Atomic force microscope tip-induced local tunable oxidation of graphene

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Chemical functionalization of graphene is an attractive method for opening a transport gap in graphene because this method provides wide-range tunability of the transport gap. In this work, we report on a method for oxidizing graphene in nanometer scale region with variable extent of functionalization by using atomic force microscope (AFM) tip-induced local anodic oxidation (LAO) lithography.

The schematic setup for the LAO lithography is shown in Fig. 1. We used the contact-mode AFM in the enclosure box. The relative humidity was maintained at 75%. A negative bias voltage $V_{\text{tip}}$ was applied to the conducting silicon cantilever with graphene and silicon substrate grounded in order to oxidize graphene. Fig. 2 shows the width of oxidized area $w$ as a function of the scanning speed of AFM cantilever $v_s$ from 5 to 200 nm/s and $V_{\text{tip}}$ from 0 V to -9.0 V. The value of $w$ depends systematically on $v_s$ and $V_{\text{tip}}$. The minimum value of $w$ was as small as 18 nm.

We fabricated graphene/graphene oxide/graphene (G/GO/G) junctions and conducted the transport measurements at $T = 4.2$ K. The current-voltage characteristic of the G/GO/G junction fabricated with the sufficiently large bias voltage $V_{\text{tip}} = -8$ V exhibits strong nonlinearity, which is qualitatively consistent with the results in metal/graphene oxide/metal junction. The size of the transport gap derived from the width of plateau in current-voltage curve is 2 eV. The size of transport gap in the junction is independent of the width of oxidized area $w$. The current-voltage characteristics are explained by the thermionic emission over Schottky barriers at G/GO interface.

When $V_{\text{tip}}$ is increased from -8 V to -5.5 V so that the moderate oxidation takes place, the conductance of G/GO/G junction increases from 0.4 $\mu$S to 136 $\mu$S. The size of transport gap decreases from 2 eV to 0.1 meV. This result indicates that the transport gap of graphene/graphene oxide/graphene junction changes depending on the extent of oxidation in GO.

Thus the transport gap of G/GO/G junctions can be tuned by using LAO lithography using AFM. Since AFM lithography provides extremely fine oxidized patterns in graphene, one can form various sizes of transport gaps in graphene in nanometer scale region by using this technique. The results presented above indicate that LAO lithography using AFM is an attractive method for fabricating graphene-based nanoelectronic devices.

References
Figure 1 Schematic setup for the local anodic oxidation of graphene. A graphene flake is deposited on a SiO$_2$/Si substrate and is contacted by the metal electrodes. A negative bias voltage ($V_{\text{tip}}$) is applied to the conductive silicon cantilever with the graphene sheet and silicon substrate grounded.

Figure 2 The width of oxidized area $w$ as a function of (a) the bias voltage applied to AFM tip $V_{\text{tip}}$ for $v_s = 50$ nm/s and (b) the scanning speed of the AFM tip $v_s$ for $V_{\text{tip}} = -8$ V.