Hybrid graphene nanoribbon-nanopore devices for biolomolecule detection and DNA sequencing

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Abstract

We present a study of hybrid graphene nanoribbons-nanopore devices[1] for biomolecule detection and ultimately DNA sequencing. When a graphene nanoribbon is constricted to nm sizes, the variation of the potential created by the different bases of the DNA strand passing through the adjacent nanopore will create a variation of the ribbon conductivity, enabling an electrical discrimination between DNA bases.
We realized devices (Figure 1) comprised of nanopores with diameters in the range of 2−10 nm at the edge or in the center of graphene nanoribbons (GNRs), with widths between 5nm and 200 nm, on 40 nm thick silicon nitride (SiNx) membranes.

We discuss the challenges encountered in the manufacturing of these nanoconstrictions (by lithograph or by electron beam sculpting) and the irradiation effects of the electron beam during the nanopore formation. GNR conductance is monitored in situ during electron irradiation-induced nanopore formation inside a transmission electron microscope (TEM) operating at 200 kV.

We identify and study a linear and a supralinear regime for the increase of GNR resistance with the electron dose(Figure 2a), and correlate with the decrease by a factor of ten or more in mobility(Figure 3b) when GNRs are imaged at relatively high magnification with a broad beam prior to making a nanopore.

Based on our findings we devise a scanning TEM procedure in which the position of the converged electron beam can be controlled with high spatial precision via automated feedback and we are able to quantify GNR electron induced damage (Figure 2b). This method minimizes the exposure of the GNRs to the beam before and during nanopore formation. A statistic of the resistances of the GNR-NP devices obtained by the TEM and STEM method(Figure 3a) shows that the TEM method severely damage the ribbons (increase in resistance on average 15 times), while the resistance of the STEM drilled ribbons remains virtually unchanged during the process. The resulting STEM GNRs can sustain microampere currents at low voltages (~50 mV) in buffered electrolyte solution and exhibit high sensitivity and mobility, similar to pristine GNRs without nanopores(figure 3b).

We finally present the operation of this sensor for biomolecule detection and DNA sequencing, correlating the electric signal measured in the GNR to the ionic current measured through the nanopore. The higher current(~uA) which can be driven through a GNR compared to the ionic current(~nA)[2] enable us to obtain a hundredfold increase in the measuring speed, making possible DNA sequencing without slowing the molecules, for a projected 10 minutes gull genome sequencing.

References

[1] Towards sensitive graphene nanoribbon-nanopore devices by preventing electron beam induced damage. M. Puster*, J. A. Rodríguez- Manzo*, A. Balan*, M. Drndić. ACS Nano, **7(12)** 2013 pp 11283-11289 , *equal contribution
[2] Differentiation of short, single-stranded DNA homopolymers in solid-state nanopores. K. Venta, G.Shemer, M. Puster, J. A. Rodríguez- Manzo, A. Balan, J. K. Rosenstein, K. Shepard, M. Drndić. ACS Nano, **7(5)** 2013 pp 4629-4636.

Figures

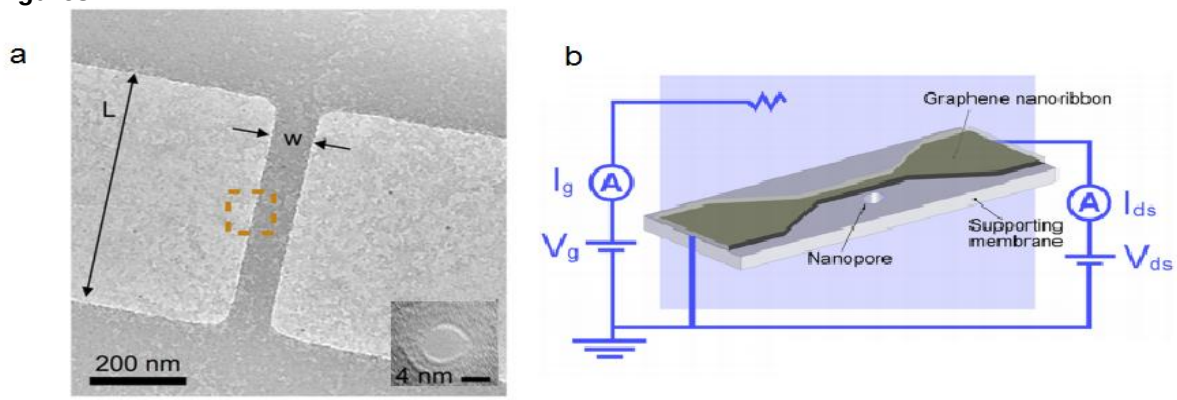

Figure 1 : a) TEM images of GNR devices. The dark gray areas are graphene covered with a 15 nm thick layer of hydrogen silsesquioxane (HSQ). Light gray areas are the bare 40 nm thick supporting silicon nitride (SiNx) membrane. Inset: Nanopore formed in the center of the GNR b) Schematic showing the GNR-NP device and the circuit diagram used for electrolytic gating in KCl solution.

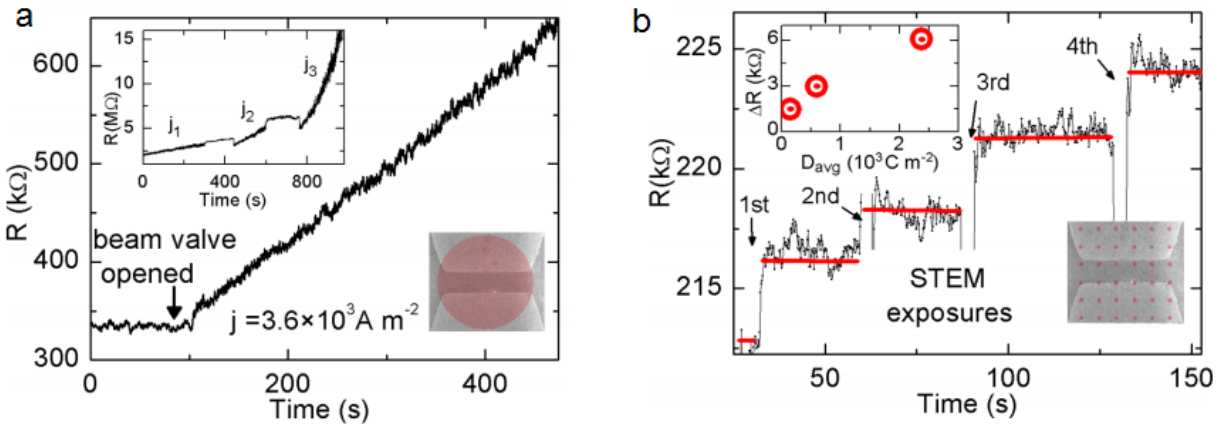

Figure 2 : a) In situ TEM electrical measurement of GNR resistance vs. time for broad beam TEM imaging showing a linear increase. Top-left inset : the rate of change of resistance increases with current density (j1, j2, and j3 are 3, 9, and 23 × 10^4 A m^{-2}, respectively). Bottom-right inset: illustration of a GNR exposed to a broad beam (red circle) in TEM imaging mode. b) In situ STEM electrical measurement of GNR resistance vs. time for converged beam STEM imaging. GNR resistance increases in a step-like fashion after each 330 ms scan in between the four steps, indicated by arrows. Top-left inset: average increase of resistance (ΔR) per STEM scan exposure as a function of average dose (Davg). Bottom-right inset: illustration of the STEM scan over a GNR.

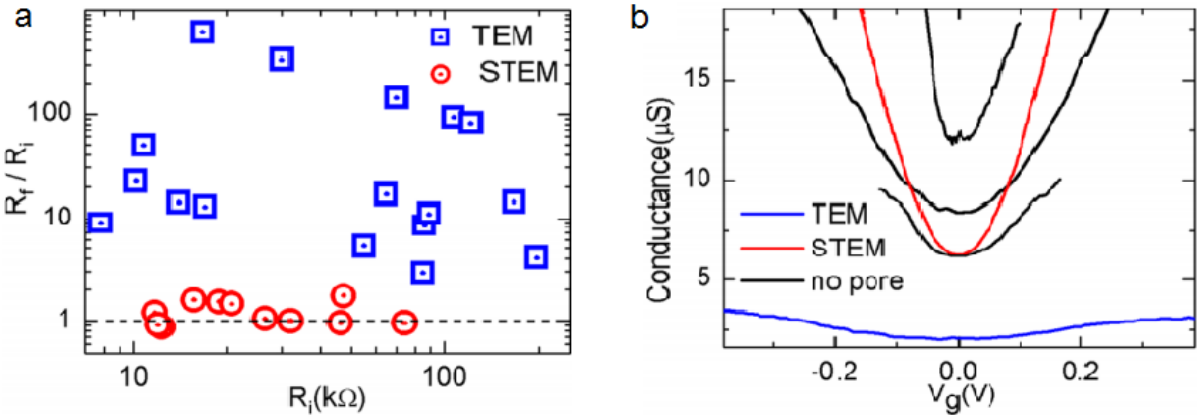

Figure 3. Comparison of GNR electrical properties after TEM and STEM nanopore formation methods. (a) Relative increase in resistance before (Ri) and after (Rf) nanopore formation for 28 GNR-NP devices made with a TEM method (17, blue squares) and STEM method (11, red circles), as a function of initial resistance Ri. (b) GNR conductance vs. gate voltage (Vg) measured in 1M KCl solution for representative devices before (black curves) and after nanopore formation with TEM (blue) and STEM (red) methods. For clarity, these curves were shifted so that the charge neutrality point is at Vg = 0 V.