Mechanical fabrication of graphene devices using focused-ion beam: deposition and milling

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ABSTRACT

Graphene, due to its material stability, mechanical strength, unique band structure and excellent electrical properties, holds great promise for future nanoscale electronic devices. Here, we report a novel mechanical fast-prototyping method to fabricate graphene-based electronic structures on few-layer graphene (FLG) using focused-ion beam (FIB). FIB is a versatile tool and FIB-assisted deposition and milling processes feature mask-free patterning, quick turnaround time as well as high precision. Specifically, in this study, the parameters of FIB are well controlled. The platinum wire, which is induced from the gaseous precursors by the fine focused gallium ions of low energy, is conveniently deposited on the substrate to connect FLG. The measured channel resistance of around 50 kΩ indicates good electrical contact between platinum wire and FLG. With the computer controlled alignment and patterning, the accuracy of deposition and milling can be as high as 15-20 nm. For the milling process, 2-D ribbon structures can be easily produced by milling the patterned area, followed by an annealing process. Raman spectra are used to examine the quality of graphene after the FIB and annealing processes.

Keywords: graphene, nanoribbon, focused ion beam, depositon, milling

1 INTRODUCTION

Graphene, a single layer of carbon atoms arranged into a two-dimensional honeycomb lattice, has rapidly received significant attention since its discovery in 2004 by Novoselov and co-workers [1]. It holds great promise for the next-generation electronics due to its unique electrical properities, such as massless Dirac quasiparticles[2], high carrier mobilities and capacities [1-6]. To date, several different experimental methods have been developed to produce single layer (or few layers) graphene (SLG or FLG), including mechanical exfoliation of highly oriented pyrolytic graphite [1], thermal decomposition of SiC wafer under ultrahigh vacuum (UHV) conditions or CVD growth on metal (e.g. Ru, Ni, Cu) substrates [7–8], and chemical [9–11] or thermal reduction [12] of graphite oxide (GO). The success in fabricating single layer graphene has stimulated the extensive research efforts in graphene related research area.

The ultimate goal to use graphene in next-generation electronics is to realize all-graphene circuit with functional devices built from graphene layers or graphene nanoribbons (GNRs) [13, 14]. Single layer graphene is a semimetal without bandgap. As a result of quantum confinement and edge effects, graphene nanoribbons, several-nm wide stripes of graphene, exhibittunable bandgap [4], which is more desirable in electronics. However, the application of graphene nanoribbons is limited by their availability. Although they have been made by chemical [15], sonochemical [16] and lithographyic [17,18] methods, they are particularly sensitive to surface contaminants, including resist residues left from lithographical and chemical processes, modifing the local properties and providing extra scattering sites.

Here in this paper, we propose a novel mechanical fast-prototyping method to fabricate full-graphene electronics with focused-ion beam (FIB). Basically, there are two indispensable processes involved in the fabrication of such devices. At first, single layer graphene (or graphene layers) are patterned to be a basic functional part, such as a single memory cell. Then the patterned graphene devices can be connected with outer circuit to realize certain functions, such as a computer memory. Correspondingly, the FIB technique is utilized to pattern graphene and deposit electrodes, using milling and deposition functions, respectively. An annealing process is followed to improve the quality of graphene and the patterns. We believe that the methods proposed here will have great impact on the development of full-graphene electronics.

2 MATERIALS AND METHODS

Graphene sheets were made by mechanical exfoliation of bulk graphite (Kish Graphite, purchased from Graphene Supermarket) with scotch tape and randomly deposited onto a 285 nm SiO_2 layer on a silicon wafer (P/B(100), 1-10 Ω-cm purchased from universitywafer.com). An optical microscope and a camera (Nikon MM-40 and DXM 1200) were then used to identify and locate individual graphene sheets. Then graphene samples were transferred to a dual beam system (FEI Quanta 3D 200i) for the fabrication

process and characterization of the sample: an ion beam for electrode deposition and an electron beam for imaging. The region of interest on the sample was positioned at the eucentric point where the two beams cross, as shown in Figure 1a. At this location, the electron beam could provide *in situ* imagining of the graphene. The sample was tilted at 52 °for optimized ion incident angle.

2.1 Electrodes Deposition

During the process of electrode deposition, the FIB uses a highly focused Ga^+ ion beam to scan over the desired areas of the sample surface. The gaseous Pt metalorganic precursor (methylcyclopentadienyltrimethyl platinum) is introduced during the ion beam scanning, providing the well-controlled ion-assisted deposition of the metal material. Therefore, the Pt lines can be fabricated directly over the desired area of a graphene with approximately 20 nm precision. In the deposition process, to prevent the graphene from being damaged by the Ga^+ ions, two rectangular electrodes (200 μm × 10 μm × 100 nm) were deposited on two sides of graphene under relatively small ion energy (16.0 kV acceleration voltage and 4.0 pA ion current). Figure 1b shows a microscopic image of the fabricated electrodes on both sides of a graphene sheet.

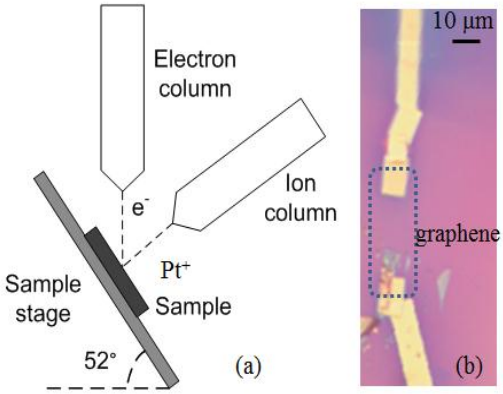

Figure 1: a) the eucentric location of the ion beam; b) the image of fabricated electrodes on two siede of graphene.

2.2 Graphene patterning

During FIB milling process, Ga^+ ions interact with the electrons and nuclei of the target atoms. If the energy transferred to the nuclei exceeds the displacement energy, the atoms will be dislodged from their sites. Since it has been demonstrated that the radiation of ions can damage the graphene seriously, [19] we used a small ions energy, 2 keV, to conduct the milling process. To study the effect of ions milling, rectangular trenches 5 × 5 μm^2 were milled using different emission current and deposition time. Based on extensive expereiments with various parameters, we found that 2 keV acceleration voltage and 0.7 pA beam current produce the best results. The dose in ions cm^{-2} was calculated according to equation (1) for a target ion current (I) of singly charged Ga ions in pA, exposure time (t) in s, and pattern area (A) in μm^2:

$$dose = \frac{I_{ion} \times t_{exposure}}{A_{pattern} \times 1.602 \times 10^{-15}} \qquad (1)$$

The dose used in the expereiments under 15 seconds, 30 seconds and 40 seconds, are 2.6×10^{14}, 3.5×10^{14} and 5.2×10^{14} ions cm^{-2}, respectively.

2.3 Annealing Process

Annealing has been proven effective to repair the defects and damage in graphene sheets [20] leading to the recrystallizaton of damaged graphene sheets. Therefore, we annealed the graphene patterns fabricated from focused ion beam technique. Graphene sheets were annealed in argon gas environment for 12 hours at 700 ℃ in a furnace (Neytech Vacuum Muffle Furnace, 110-120 VAC).

3 RESULTS AND DISCUSSION

3.1 Deposited Electrodes

A semiconductor analyzer (Agilent Technologies B1500A) was used to test the resistance of the Pt-graphene-Pt device. The *I-V* curve, as shown in Figure 2, shows that the resistance of the device is about 50 kΩ, demonstrating a good connection between platinum electrodes and graphene. Multiple experiments are done for different graphene layers, and the results are in good consistency.

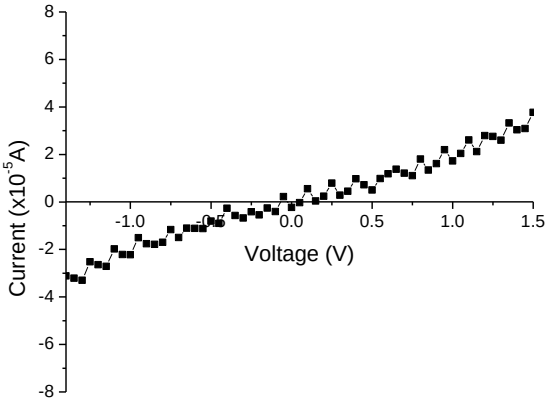

Figure 2: *I-V* curve of a typical Pt-graphene-Pt device.

3.2 2-D Graphene patterns

Typical graphene patterning processes for FLG are shown in Figure 3. FLG was first observed under optical microscope (Figure 3 (a)). Milling time periods set for patterns 1, 2 and 3, are 30 s, 15 s and 40 s, respectively, while all the patterns are milled under 2 keV acceleration voltage and 0.7 pA beam current. After the milling process, we can only observe a little difference on the milled patterns, (Figure 3(b)). After that the FLG was annealed in argon gas environment at 700 ℃ for 12 hours. Interestingly, the edge of milled area became sharper. Furthermore, for different milling parameters, even though the defined square areas are the same before milling, the resulted area sizes are different, (Figure 3(c)). More specifically, area 2 matches best with its defined area, while area 1 and 3 are enlarged to some extent. This suggests that 15 second is an optimized milling time for 2 keV voltage and 0.7 beam current. To study the property of FLG after the milling process, the Raman spectra of several different spots were examined, as shown in Figure 3(d). The flat lines 1, 2 and 3 indicate that there is nothing left at the milled area. The clear defect-induced Raman bands, D (1350 cm^{-1}) was observed after the milling process. This may be caused by the damage on the sample during the scanning of SEM and FIB.

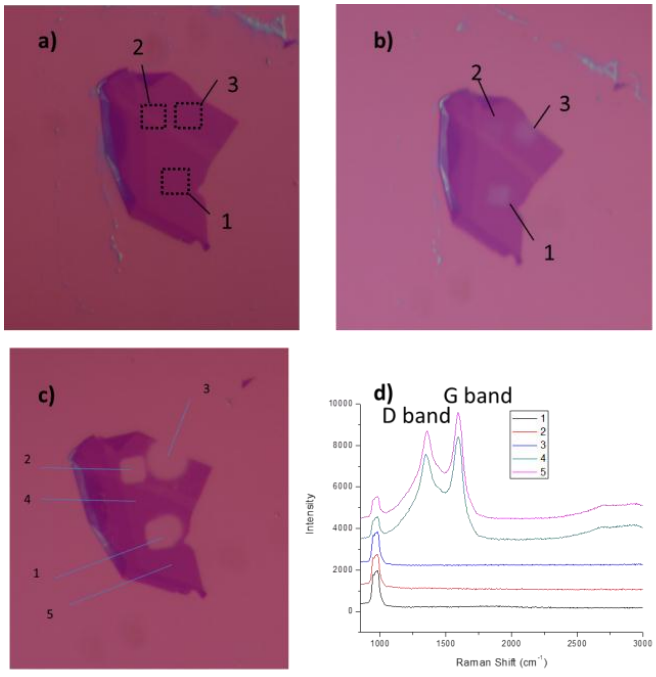

Figure 3: a) Few layer graphene (FLG) before FIB milling; b) FLG after FIB milling using different exposure time for pattern 1, 2 and 3; c) FLG after annealing process; d) the Raman spectra of FLG at spot 1-5 after annealing. Spectra are offset for clarity.

3.3 Graphene Nanoribbon

Based on the experiments in the last section, we chose 2keV acceleration voltage, 0.7 pA beam current and 15 second milling time as the parameters for creating graphene nanoribbons. The procedures are similar to those for graphene patterning, as shown in Figure 3. Since the resolution for milling process of FIB is around 15-20 nm, we can get nanoribbon by accurately control the position of two milling patterns (case 1, Figure 3), or the distance between the edge of graphene and milling patterns (case 2, Figure 4). And the graphene nanoribbons we created in the second case are shown in Figure 5. The widths of two nanoribbons were 800 nm and 300 nm, respectively, as determined from AFM. By comparing the Raman spectra of the two cases, we found the D band in case 2 are much lower than that in case 1, which indicate that the damage in case 2 is much smaller. There might be two reasons for the difference. First, the graphene in case 2 is a single layer graphene, which has much less defect than Few layer graphene. Second, the graphene in case 2 has shorter exposure time to SEM and FIB during the whole process, because less patterns were milled.

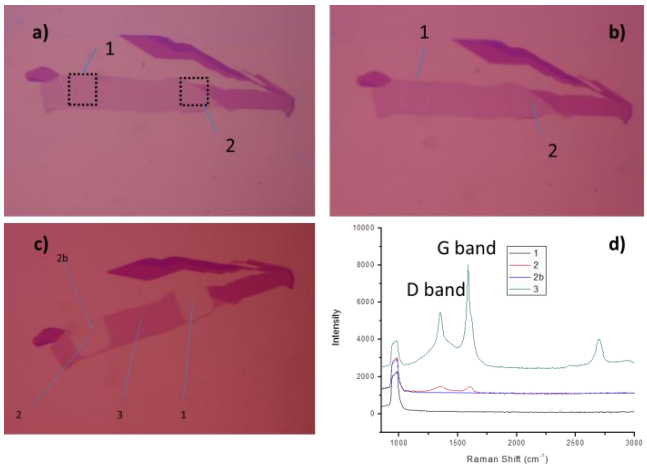

Figure 4: a) Single layer graphene (SLG) before FIB milling; b) SLG after FIB milling using different parameters for pattern 1and 2; c) SLG after annealing process; d) the Raman spectra of FLG at spot 1-3 after annealing.

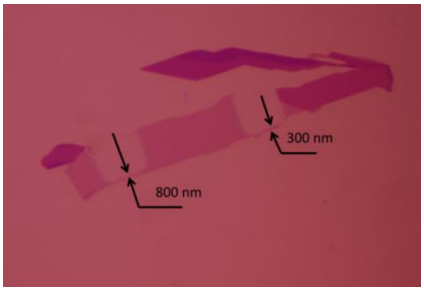

Figure 5: Nanoribbons created with focused ion beam.

4 CONCLUSION

In this paper, we demonstrated a new technology, FIB, for fabricating graphene devices. More specifically, we deposited platinum electrodes, patterned graphene and graphene nanoribbons using a focused ion beam system. An annealing process was used to clean the milled patterns and repair graphene defects. The method proposed here is low-cost, and easy to conduct. Future experiments will be focused on the optimization of the processes in order to achieve damage/defect-free fabrication. Two methods will be approached for the optimization. The first one involves minimum exposure of graphene to electrons and ions during the scanning of SEM and FIB. At the same time, higher resolution can be achieved on the size, location and height/depth. The second one is to completely remove any possible defects or damage during milling.

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