Wafer-scale fabrication of solution-gated graphene field-effect transistors for biosensing applications

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Abstract

Graphene is a 2-dimensional material with a honeycomb structure formed by sigma bonds between three fourths of its valence electrons, the remaining fourth forming a huge conjugated electronic π system. Its transport properties are therefore extremely sensitive to the charge environment or to electric fields in its vicinity. Graphene field-effect transistors (GFETs) take advantage of this fact which, together with the well-known density of states in the shape of Dirac cones close to the valence and conduction band edges, allows GFETs to be operated in n-, p- or ambipolar channel mode by shifting the Fermi level with the simple application of the appropriate gate voltage. Combining such electronic properties with a high chemical stability in biological and chemical solutions, graphene is a promising material for biosensing applications [1]. In order to detect specific biomarkers graphene surface must be functionalized with recognition biomolecules, such as antibodies [1]. For graphene-based devices to be usable in real applications it is necessary to process them at the wafer-scale. However, the integration of graphene with microelectronic devices has proven to be a difficult task due to poor adhesion of graphene to its insulating substrate after the transfer process using a temporary polymeric substrate, normally PMMA. This sets limits to the microfabrication processes that can be used to pattern devices on transferred graphene. The transfer of graphene on top of pre-patterned contacts, without need for further process, simplifies the fabrication. Graphene produced over large areas by chemical vapor deposition (CVD) shows a high crystalline quality and almost 100% surface coverage which allows it to be used as an electric insulation layer in aqueous environment. This makes it possible to operate GFETs in solution-gated configuration (SG-GFET), more suitable for biosensing than the traditional back-gated configuration due to a lower operational voltage [1, 2]. More important, the transistor transfer curve is highly sensitive to the liquid gate electrolytic properties, such as ionic strength, pH or any other parameter that changes the electrical double layer that forms at the solution-graphene interface.

Device Fabrication

In this work, a 200 mm silicon wafer with 200 nm of thermal oxide and 3 nm of chromium as adhesion layer was covered with 30 nm of gold. Using optical lithography and ion milling, the wafer was patterned with 126 devices (figure 1), each with source and drain contacts separated by a gap between 12.5 and 50 µm. An insulating layer of 320 nm of aluminium oxide was deposited on top of the contacts using a lift-off technique, leaving only 10 µm uncovered at the extremity of the contacts.

Copper foil of 25 µm thickness and 99.999% purity was cut into pieces with ≈20 mm side. These substrates were loaded into a quartz tube and heated to 1020 °C under low pressure argon flux. Methane was introduced into the chamber, thermally decomposing to produce monolayer graphene on the copper catalyst. After deposition, the conventional copper dissolution process using PMMA as a temporary substrate was applied to the samples. The floating graphene/PMMA samples were then transferred onto different areas of the prepared wafer, such that graphene entirely covers the area of source and drain contacts, but does not short-circuit the source and drain pads. The PMMA temporary substrate was removed using acetone. The quality of graphene was assessed using Raman spectroscopy. The finished set of devices was characterized electrically at wafer-scale without further process.

Device Characterization

Phosphate buffered saline (PBS, pH 7.4, 150 mM) solution was used in the measurements. A platinum (Pt) wire was used to gate the transistor through the solution. The experiments were conducted by dipping a known volume of PBS onto the graphene transistor sensing area.

Figure 2a shows the characteristic output curve at different gate voltages (V_G = -0.2, 0 and 0.2 V). A linear behavior is observed, which is indicative of ohmic contacts between graphene and the contacts underneath.

The transfer curves of the devices show that the graphene is unintentionally p-doped. The p-doping is related to the process and the substrate (Figure 2b). Figure 2b shows that the transfer curve changes with the ionic strength of the PBS (1X = PBS 150 mM) in such a way that the minimum conductivity point is shifted to lower V_G when the ionic strength increases. This is consistent with the shorter Debye length in solutions with high ionic strength [1].

G.M.J thanks CNPq for a PhD grant. N.V is thankful to FAPESP for a post-doctoral grant.

References

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Figures

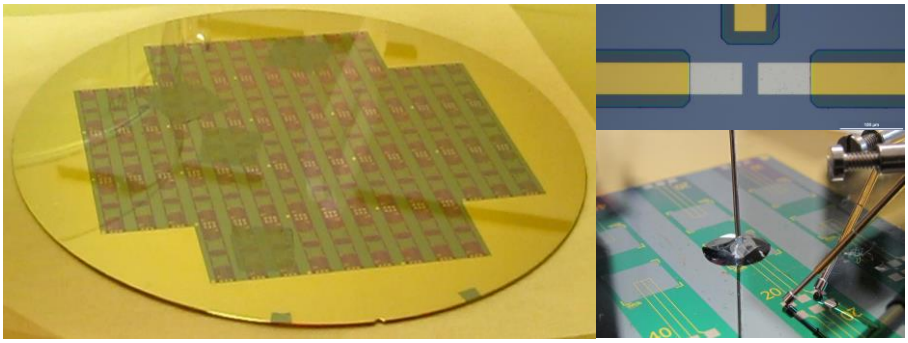

Figure 1. (left) Picture of the microfabricated 200 mm wafer with 130 sets of contacts for graphene devices and several graphene/PMMA pieces transferred. (top-right) Picture of the source and drain contacts (left and right respectively) in a device with a gap of 20 µm during the fabrication process (before graphene transfer) and an accessory insulated voltage line (not used in this work). (bottom-right) Picture of a device during a measurement with a platinum wire as a gate.

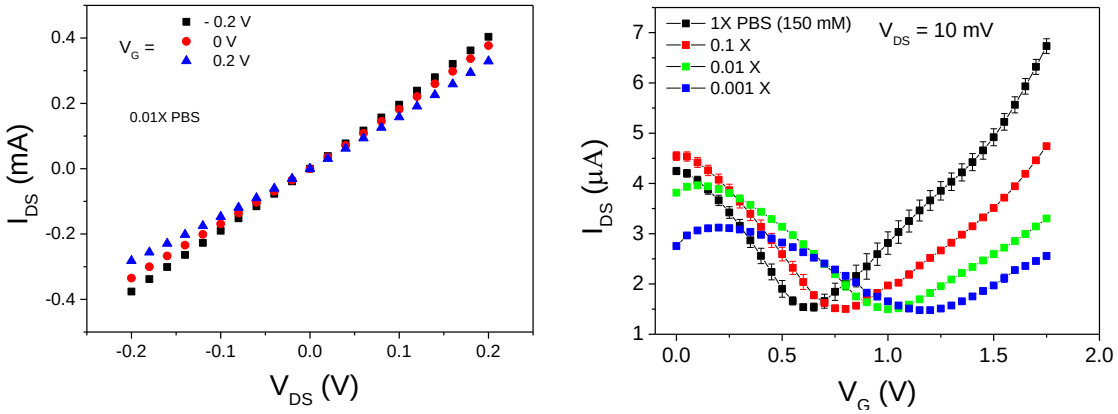

Figure 2. 2a (left) Output curves of the device for several V_G values. 2b (right) Evolution of V_G min as function of PBS ionic strength.