Enhancing Device Sensitivity of Graphene Field Effect Transistor DNA Biosensors via Single Layer Boron Nitride

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ABSTRACT

When it comes to nanoscale devices, graphene continues to demonstrate its great potential as an ideal material for various nanostructures. This is in part due to graphene's physical and electrical properties, which include high electrical conductivity, sensitivity, and flexibility. As such, graphene proves to be useful for developing highly-sensitive sensors and electronics of various kinds. Here we present our progress on developing highly-sensitive graphene field-effect transistor (GFET) sensors capable of detecting single-stranded DNA sequences and provide electrical readouts from these sequences. Here we chose to examine the effects of adding monolayer BN to our GFET devices to improve our GFET device sensitivity and performance. Our devices made with a protective monolayer of BN deposited on top of the graphene layer showed higher sensitivity and a lower Dirac voltage than our standard GFET devices. Furthermore, our fabrication method is scalable and reproducible. These highly-sensitive and accurate bio-sensors show great potential for medical sensing applications such as disease detection and DNA sequencing.

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1. INTRODUCTION

When it comes to nanoscale devices, graphene continues to demonstrate its great potential as an ideal material for various nanostructures. The term graphene refers to a 2D monolayer of carbon atoms arranged in a hexagonal-shaped lattice that makes up the basis of graphitic materials. Graphene is a particularly interesting material due to its 2D structure and high quality crystal and electronic properties. However, it was not until 2004 that graphene was discovered. Prior to 2004, it was presumed that 2D materials did not exist. This is because 2D atomic monolayers were typically incorporated into 3D structures (e.g. epitaxial growth on substrates) and it was believed that 2D crystals were too unstable to exist [1]. However, this belief changed once free-standing 2D atomic monolayer crystals, such as graphene, and single-layer boron nitride were experimentally discovered [1].

Regarding device fabrication, graphene is seen as an ideal substance for nanoscale devices due to its unique range of physical and electrical properties, durable mechanical strength, and flexibility [1]. Given its aforementioned flexibility, graphene can also be rolled into 1D tubes known as carbon nanotubes or stacked to form 3D graphite. In addition to this, graphene can be shaped to create several other allotropes of carbon not mentioned here [2]. Graphene based nanostructures also have the added benefits of having their atoms directly interact with their immediate environment and tunable electronic properties. It is for these combined reasons that graphene-based nanostructures show great potential in allowing one to develop a large variety of robust sensor devices [1].

We are currently investigating the effects of adding monolayer BN to our graphene field-effect transistors (GFET). More specifically we are testing two variations of our GFET devices. The first variation involves depositing a layer of BN on top of the layer of graphene used for our GFET device. This ought to allow the layer of BN to serve as a protective layer to insulate our graphene for the external environment. The second variation of our device involves placing the same kind of BN layer underneath the graphene layer. With this method we suspect that the BN layer will be useful for blocking undesired interactions resulting from dangling bonds between the SiO_2 and graphene layer interface. BN is desirable for these specific functions because it shares similar properties with graphene (e.g. stability, high crystal and electronic qualities, etc.) and its physical structure is very similar to that of graphene. Thus, BN can serve as a useful

intermediary or exterior layer for our GFET devices that we suspect may be able to enhance our device performance.

2. GRAPHENE GROWTH AND DEVICE FABRICATION

To make our devices, we start by growing graphene in a 4'' furnace. This is done by baking a copper film on a glass plate in our furnace tube with methane, nitrogen, and hydrogen in accordance with our growth recipe. After baking the copper film, we are left with a very thin film of graphene. Prior to transferring the graphene onto our SiO_2 substrates, we pattern a wafer of SiO_2 with standard photolithography steps and evaporation. This allows us to deposit Au/Cr electrical contacts onto our patterned SiO_2 wafer, which contains our device substrates. To transfer the graphene from the copper film, we perform a bubble transfer in NaOH solution. This allows us to easily transfer the graphene film onto SiO_2 substrates which serve as the basis of our GFET devices. An example of one of our SiO_2 substrates is shown in Figure 1.

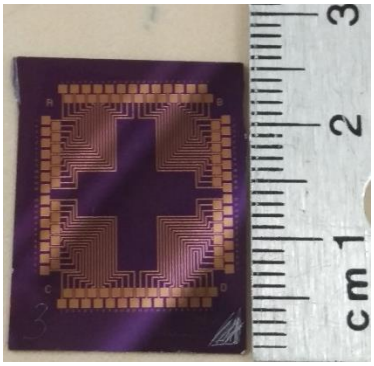

Figure 1. 2 x 2.5 cm SiO_2 substrate for our GFET devices.

Once the substrates with graphene have dried after the bubble transfer, we thoroughly clean these devices with acetone and IPA. This procedure allows us to make our standard GFET devices. When we choose to add an intermediary or protective BN layer to these devices we simply alter a few steps in the process as described below. The various device structures are illustrated in Figure 2.

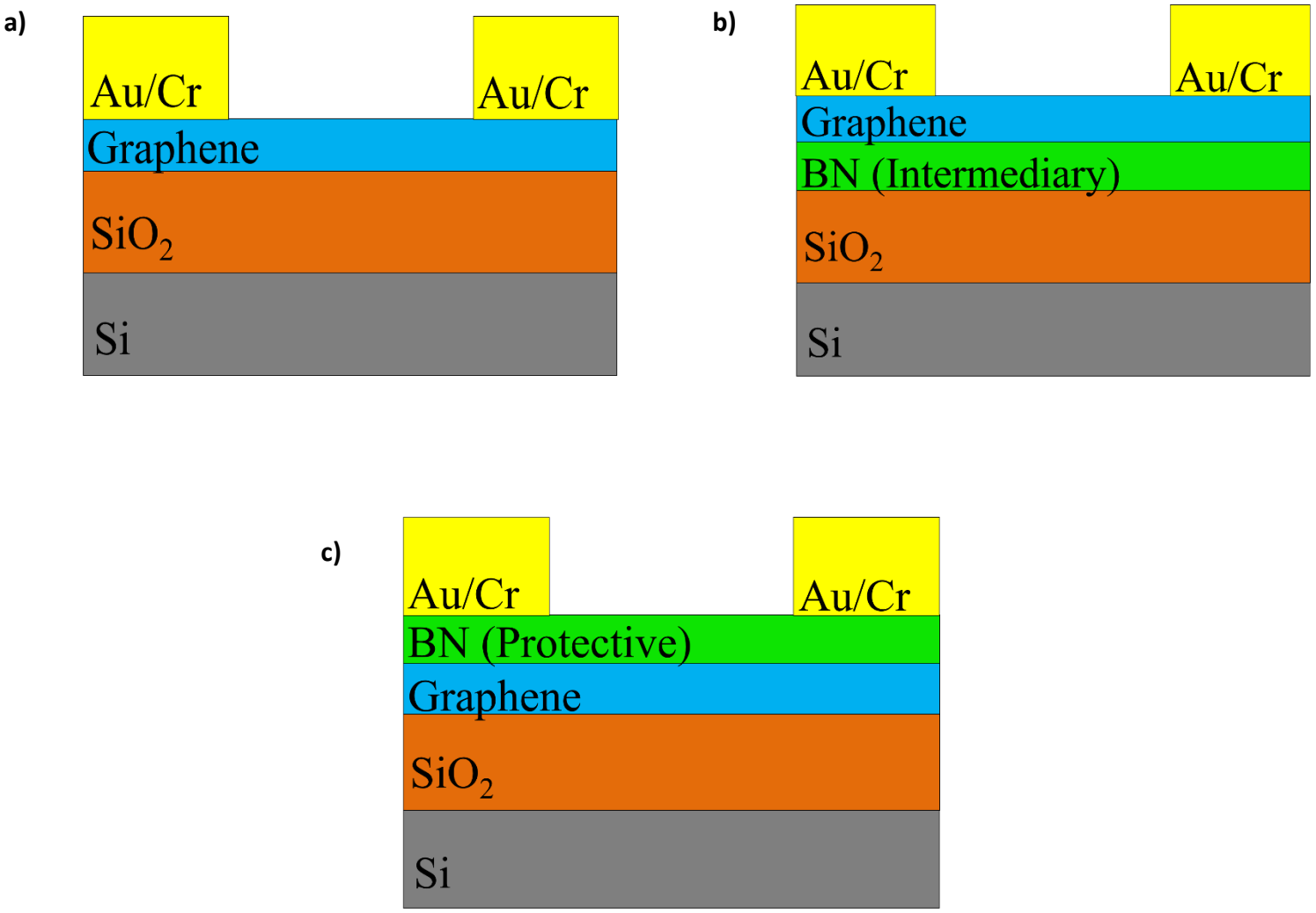

Figure 2: a) Schematic of standard GFET device. b) Schematic of intermediate layer BN GFET device. c) Schematic of protective layer BN GFET device.

To insert an intermediary layer of BN between our graphene and SiO_2, we perform a bubble transfer with BN film prior to transferring graphene. This allows us to place the BN layer on top of the SiO_2 substrate which is then baked on a hot plate, cleaned with IPA and acetone, and annealed after depositing the BN layer. Afterwards we perform standard photolithography to evaporate gold contacts onto the device and then perform a graphene bubble transfer as described above. In the case of the external BN layer, we perform our standard procedure for fabricated

GFET devices and perform a bubble transfer for the BN film on top of the GFET device. In this case, BN serves as a protective layer for the graphene which is now between the SiO_2 substrate and the outer BN film.

Once our various devices have been made, we functionalize them by binding ssDNA to them. When doing this, we use several identical devices, and bind varying concentrations of twenty-two base-pair ssDNA to them. For our control group we use deionized water on GFET devices. By doing this, deionized water serves as a baseline with no ssDNA with which we can compare our GFET devices with ssDNA bound to them. We also have a probe DNA sequence, which is a complimentary strand of DNA to the specific DNA sequence called the target DNA that we wish to detect. For our sensing experiments we bind varying concentrations and variations of our target DNA to our GFET devices and compare their electrical outputs.

3. RESULTS AND DISCUSSION

To determine how our various devices perform, we probe them for their electrical output signals and observe how the Dirac point shifts for each device being compared. A perfect graphene device would display a Dirac point at 0 V. Our standard GFET devices are very close to mimicking this performance—as they tend to have Dirac points falling within the range of 5-10 V. Of course, our devices are not perfect and exhibit some forms of defects such as film tears and undesirable residual materials such as PMMA or photoresist.

Based on the results of the experiments that we have conducted up to this point, we have found that adding an intermediary BN layer to our GFET devices was not particularly useful for enhancing device performance. From our initial devices with BN layers in between the graphene and SiO_2 we noticed that our Dirac point shifted to about -30 V and that they also exhibited low carrier mobilities. This was in part caused by residual PMMA and photoresist on the BN layer which, despite our standard cleaning process, was not completely removed. Compared to our SiO_2 substrates, it was far more difficult to remove the residual photoresist from the BN layer and we found that it would take significantly more effort to try to get our intermediary BN-graphene interfaced devices to exhibit the same performance as the SiO_2-graphene interfaced devices. In summation, these devices tended to perform worse than our standard GFET devices

in which our graphene layers were able to make direct contact with the SiO_{2-} substrate. On the other hand, our devices with an external, protective layer of BN showed much more promise. Our preliminary devices showed Dirac points very close to 0 V and exhibited higher sensitivity and carrier mobilities than our standard GFET devices. In addition to this, these BN-GFET devices have the added benefit of having a layer of BN protect the graphene layer from external contaminants.

Our methods presented here allow us to fabricate GFET devices on a large scale with upwards of 100 devices per fabrication process. Additionally, our fabrication methods can be easily reproduced and are scalable. At the moment we are still continuing our work on this project and are examining the effects on device performance that intermediary and external BN layers can have on our GFET devices.

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5. REFERENCES

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