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Sensors and Actuators A: Physical

journal homepage: www.elsevier.com/locate/sna

Nanostructured graphene–Schottky junction low-bias radiation sensors $^{\scriptscriptstyle{\updownarrow}}$



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ARTICLE INFO

Article history: Received 31 October 2014 Received in revised form 10 April 2015 Accepted 10 April 2015 Available online 10 June 2015

Keywords: Radiation sensor Graphene Gamma-radiation Schottky diode Low-bias Nanosensor

ABSTRACT

We present a key idea of using the graphene-based Schottky junction to achieve high sensitivity and wide detection range radiation sensors. Nanostructured Schottky junction is formed at the interface between a graphene, metal electrode, and a semiconductor. The current flowing through the junction is mainly controlled by the barrier's height and width. Therefore, the detection principle is based on Schottky barrier height (SBH) modulation in response to different materials and stimuli. We have illustrated the concept for gamma (γ) radiation sensors. It's demonstrated that the integration of graphene leads to a great enhancement in sensitivity of up to 11 times coupled with 5 times increase in the sensing range as compared to conventional Schottky junctions. Furthermore, it was demonstrated that for proposed sensors, that the change in SBH could be fairly linearized as a function in the radiation dose unlike the SBH of comparable conventional junctions. The new concept opens the door for a novel class of minitiuarized, low biased, nanoscale radiation sensors for wireless sensor networks. The devices are based on new nanostructured Schottky junctions made by growing graphene on ultrathin platinum catalytic layer grown on different silicon substrates. Graphene high uniformity film with small flakes size embedded with platinum particles was synthesized using two deposition steps. The integration of graphene layers on regular M–S junctions was only possible by using an ALD grown platinum thin film (10–40 nm) and then growing graphene in PECVD at temperatures lower than platinum silicide formation temperature. The radiation sensing behaviors were investigated using two different substrate types. The first substrate type is a moderately doped *n*-type $(n \approx 2 \times 10^{15} \text{ cm}^{-3})$ silicon substrate in which a Schottky rectifier response with different threshold voltages was observed. A device that is based on Pt/n-Si conventional Schottky junction was used as a reference. The various devices were exposed to a range of γ -irradiations (2–120 kGy) using Co⁶⁰ source, and a change in terminal voltages before and after radiation were measured accordingly. A sensitivity of 3.259 µA/kGy cm² at 1 V bias over a wide detection range has been realized. The charge transport mechanisms are interpreted on the basis of testing the detectors at elevated temperatures and theoretical models, both of which both verified tunneling as the dominant charge transport in the device. Tunneling allowed the operation of the detectors at low bias voltages with good sensitivity. The detector's realized sensitivity at low bias voltage is a significant advantage, allowing the sensor to operate on a small battery or an energy-harvesting source. This is ideal for low-cost wireless sensor networks.

The obtained responses, increase in sensitivity, and increase in detection range, were explained by studying the band diagrams of the graphene–Schottky junction in comparison to that of the conventional junction. Further, the fact that graphene layer was grown on the M–S junction adds to the uniqueness of this research since exfoliated graphene will result in increased contact resistance and lower carrier mobility which might not yield the desired sensing response.

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http://dx.doi.org/10.1016/j.sna.2015.04.031 0924-4247/© 2015 Elsevier B.V. All rights reserved.

1. Introduction

In the aftermath of the devastating nuclear disaster at the Fukushima reactor facilities in Japan, the development of high-

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performance ultrasensitive radiation sensor technologies has become paramount. There is a critical need for devices that will actively detect all forms of radiation species (i.e., alpha, beta, gamma, X-ray, and neutrons) and provide continuous monitoring of radioactive material. Moreover, the American National Academy of Engineering has listed the ability to detect nuclear materials at a distance as one of the five major obstacles to preventing nuclear terror [1]. Thus, not only would the implementation of these devices near nuclear facilities be especially advantageous, but it would also be beneficial to install them at national borders to counteract the increased threat of radiological or nuclear terrorist attacks. To accomplish this, the detectors must be small and reliable enough to discriminate among the various types of radiological sources. They would be implemented in portable devices and sensor networks that could be deployed over large geographical areas vulnerable to nuclear radiation or cross-border weapons smuggling.

Detecting small doses of radiation in the environment is critical for communities living close to nuclear plants or in the event of nuclear disasters with International Nuclear Event Scale (INES) of 5 or more, e.g., Fukushima (2011), Chernobyl (1986), and Three Mile Island reactor (1979). With more than 498 power reactors currently operating or under construction in 30 countries [2], accompanied by the unsolved problem of post-process storage of nuclear wastes and reliable controlling of potential environmentally induced leakages, there is an urgent need for relatively cheap and simple to used sensor of a wide range of radiation doses [3,4].

There are three main types of radiation detectors that are commonly used, each having its own method to detect the radiation particles. They are divided into gas-based detectors [5-9] which use a gas that is electrically biased to collect ionized particles, which are a result of the interaction between the radiation particle and the fill gas, semiconductor detectors [10-14] which rely on a reverse biased diode junction to create electron hole pairs when the depletion region is excited by incoming radiation particles, and scintillator detectors [15-22] which interact with incoming radiation particles and, as a result, release light photons that are detected using an optical transducer. Nanostructured materials on MEMS integrated devices could replace most current conventional radiation sensors, the majority of which rely mainly on lattice defects in single crystal silicon structures that are induced by irradiation [23]. These defects are detected through resistance or capacitance changes. The current MEMS based radiation techniques, however, have substantial drawbacks: (1) limited sensitivity. (2) Limited detection range (efficient at high doses of 60 kGy or higher). (3) Limited efficacy (one time use), and (4) High probability of error.Increasing surface to volume ratio and increasing material's selectivity could potentially overcome the limited detection range and low sensitivity of conventional MEMS based capacitive radiation sensors.

P-n structured diodes and Schottky diode-based radiation sensors are gaining much attention since the 1990s, in particular for biomedical and imaging applications [24,25]. However, they normally operate at high reverse voltage (500–1500 V) such that the space charge region is wide enough to increase the generation/recombination currents.Therefore, the problem of leakage current should be overcome [26].

Integration of nanomaterials with solid-state radiation sensors is undertaken in order to enhance sensitivity and improve device efficiency and capacity [27–31]. More importantly the devices can operate at low bias and therefore increase the possibility of operating these detectors on small batteries or energy harvesting sources, which is not possible with the current state-of-the-art detectors. However, along with the ability to scale down devices come challenges with designing and developing nanostructures that must operate properly in an increasingly smaller environment. Schottky diode can replace conventional PN junctions and bipolar transistors in the scaled down solid-state sensors due to their fast response and low threshold voltages. Schottky diode based sensor generally consists of a metal-semiconductor (M–S) junction. The most common choices of metals are catalytic type noble metals (e.g., Au, Pt, and Pd) [32–35]. Schottky diodes have been implemented for developing highly efficient sensors, solar cells and electrochromic devices [36–40]. Graphene ultrathin sheets are emerging as ideal candidates for thin-film devices and combination with other semiconductor materials such as silicon. They have been produced in the form of ultrathin sheets consisting of one or a few atomic layers directly grown by chemical vapor deposition (CVD) [41-43] or by solution processing [42,45] and then transferred to various substrates.

In this paper we have implemented a direct, simple, reusable, isotropic, wide range, and ultrahigh sensitive radiation sensor. The sensor is based on a nanostructured array of graphene grown on platinum/n-Si substrate. The new sensor structure (Fig. 1) is a Schottky barrier diode based on forming an M–S junction of *n*-type silicon substrate and ultrathin film platinum integrated with graphene layer. The high carrier mobility in graphene and the ultrathin platinum film facilitate generating a strong forward current in response to γ -irradiations. The results show that this new structure gives a signal that is an order of magnitude higher than a conventional Schottky barrier diode exposed to the same radiation doses.

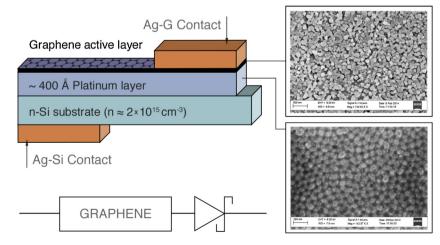


Fig. 1. Schematic diagram of the working principle of the graphene based Schottky junction sensor showing SEM images of the top view of the graphene and platinum thin films.

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