laser plasma deposition (PLPD) [33] were successfully utilized to obtain the desired BN nanosheets, though both these techniques are not without their limitations. The CVD technique requires ultrahigh vacuum conditions and high temperatures of more than 1000 °C in order to produce extremely stressed BN nanosheets, significantly increasing the cost of fabrication. Furthermore, Rivera et al. [34] and Aldalbahi et al. [35] observed that BN nanosheets fabricated from the PLPD technique demonstrated less than desirable properties during tests at temperatures greater than 200 °C, due to the significant diffusions of atoms at the interface of the substrate and BN nanosheet.

In this work, a low-cost drop-casting method is used to fabricate a planar PD with a BN/GO composite layer between two silver (Ag) electrodes. While the GO heterojunction PD and other reported work discussed above were focused specifically towards deep UV photodetection, the fabricated BN/GO composite PD in this work is tested for both UV and visible photoconduction. The broadband detection shows that the dependency of the PD on wavelengths and laser powers are in good agreement with the relatively thick BN/GO layer to absorb broadband wavelength photons. As such, the difficulty of photoconduction in the bulk GO layer is suppressed by the BN nanoparticles introduced into the GO bulk layer in order to improve the generation of electron-hole (*e-h*) pairs, and thus allowing photodetection over a broad wavelength range.

2. Experimental setup

2.1. Preparation of BN/GO solution

Fig. 1(a) shows the BN/GO composite layer on SiO_2/Si substrate preparation flowchart. A solution of BN and GO in a volume ratio of 0.2:1 was used as source material for converting the light signal into an electrical signal. The GO nanoparticles were obtained from the oxidation of bulk graphite using a modified Hummer's method [36,37]. For this process, a mixture 5 g of Mesh 7-graphite flakes and an oxidizing solvent encompassing 200 ml of 98% sulfuric acid (H_2SO_4) and 30 ml of 68% nitric acid (HNO_3) in an ice bath and stirred rigorously at 200 rpm for an hour to pre-oxidize the graphite flakes. The oxidation process is then enhanced by adding 30 g of potassium permanganate ($KMnO_4$) to the mixture, with the temperature being maintained at 40 °C for half an hour while stirring continues. Subsequently, 100 ml of 10% hydrogen peroxide (H_2O_2) was added to stop the oxidation process by neutralizing any excess active oxidizing species from the added $KMnO_4$. Finally, the solution was centrifuged for half an hour at 15000 rpm in order to obtain a supernatant. The BN solution on the other hand is prepared by sonicating a mixture of 200 mg of BN and 100 ml of deionized (DI) water for 8 h at 40 °C followed by centrifugation at 5000 rpm for half an hour. Any residue in the prepared solution was removed and a pure BN solution was separately mixed with the collected GO supernatant before being further sonicated for an additional of 2 h. The final solution containing BN to GO nanoparticles at a volume ratio of 0.2:1 is sonicated for 6 h at 40 °C before being ready for use.

A Si substrate consisting of a 1500 nm thick SiO_2 layer was obtained by thermal oxidation at a temperature range of 800–1200 °C to be used as a base for the deposition of the silver (Ag) source and drain electrodes. The cleansing process on the substrate surface was carried out using an ethanol solution followed by rinsing with DI water. After that, nitrogen was blown on the substrate to completely rinse off any ethanol stains that may have remained. Two Ag electrodes with a gap of 3 mm were deposited onto the substrate using an electron beam evaporation system, after which the SiO_2/Si substrate with the Ag electrodes are placed on a hotplate with the temperature set to a reducing rate of 1 °C/s from an initial temperature of 650 °C. When the temperature reaches approximately 550 °C, the prepared BN/GO composite solution of 1.0 µl was drop casted on the substrate to allow for fast drying and ensure strong adhesion of the material to the substrate and Ag electrodes. The area of the BN/GO composite layer was estimated to be around 9 mm^2.

2.2. Material characterization

The fabricated BN/GO composite based PD was analysed using FESEM, EDX and Raman scattering prior to photoconduction characterization. A Quanta 450 FESEM was used to obtain the morphological and elemental composition, while the intrinsic localized vibration properties of the sample were analysed using a Renishaw in-Via micro Raman Microscope at 532 nm green laser illumination. The current-voltage (*I-V*) characteristics of the fabricated BN/GO PD under laser/light illumination was characterized for DC bias voltages ranging from −10 to +10 V using a Keithley 2410 sourcemeter as shown in Fig. 1(b), with a red laser at 650 nm, UV laser at 405 nm and UV light emitting diode (LED) at 380 nm used as illumination sources. The photoresponsivity of the BN/GO PD was determined by a Yokogawa DLM2054 mixed signal oscilloscope (OSC) unit as shown in Fig. 1(c).

3. Results and discussion

3.1. FESEM and EDX

The morphology and composition of elements in the drop casted BN/GO layer on the SiO_2/Si substrate was collected using the FESEM. The images obtained as given in Fig. 2(a) shows the discrete distribution of BN nanoparticles on the surface of the GO composite layer. This was due to the ultrasonification process of the BN and GO mixture for 6 h which allowed for the formation of BN nanoparticles and its random distribution in the bulk GO layer. EDX scanning as shown in Fig. 2(b) shows B, N, O and C elements, which have weight percentage of 16.91, 22.05, 18.83 and 42.21 respectively as indicated by the EDX spectrum of Fig. 2(c). High counts per second (cps) on the C element followed by the O element indicates a bulk GO layer on the SiO_2/Si substrate, while the low cps values for the B and N elements show the discrete distribution of BN nanoparticles. No detection of the Si element also signified that the drop casted GO layer is dense and thick enough to disallow penetration of x-ray. These observations augur well with the FESEM image of Fig. 2 (a). But the cps value for B is very low compared with other elements due to high cps detection of C element which has overlapped the peak related to B element. Detailed information about the configuration of elements is presented in Table 1. It shows detection

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