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Fabrication of 10-nm-Scale Nanoconstrictions in graphene using AFM-Based Local Anodic Oxidation Lithography

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Abstract

We performed local anodic oxidation (LAO) lithography of highly oriented pyrolytic graphite (HOPG) and monolayer graphene using atomic force microscopy (AFM). Auger electron spectroscopy (AES) measurements in the oxidized area formed on the HOPG revealed that the number of oxygen atoms systematically increased with the bias voltage applied to the AFM cantilever $|V_{tip}|$, which demonstrates the tunability of the extent of oxidation with $|V_{tip}|$. By utilizing the LAO lithography technique, we fabricated monolayer graphene nanoconstrictions with a channel width as low as 10 nm, which was the smallest graphene nanoconstriction achieved so far using LAO lithography.

1. Introduction

Graphene has attracted considerable attention as a candidate material for next-generation nanoelectronics owing to its unique band structure and exceptionally high charge carrier mobility. Thus, the fabrication of graphene nanostructures is a key process element for studying graphene-based nanoelectronics devices. To date, fabrication of graphene nanostructures has typically been performed by using electron beam lithography followed by oxygen plasma etching. However, the resulting rough edges induced by oxygen plasma etching have posed challenges in the fabrication of nanostructured graphene.

In this regard, local anodic oxidation (LAO) lithography based on atomic force microscopy (AFM) provides an alternative method for fabricating graphene nanostructures. Because AFM tips can directly contact graphene flakes, AFM lithography enables high-resolution nanolithography of graphene flakes. Thus far, electrochemical etching [1, 2] and oxidation [3-5] of graphene flakes have been demonstrated by employing AFM-based LAO lithography techniques. By electrochemically etching, graphene nanoribbons [1] and Aharanov–Bohm rings [2] have been fabricated. Fabrication of graphene/graphene oxide/graphene (metal/semiconductor/metal) in-plane junctions [3], single-electron transistors [4], and graphene nanoconstrictions [5] have been demonstrated by electrochemical oxidation. These results demonstrate the effectiveness of AFM lithography as a method for fabricating graphene nanostructures; thus, further investigation of the AFM lithography techniques is desired for the advancement of nanofabrication and the application of graphene-based nanoelectronics devices.

2. Auger electron spectroscopy measurements

In order to study the chemical properties of the oxidized area, we oxidized the surface of the HOPG and performed an Auger electron spectroscopy (AES) analysis of the oxidized area. HOPG was chosen as a base material rather than monolayer graphene on SiO_2 because the AES analysis of the oxidized area of monolayer graphene on a conventional SiO_2/Si substrate could not be performed owing to the strong background signal from the oxygen atoms residing in the SiO_2 layer. We oxidized 4×4 μm^2 square regions on the HOPG by scanning the AFM cantilever in a zigzag path with a 100-nm pitch for various bias voltages of $|V_{tip}|$ = 12, 11, 10, 9, 8, 7, and 6 V with the scanning speed of the AFM cantilever of v = 100 nm/s. The resulting oxidized area was observed by measuring the frictional force image as shown in the inset of Fig. 1(b), in which the dark (bright) areas correspond to a large (small) frictional force and to the oxidized (intact) graphite surface. After oxidizing the surface of the HOPG, the sample was transferred to an AES system (JEOL JAMP 7830F) with a Schottky field emission electron source. Auger electron spectra were collected with an accelerating voltage of 10 keV.

Figure 1(a) shows the Auger electron spectra as a function of the kinetic energy for the samples oxidized with $|V_{tip}|$ = 12, 11, 10, 9, 8, 7, 6, and 0 V (top to bottom). When oxidation was performed with $|V_{tip}|$ = 12 V, a discernible amplitude in the Auger electron spectrum for the O-KLL transition was detected [top curve in Fig. 1(a)]. This observation indicated that the oxidized area contained oxygen atoms bonded to the surface. In Fig. 1(b), we show the intensity of the O-KLL peaks $I_{O\text{-}KLL}$ as a function of $|V_{tip}|$. The value of $I_{O\text{-}KLL}$ was obtained by integrating Auger electron spectrum with respect to kinetic energy from 470 to 520 eV while linear backgrounds were subtracted. When the value of $|V_{tip}|$ increased from $|V_{tip}|$ = 6 V, $I_{O\text{-}KLL}$ increased with $|V_{tip}|$ and saturated for $|V_{tip}|$ > 9 V. This observation indicates that the number of oxygen atoms bonded to the graphite surface increased with $|V_{tip}|$ for 6 V < $|V_{tip}|$ < 8 V and saturated for 9V < $|V_{tip}|$.

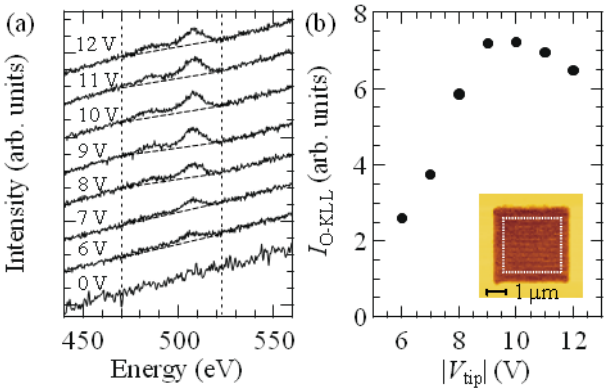

Fig. 1 (a) Auger electron spectra of the oxidized areas for varying $|V_{tip}|$ plotted as a function of the kinetic energy. O-KLL peaks were located at a kinetic energy of 510 eV. (b) Intensity of the O-KLL peaks $I_{O\text{-}KLL}$ plotted as a function of $|V_{tip}|$. (Inset) Friction image of an oxidized area of HOPG after LAO at V_{tip} = -12 V.

4. Fabrication of nanoconstrictions

We fabricated graphene nanoconstrictions by utilizing LAO lithography. In order to fabricate graphene nanoconstrictions, monolayer graphene on SiO_2 was prepared by mechanical exfoliation. LAO was conducted from one edge of graphene flake toward central region with $|V_{tip}|$ = 10 V and v = 50 nm/s. When the AFM tip reached the flake center, $|V_{tip}|$ was turned off and the scanning speed was set to 200 nm/s. After a short period, the bias voltage was resumed to $|V_{tip}|$ = 10 V and the scanning speed were set to v = 50 nm/s. Thus nanoconstrictions were carved out in the central region of graphene flake as shown in Fig. 2(a). We fabricated graphene nanoconstrictions with W = 40, 30, and 10 nm. The width of our smallest graphene nanoconstriction (W = 10 nm) was smaller than the smallest channel width achieved so far by using LAO lithography (W = 25 nm [2]) by a factor of 2.5.

In order to confirm the presence of the confinement effect in the graphene nanoconstriction devices, we measured the two-terminal differential conductance by applying a small alternating voltage V_{ac}. The application of a back-gate bias voltage V_g allowed for tuning of the Fermi energy of the graphene layers. Fig. 2(b) shows the two-terminal conductance G as a function of V_g measured at 4.2 K for the devices with W = 10 nm. A region of suppressed conductance G < 20 μS, i.e., a transport gap, was observed [indicated by the vertical dashed lines in Fig. 2(b)]. The emergence of zeros conductance demonstrated the opening of a transport gap in the graphene nanoconstriction devices due to lateral confinement of the charge carriers. In addition, these observations demonstrated that LAO lithography based on AFM is capable of producing graphene constrictions with a resolution better than 10 nm that function as graphene nanoelectronic devices. The high spatial resolution of the AFM lithography technique and the tunability of W with nanometer-scale precision demonstrate that AFM-based LAO is a promising method for the nanofabrication of graphene.

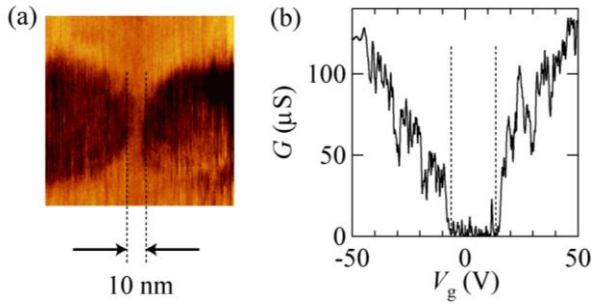

Fig. 2 (a) Friction images of the quantum point contact devices with channel widths of W = 10 nm. (b) Differential conductance G for W = 10 nm as a function of the back-gate bias voltage V_g measured at 4.2K.

5. Conclusions

We performed LAO lithography of HOPG and monolayer graphene surfaces using AFM. The AES measurements in the oxidized area of the HOPG showed that the number of oxygen atoms systematically increased with $|V_{tip}|$. By utilizing an AFM lithography technique, we fabricated graphene nanoconstrictions with W = 10 nm, which was the smallest nanoconstriction achieved that utilized an LAO lithography technique. These observations indicated that AFM lithography allows for fabrication of ultra-small graphene nanostructures with features less than several tens of nanometers, which demonstrates that AFM lithography is a promising nanofabrication technique for nano structured graphene devices.

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