

All-carbon photodetectors

F. Wang^{1*}, Y. D. Liu¹, X. M. Wang^{2*}, E. Flahaut^{3,4}, Y. Li¹, X. Z. Wang⁵, X. R. Wang¹, Y. B. Xu¹, Y. Shi¹, R. Zhang¹

¹School of Electronic Science and Engineering and Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210093, China

²Department of Electrical Engineering, Yale University, New Haven, CT 06511, US

³Université de Toulouse; UPS, INP; Institut Carnot Cirimat; 118, route de Narbonne, F-31062 Toulouse cedex 9, France

⁴CNRS; Institut Carnot Cirimat; F-31062 Toulouse, France

⁵School of Chemistry and Chemical Engineering, Nanjing University, Nanjing, 210093, China

E-mail address: fwang@nju.edu.cn, xiaomu.wang@yale.edu

Abstract: We demonstrate a graphene/nanotube hybrid phototransistor, in which photogating provided by the nanotube layer leads to a dramatically enhanced photoresponsivity ($>100\text{A/W}$) in the visible range, corresponding to $\sim 10^4$ enhancement with respect to a graphene-only device.

OCIS codes: (040.5160) Photodetectors; (160.1890) Detector materials; (040.6070) Solid state detectors

1. Introduction

Due to ultra-broadband absorption and intrinsic high operation frequency [1-3], graphene based photodetectors have received considerable research attention in recent years [2-9]. However, the relatively low absorbance of a single sheet of carbon atoms adversely limits the photoresponsivity of graphene-based photodetectors to a level of $\sim 10^{-2}\text{A/W}$ [3]. A number of approaches have been proposed and demonstrated to enhance the device photoresponsivity [4-9]. However, enhancement in these device proposals either requires sophisticated fabrication steps that are not manufacturing scalable or is at the expense of the devices' optical bandwidth or response time.

Carbon nanotubes (CNTs) are the one-dimensional (1D) quantum-confined form of carbon allotropes, which also possess intriguing optoelectronic properties. For example, CNTs are effective light absorbers in a wide spectral range [10, 11]. In addition, free carriers in semiconducting tubes can have mobilities as high as $10^5\text{cm}^2\text{V}^{-1}\text{s}^{-1}$ [12]. The compatibility with solution processing further enables the deposition of nanotube thin-films onto flat and flexible optical substrates in a cost-effective manner [13]. The combination of 1D CNTs with 2D graphene represents an interesting and promising all-carbon composite material for optoelectronic applications. Recently, efficient charge transfer has been identified at junctions formed by graphene and CNTs [14]. This prompts us to investigate the graphene/nanotube composite materials for photodetector operation. In this paper, large-scale CVD grown graphene and liquid-processed CNT dispersions are used to fabricate a phototransistor with a graphene/nanotube hybrid channel. At visible wavelengths, i.e. $\sim 650\text{nm}$, the hybrid photodetector exhibits $\sim 10^4$ responsivity enhancement with respect to a graphene-only device. Our results demonstrate that graphene/nanotube composites have great potential in light harvesting and related optoelectronic applications.

2. Graphene/nanotube hybrid phototransistor fabrication

We use single-walled CNTs from a commercial supplier (Carbon Solutions Inc.). The graphene sample is grown by CVD method. Raman spectrum combined with optical microscope characterization points to a high quality single-layer sample. The phototransistor is fabricated as follows: CNT suspensions are produced by ultrasonically dispersing 2 mg nanotube in 20 mL NMP. The resulting suspensions are ultra-centrifuged with 10,000 g for 1h before the supernatant is collected for the deposition of CNTs thin films on a SiO_2/Si wafer. CVD graphene is transferred on top of the CNT layer using the standard PMMA supported procedures. Electrodes are patterned by standard photolithography. Different metal composition (Ti/Au and Pd/Au) are subsequently deposited. The electrical measurements are carried out in a cryogenic probe station under vacuum (10^{-6}Torr) at room temperature. Fig.1 (a) shows an optical micrograph of the device with the inset illustrating the underlying CNT thin film. Fig.1 (b) shows the Raman spectra of the top graphene layer. UV-vis-IR absorption of the underlying CNT film is shown in Fig.1(c).

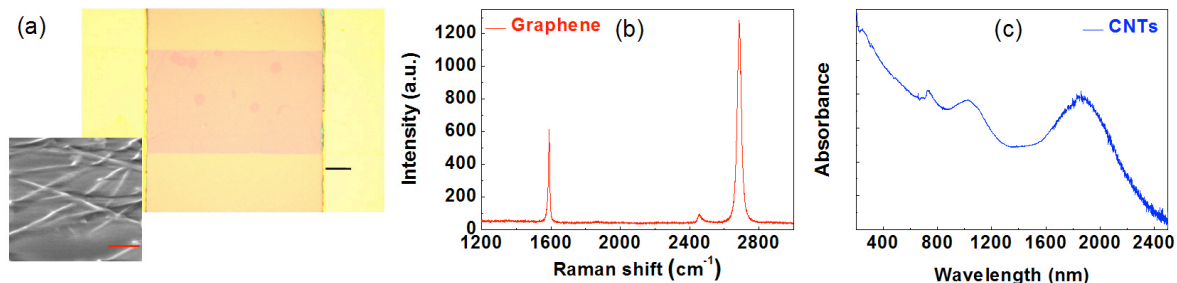


Fig.1 (a) Optical micrograph of an as-fabricated device (scale bar: $10\mu\text{m}$), Inset: SEM of CNTs (scale bar: 100nm), (b) Raman spectrum for the graphene top layer, (c) absorption curve of the CNT film.

3. Results for photoresponsivity enhancement

The photoresponse of the graphene/nanotube hybrid phototransistor is investigated under vacuum of 10^{-6} Torr at room temperature using a 650 nm visible diode laser. For reference, we have also fabricated devices with graphene-only and CNT-only channels. For the bare graphene channel device, a $0.17 \mu\text{A}$ photocurrent is measured under 200 μW illumination, while the CNT-only device exhibits no photoresponse due to the discontinued electrical pathway. Fig.2 (a) shows the transfer characteristics of a graphene/nanotube hybrid device with different incident power (at a fixed source-drain voltage of 0.5 V). The photocurrent as a function of the gate voltage is shown in Fig. 2(b). The maximum photocurrent of $93.7 \mu\text{A}$ is obtained at a gate voltage of -23V under 0.2 mW illumination. It should be noted that the photocurrent is much higher (~ 550 fold enhancement) than that of a graphene-only phototransistor ($0.17 \mu\text{A}$) under the same illumination conditions, highlighting the role of the underlying CNT film in enhancing the photoresponse. For the hybrid device, photocarriers generated in the graphene layer are not expected to contribute substantially to the photocurrent due to ultrafast carrier relaxation and recombination in graphene. The pronounced shift of the Dirac point under light illumination indicates the presence of photogating effect [6]. Fig. 2(c) illustrates the Dirac-point voltage shift of the device, which is characteristic of photogating effect in graphene phototransistors [6]. Therefore, we believe photocarriers are mostly generated in the CNT layer, with electrons subsequently transferred to graphene due to the internal electric field, caused by band alignment at the interface between graphene and nanotube layers. Investigation of the exact photoresponse mechanism of the hybrid system, through spatial photocurrent mapping, wide-wavelength photoresponsivity characterization is currently underway. As shown in Fig. 2(c), at relatively low incident power ($<10 \mu\text{W}$), the demonstrated all-carbon device exhibits photoresponsivity ($\sim 116 \text{ A/W}$) that drastically outperforms commercial Si detectors ($\sim 1 \text{ A/W}$).

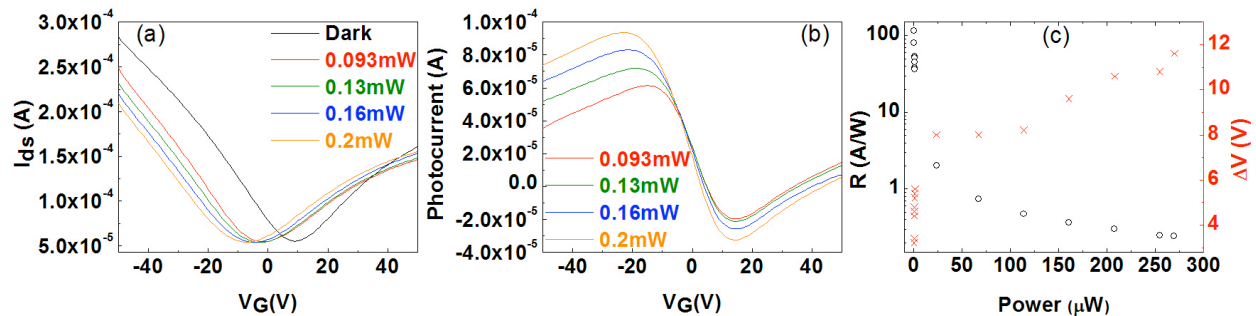


Fig.2 (a) Transfer curves and (b) photocurrent curves as a function of gate voltages, at different 650 nm incident power. (c) Responsivity and Dirac-point voltage shift (ΔV) as a function of incident power.

4. Conclusion

We have for the first time demonstrated an all-carbon phototransistor with a CVD graphene/nanotube hybrid channel. The underlying single-walled CNT layer is found to effectively photogate the graphene channel, providing significant ($\sim 10^4$) enhancement in photoresponsivity with respect to a graphene-only device. Such a graphene/nanotube composite photodetector is envisaged to be important for optical communication, spectroscopy and imaging applications, paving the way for the use of all-carbon composite nanomaterials in optoelectronics.

- [1] F. H. L. Koppens et al., "Photodetectors based on graphene, other two-dimensional materials and hybrid systems," *Nat. Nanotechnol.* **9**, 780 (2014)
- [2] F. Bonaccorso et al., "Graphene photonics and optoelectronics," *Nature Photonics* **4**, 611 (2010)
- [3] F. Xia et al., "Ultrafast graphene photodetector," *Nature Nanotech.*, **4**, 839 (2009).
- [4] T.J. Echtermeyer et al., "Strong plasmonic enhancement of photovoltage in graphene," *Nature Comm.* 2:458 (2011)
- [5] M. Furchi et al., "Microcavity-Integrated Graphene Photodetector," *Nano Lett.* **12**, 2773 (2012)
- [6] G. Konstantatos et al., "Hybrid graphene-quantum dot phototransistors with ultrahigh gain," *Nature Nanotechnol.* **7**, 363 (2012)
- [7] X. Wang et al., "High-responsivity graphene/silicon-heterostructure waveguide photodetectors," *Nature Photon.* **7**, 888 (2013)
- [8] Y. Zhang et al., "Broadband high photoresponse from pure monolayer graphene photodetector," *Nature Comm.* 4:1811 (2013)
- [9] C. O. Kim et al., "High photoresponsivity in an all-graphene p-n vertical junction photodetector," *Nature Comm.* 5:3249 (2014)
- [10] H. Kataura et al., "Optical Properties of Single-Wall Carbon Nanotubes," *Synthetic Metals* **103**, 2555 (1999)
- [11] F. Wang et al., "The Optical Resonances in Carbon Nanotubes Arise from Excitons," *Science* **308**, 838 (2005)
- [12] T. Durkop et al., "Extraordinary mobility in semiconducting carbon nanotubes," *Nano Lett.* **4**, 35 (2004)
- [13] T. Hasan et al., "Nanotube polymer composites for ultrafast photonics," *Adv. Mater.* **21**, 3874, (2009)
- [14] G. L. C. Paulus et al., "Charge Transfer at Junctions of a Single Layer of Graphene and a Metallic Single Walled Carbon Nanotube," *Small* **9**, 1954 (2013)