

Nitrogen plasma-treated multilayer graphene-based field effect transistor fabrication and electronic characteristics



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ABSTRACT

Chemical doping with hetero-atoms is an effective method used to change the characteristics of materials. Nitrogen doping technology plays a critical role in regulating the electronic properties of graphene. Nitrogen plasma treatment was used in this work to dope nitrogen atoms to modulate multilayer graphene electrical properties. The measured I-V multilayer graphene-base field-effect transistor characteristics (GFETs) showed a V-shaped transfer curve with the hole and electron region separated from the measured current-voltage (I-V) minimum. GFETs fabricated with multilayer graphene from chemical vapor deposition (CVD) exhibited p-type behavior because of oxygen adsorption. After using different nitrogen plasma treatment times, the minimum in I-V characteristic shifted into the negative gate voltage region with increased nitrogen concentration and the GFET channel became an n-type semiconductor. GFETs could be easily fabricated using this method with potential for various applications. The GFET transfer characteristics could be tuned precisely by adjusting the nitrogen plasma treatment time.

1. Introduction

Considerable relevant graphene researches have been conducted in recent years. The two-dimensional sp²-hybridized carbon form shows significant characteristics [1], such as high carrier mobility [2], high sensitivity [3], optical transparency [4], thermal stability and excellent mechanical strength [5]. Graphene has been widely studied in many research fields, including lithium batteries [6], gas sensors [7,8], electrochemical electrodes and p-n junction diodes [9,10]. Graphene-based electronic devices are very attractive for the next generation nano-electronics because of their unique physical properties. Graphene-based field effect transistors (GFETs) are considered a promising device for nano-electronics [11–13]. In general, the graphene sample exhibits p-type behavior in a nitrogen atmosphere. The p-doping formation was due to the presence of water vapor and oxygen in the atmosphere [14,15]. In a transistor channel or semiconductor element application the n-channel FET operating speed is faster than the p-channel speed [16]. N-type graphene semiconductors have a wider range of applications. Therefore, group IV elements used as

dopants in graphene serve as electron donors and hole traps, resulting in p-type transformation into n-type conduction [17]. Nitrogen atoms are considered an ideal element for carbon doping because of its five valence electrons and suitable atomic size [18]. It was reported that graphene could be converted into n-type semiconductors using chemical vapor deposition (CVD), arc-discharge approaches, segregation, solvothermal methods, and plasma treatment [19–22]. Compared with these techniques plasma treatment is viewed as an easy surface modification method to introduce atoms [23]. Nitrogen plasma allows controlling the nitrogen content in the graphene structure. We developed GFETs motivated by the enhanced ionic binding capability based on N-doped multilayer graphene using a simple plasma process. When nitrogen atoms are doped into graphene, it usually expresses three types of C–N bonding: pyridinic-N, pyrrolic-N, and graphitic-N. The pyridinic-N bands with two C atoms at the graphene defects and provides one p electron to the π system [19,24]. The pyrrolic-N bands form a five-membered ring that provides two p electrons to the π system [19,24]. The graphitic-N refers to C atoms substituted for N atoms in the hexagonal ring in the graphene layers [24]. The pyridinic-

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N and graphitic-N are sp^2 -hybridized into the graphene and the pyrrolic-N is sp^3 -hybridized into the graphene. Jin et al. reported using pyridine as the carbon and nitrogen source to synthesize N-doped graphene using CVD. The results showed that the C–N bonding in the graphene occurs mainly in the pyridinic-N form and exhibits n-type material characteristics [23]. Gao et al. showed that the pentagon N (pyrrolic-N) can introduce strong electron donor states near the Fermi energy level and the as prepared N-doped graphene shows n-type behavior [25]. Pyrrolic-N bonding is most easily generated by plasma processing in graphene doped with nitrogen. The graphene nitrogen content is increased when the nitrogen plasma doping duration is increased [26]. Pyrrolic-N is the major factor resulting in p-type transformation into n-type conduction in N-doped graphene.

Multilayer graphene was directly grown onto Cu foils in this study using thermal CVD. The nitrogen plasma treatment was used to introduce nitrogen atoms into the graphene. The plasma treatment duration was the parameter used to control the multilayer graphene nitrogen content. In the semiconductor materials analysis we confirm whether the multilayer graphene is p- or n-type by measuring the minimum in current-voltage (I-V) GFET characteristics. Generally, the nitrogen content increased with the increase in nitrogen plasma treatment time. The pyrrolic-N content in the sample would obviously increase with the XPS results in this work.

2. Experimental

Cu foil (10 mm×5 mm) was used as the substrate. The Cu foil was placed into a quartz tube with a 50.8 mm diameter. the quartz tube was then placed horizontally inside a furnace. Fig. 1 shows the GFET formation procedure. The Cu foil was annealed with an Ar (200 sccm) and H₂ (4.5 sccm) mixture at 1000 °C for 60 min. After annealing, CH₄ was introduced into the quartz tube at a flow rate of 10 sccm for 5 min under atmospheric pressure. At atmospheric pressure, there were more gas molecules to increase the deposition rate of graphene layer. Therefore, the graphene synthesized was a multilayer structure. The

Cu foil was etched using FeCl₃ solution to define the FET channel. The multilayer graphene was transferred onto SiO₂/Si substrate, followed by baking at 95 °C for 180 min. The multilayer graphene/SiO₂/Si sample was subsequently treated with nitrogen plasma in a vacuum chamber. A 7-layer graphene was obtained under the described experimental conditions. The result was very stable. According to the plasma treatment principle, the experimental design could confirm that the nitrogen ions directly bombarded onto the graphene surface. The nitrogen plasma was produced using an RF generator with 20 W at a pressure of 2×10^{-2} Torr, and the treatment duration was set as a parameter for controlling the nitrogen content [24]. Au (100 nm) was deposited as electrodes using ion sputtering. The multilayer graphene/SiO₂/Si was subsequently transferred into another vacuum chamber to define the FET channel size using oxygen plasma etching. The oxygen plasma was produced using an RF generator with 50 W at a pressure of 2×10^{-2} Torr for 60 min. The gate oxide layer (500 nm of SiO₂) was deposited using e-beam evaporation at a deposition rate of about 0.2 Å/s. The gate electrode (100 nm of Au) was deposited by sputtering at a deposition rate of about 3 Å/s. An additional component manufacturing process was required for the top gated structure. This was one of the reasons back gated structure development was decided. However, excessive leakage current was observed in the back gate device characteristic measurements. Therefore, top gate multilayer graphene FETs were fabricated and studied [27].

The sample structure was observed using transmission electron microscopy (TEM). Raman spectroscopy was used with a laser excitation energy of 2.41 eV (514.5 nm) to examine the multilayer graphene micro-structure. X-ray photoelectron spectroscopy (XPS) was used to study the composition. The binding energy data was calibrated with the C 1s signal at 284.6 eV before the XPS measurements. By fitting the XPS spectra, the N 1s peak could be divided into three C–N bonding types: pyridinic-N, pyrrolic-N, and graphitic-N which corresponded to the XPS results in the work. The GFET I-V characteristics were measured using a source meter for the minimum in I-V characteristics in ambient atmosphere. The GFET sample channel size has a total

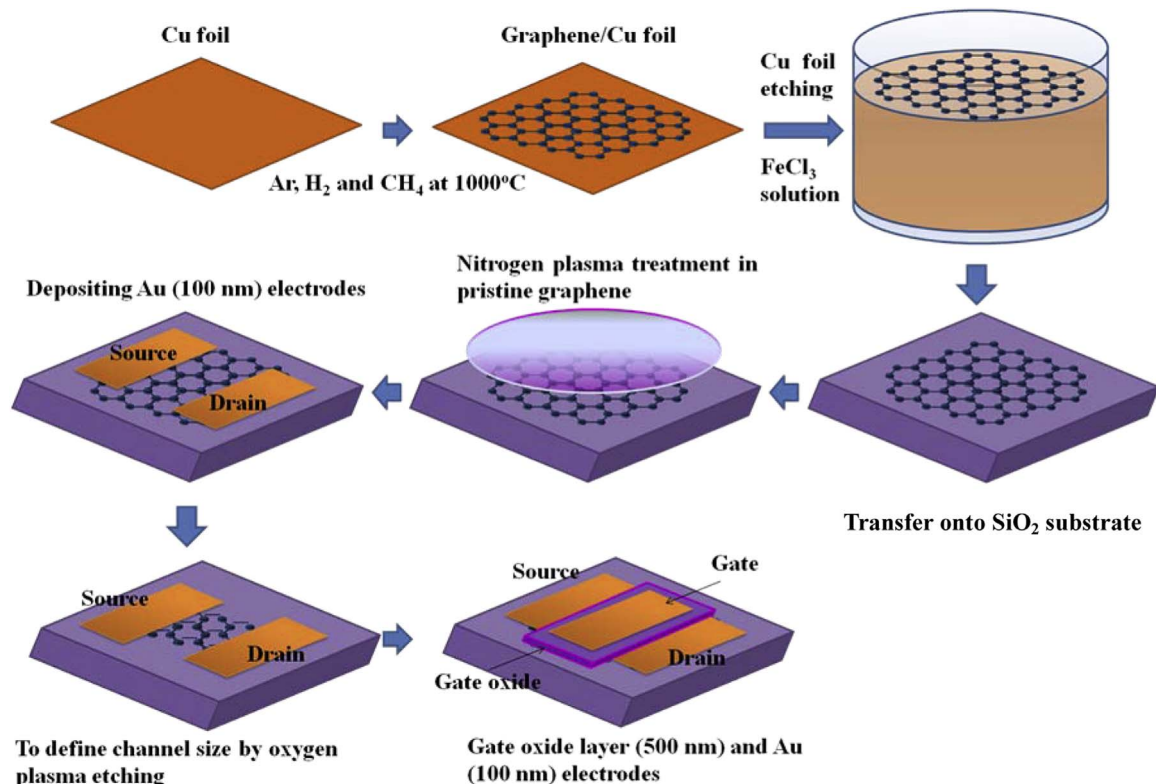


Fig. 1. Illustration of GFETs fabricated on SiO₂ substrate.

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