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Self-powered Ag-nanowires-doped graphene/Si quantum dots/Si heterojunction photodetectors

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We report Ag-nanowires (Ag NWs)-doped graphene/p-type SiO_2-embedded Si quantum dots (p-SQDs:SiO_2)/n-Si heterojunction photodetectors (PDs). It is found that the p-n junctions show excellent PD characteristics including photocurrent/dark current (on/off) ratio of 10^5 at 0 V bias, meaning "self-powered". The PDs optimized at an Ag NWs concentration of 0.1 wt % exhibit 0.32–0.65 AW^{-1} responsivity (R), ~85% external quantum efficiency (EQE), and ~4.5×10^{12} cm $Hz^{1/2}$/W detectivity in the visible range of 500–900 nm. The linear dynamic range and response time of the PDs at 532 nm are ~83 dB and ~2 μs, respectively. The loss of the R is only 15% of its initial value while the PDs are kept for 700 h in air. In particular, the EQE of the self-powered PD is comparable to that of commercially-available Si PD and better than those of previously-reported graphene/Si PDs. These results suggest that the doped graphene/p-SQDs:SiO_2/n-Si heterojunctions are promising for their applications in self-powered optoelectronic devices.

1. Introduction

Broadband photodetectors (PDs) for the ultraviolet (UV)-infrared (IR) zone are widely used in various fields such as environment, biology, chemistry, imaging, and communication [1–4]. Graphene has been extensively used as a transparent conductive electrode (TCE) in optoelectronic devices because of its unique excellence in electron mobility, current carrying capacity, and transmittance [5]. Furthermore, the conductance and Fermi level of graphene can be sensitively controlled even by a small amount of extra charge carriers due to its unique band structure [6]. Recently, it has been shown that graphene well forms Schottky junctions with conventional semiconductors such as GaAs [7], GaN [8], and Si [9–27]. In particular, graphene/Si heterojunctions have received much attention in a variety of applications including PDs [9–23], solar cells [24,25], displacement meter [26], and chemical/biological sensors [27]. Among various kinds of graphene-based structures for PDs, graphene/Si heterojunctions are attractive due to their high responsivities over a broad spectral bandwidth near the visible region [9,14]. However, the interface between graphene and Si normally contains relatively-high density of surface states pinning the Fermi level, resulting in large leakage current, thereby limiting the overall performance of the graphene/Si PDs [9,12]. In addition, the photoresponse of the graphene/Si PDs is not fast enough for practical high-speed applications, which prompted a series of studies to lower the dark current and enhance the response speed [13–19]. For all of these graphene/Si PDs, the voltage (V_a) should be supplied. If photovoltaic cells with excellent performance are developed for PDs, they can be self-powered ($V_a = 0$ V). Recently, there have been some reports on graphene/Si-based self-powered PDs showing fast photoresponse in the UV to near IR range except the visible region [20–23]. Si quantum dots (SQDs) within SiO_2 matrix allow large light absorption in the visible range based on quantum confinement effect [14,24]. More recently, dark leakage current has been effectively suppressed in silver nanowires (Ag NWs)-doped graphene/p-SQDs/n-Si heterojunction solar cells, resulting in high external quantum efficiency (EQE) in the visible range under zero applied voltage [24]. If similar device structure is employed for self-powered PDs, the key performance parameters, especially including the photoresponse in the visible region, are expected to be greatly improved. Here, we report Ag NW-doped graphene/SQDs/Si heterojunction PDs operated under self-powered condition, showing maximally 10^5 photocurrent (PC)/dark current (DC) (on/off) ratio, 94.2% EQE, 4.5×10^{12} cm $Hz^{1/2}W^{-1}$ specific detectivity (D^*), and 83 dB linear dynamic range (LDR). Furthermore, the PDs lose only 15% of their original responsivity

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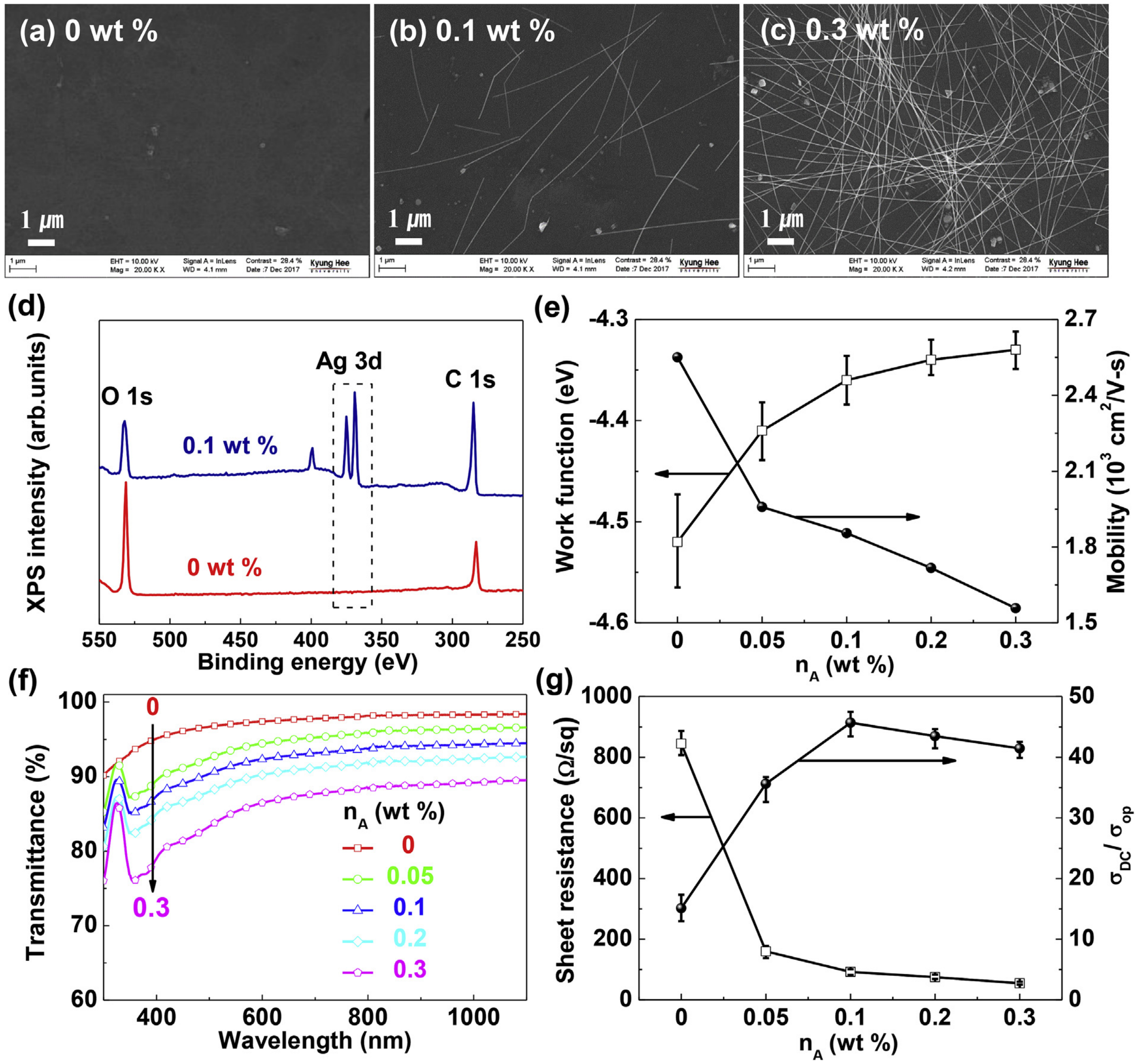

Fig. 1. FE-SEM images of (a) pristine and (b)–(c) doped graphene layers for $n_A = 0.1$ and 0.3 wt%. Here, the magnification is 20,000 times. (d) XPS spectra of doped graphene for $n_A = 0$ and 0.1 wt%. (e) Work function and electrical mobility of doped graphene as functions of n_A. (f) Transmittance spectra of doped-graphene for various n_A. (f) Sheet resistance and σ_{DC}/σ_{op} as functions of n_A.

values after 700 h, indicating excellent stabilities.

2. Experimental section

120-nm B-doped SiO_x films were grown on 6-inch n-type Si (100, 1-5 Ω-cm) wafers at room temperature using an Ar^+ beam with an ion energy of 750 eV and a n-Si target under oxygen atmosphere in a reactive ion beam sputtering system [24]. After deposition, the samples were annealed at 1100 °C in an ultra-pure nitrogen (99.999%) ambient by using a horizontal furnace to form p-type SQDs-embedded SiO_2 (*p*-SQDs:SiO_2) layers on n-type Si wafers. Single layer graphene films were grown on 70-μm-thick Cu foils (Wacopa, 99.8 purity) by chemical vapor deposition, and subsequently transferred onto the 120 nm *p*-SQDs:SiO_2/n-Si heterojunction substrates based on a conventional transfer method [24]. Ag NWs (purity: 99.5%) of 70 nm average diameter were purchased from ACS material. The Ag NWs powder was dissolved in isopropyl alcohol (98%, Sigma-Aldrich) to prepare Ag NWs solution, whose doping concentration (n_A) was varied from 0.05 to 0.3 wt%. The solution was dropped onto the substrates, coated at 1500 rpm for 1 min, and then dried on a hot plate at 100 °C for 2 min. Finally, Al (99.999%, Goodfellow) and InGa (99.99%, Aldrich) films were deposited on the top of graphene and the bottom of Si substrate as the electrodes.

The morphologies on the surface of the samples were analyzed by field emission scanning electron microscopy (FE-SEM). The atomic bonding states of the Ag NW-doped graphene were characterized by X-ray photoelectron spectroscopy (XPS) using an Al *Ka* line of 1486.6 eV. The SQDs in SiO_2 were characterized by high-resolution transmission electron microscopy (HRTEM) and photoluminescence (PL) spectroscopy. A 325 nm HeCd laser line was used as the excitation source for the PL. The transmittance, sheet resistance, work function, and electrical mobility of the doped-graphene TCEs were measured by UV–visible–NIR optical spectroscopy, 4 probe method, Kelvin probe force microscopy, and van der Pauw method, respectively. Current density-voltage (*J-V*) measurements to characterize the electrical behaviors of the PDs were carried out using a Keithley 2400 source meter controlled by a LabView