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Non-destructive photovoltaic reading of interface type memristors using graphene as transparent electrode

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The memristor device based on graphene/Nb-doped $SrTiO_3$ heterojunction was fabricated. The device exhibits obvious resistive switching between high resistance state and low resistance state. By using the graphene as the transparent electrode, remarkable photocurrent and photovoltage was achieved in the heterojunction. The photovoltage shows a clear dependence on the resistance state, whereas the photocurrent keeps unchanged. Based on these features, a new non-destructive photovoltaic reading method of the memristor was demonstrated. The photovoltaic reading method is free of external driving voltage, thereby avoiding the perturbation of the stored data. Moreover, the photovoltage output signal instead of the current is more compatible with the IC technologies.

1. Introduction

The memristor based on the resistive switching effect is wildly considered as one of the most promising devices for the next generation nonvolatile information storage and neural network simulation [1–6]. As a two terminal memory device, the memristor can be triggered to high resistance state (HRS) or low resistance state (LRS) by large writing voltages with certain polarities [7]. The resistance state of a memristor was usually read by measuring the current through it under a small reading voltage [8]. However, data reading is still a challenge for the commercial application of memristor. Long time repeated reading of the memristor under the reading voltage may change the resistance leading to the degradation of the stored data. To solve this problem, a refreshment circuit or a compensation circuit was usually used in the memristor, which inevitably increased the complexity of the device structure [9–11]. Therefore, new non-destructive readable memristors free of reading voltages are needed to enhance the reliability of the device.

Among the various types of memristor, the interface type memristor based on a Schottky contact between metals and semiconductors has been intensively studied due to its outstanding performance. It was demonstrated that the resistance state of the interface type memristors can be continuously tuned without the initial forming process. Moreover, the high writing/reading speed, long retention time and good scaling down possibility of the interface type memristor were also reported in previous studies [12,13]. In the studies of the interface type memristors, the photovoltaic effect is a useful tool for the monitoring of the device parameters. Shang et al. studied the photocurrent and photovoltage of the interface type memristor between Au and Nb doped $SrTiO_3$ (NSTO) at different resistance states, and found the inhomogenous nature of the resistive switching at the interface [14,15]. By measuring the photovoltage of the interface type memristors, it is possible to detect the resistance states without applying the reading voltage, thereby avoiding the perturbation of the stored data. However, the conventional metal electrodes have low optical transmittance, which limits the photovoltaic efficiency.

Since its discovery, graphene was widely used as the transparent electrode in solar cells, photo-detectors as well as LEDs owing to its excellent optical and electrical properties [16–19]. The photovoltaic

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effect was widely reported in the graphene contacts to n-type semiconductors. Besides, graphene is also an efficient probe for the detection of the switching mechanism of the memristor [20]. Yang et al. reported a three terminal switch device with a large on/off ratio based on the graphene/Si Schottky junction [21]. By using graphene as the electrode, Kwon et al. observed the bipolar switching in copper oxide and demonstrated the oxygen ion migration switching mechanism [22]. In this paper, we report a NSTO based interface type memristor using few layer graphene as the transparent electrode. The device exhibits bipolar resistive switching effect without the forming process. Under illumination, remarkable photovoltaic effect was achieved in the device. The open-circuit photovoltage is dependent on the resistance state, and keeps stable under constant illumination without obvious degradation. The results provide a non-destructive photovoltaic reading method for the memristors, which is more compatible with the MOSFET in the IC technology.

2. Materials and methods

The structure of the memristor device is schematically illustrated in Fig. 1a. One side polished (001) oriented STO single crystals (15 mm × 15 mm) doped with 0.7 wt% Nb were used as substrate. Prior to the device fabrication, the NSTO substrates were cleaned in acetone, ethanol and deionized water respectively, and then annealed at 800 °C in oxygen for 5 h to obtain a defect free atomically flat surface. According to the previous works, the surface is terminated with TiO_2 layer [23]. After the substrate treatment, a stripe of polytetrafluoroethylene (PTFE) film, 5 mm in width and 10 μm in thickness, was stuck on the NSTO surface at 200 °C as an insulating layer. This insulating layer is to prevent the direct vertical tunneling of electrons from the top metal electrode to the NSTO through graphene since the graphene sheet is extremely thin.

The polymethylmethacrylate (PMMA) coated freestanding few layer graphene sheets were purchased from Vigon Material Corporation, which were grown by the conventional chemical vapor deposition method. The device was fabricated by transferring a 10 mm × 10 mm graphene sheet on the substrate across the NSTO-PTFE border in deionized water. During this step the graphene/NSTO junction with an area of ~70 mm^2 was obtained. Then the PMMA was dissolved in acetone. Subsequently, the device was dried in air at 80 °C for 1 h to achieve a close contact. Finally, a silver paint Ag top electrode was spread onto the graphene on the PTFE film and an Al wire was bonded to the NSTO substrate as the Ohmic back electrode.

Raman spectrum of the device surface was measured by a Renishaw inVia Raman System with a 514.5 nm laser. The optical images were obtained by an optical metallographic microscope. The surface morphology was measured by a SII E-Sweep scanning probe microscopy. A Keithley 2400 sourcemeter connecting the top and back electrodes of the device was used to apply the writing/reading voltage and record the reading signal. In order to study the photovoltaic effect, a violet light beam was focused on the graphene/NSTO junction by an LED with the central wavelength of 395 nm. The light power was fixed as 700 μW during the illumination.

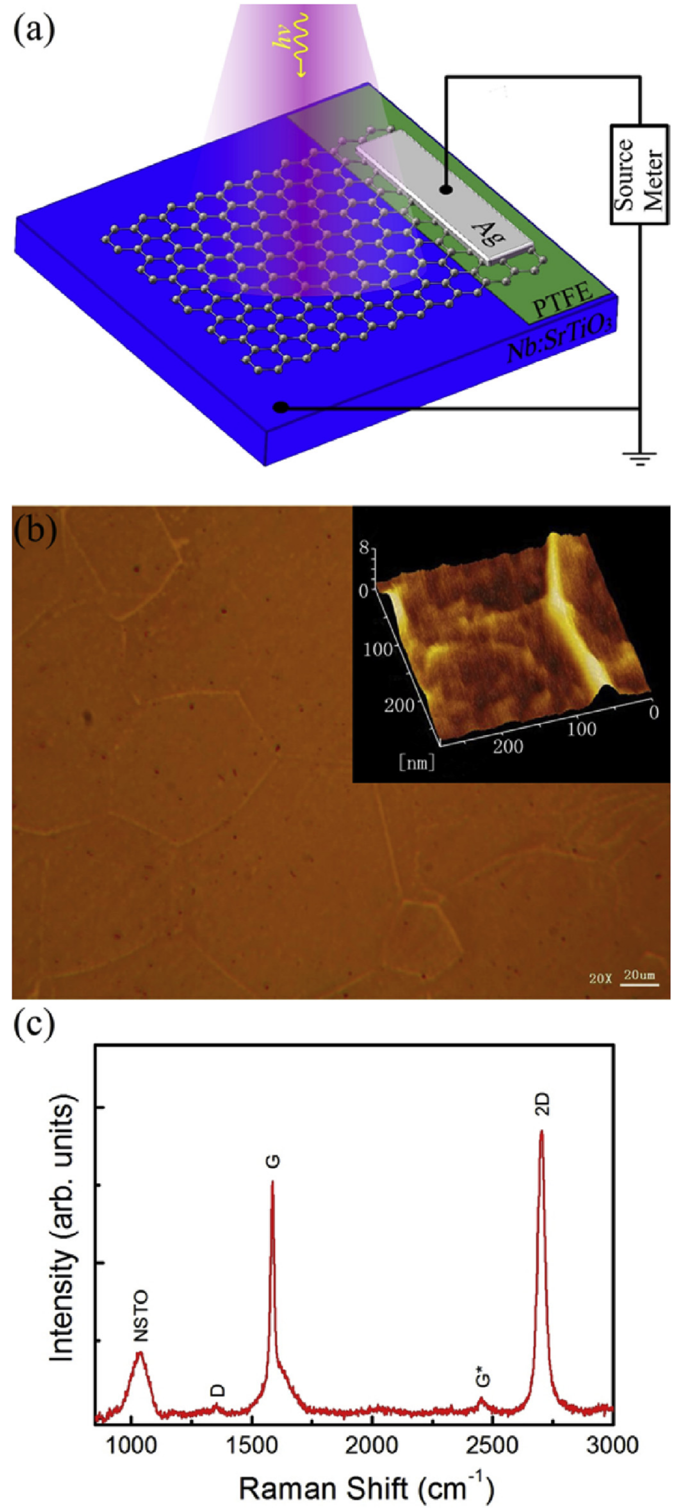

Fig. 1. (a) Schematic illustration of the device structure and measurement configuration. (b) Optical microscope image of the graphene on NSTO substrate. The inset is the AFM topography of the graphene surface near the wrinkle. (c) The Raman spectrum of the graphene/NSTO surface under 514.5 nm laser.

3. Results and discussion

The optical microscope image of the graphene sheet on NSTO is shown in Fig. 1b. It can be observed that the graphene sheet covers the NSTO substrate continuously without obvious holes. Most of the graphene surface is flat indicating good contact to the substrate. Some wrinkles exist on the surface as reported in previous works [16]. The inset of Fig. 1b shows the AFM topography image near the wrinkle. One can see that the graphene sheet is continuous at the wrinkle and no cracks can be found, which ensures the good connection of the whole graphene electrode. As plotted in Fig. 1c the Raman spectrum of the sample shows clear peaks from the graphene and NSTO substrate [24,25]. The intensity ratio between