Wednesday Morning, October 20, 2010

Graphene Focus Topic

Room: Brazos - Session GR+EM+MS+TF+MI-WeM

Graphene and Carbon-based Devices

Moderator: N.P. Guisinger, Argonne National Laboratory

8:00am GR+EM+MS+TF+MI-WeM1 Epitaxial Graphene: Effects of Dielectric Overlayers and Device Design on FET Performance, *M.J. Hollander, R. Cavalero, D. Snyder, M. LaBella, K. Trumbull, Z. Hughes, J. Robinson*, The Pennsylvania State University

The realization of a graphene-based electronic technology necessitates large-area graphene production, as well as the ability to integrate graphene with highly insulating films that act as the gate dielectric in field effect transistors (FETs). Graphene's two dimensional nature allows for phenomenal electronic properties and ultimate scalability, but also makes it susceptible to doping and scattering by charged impurities, dangling bonds, and other defects that may derive directly from choice in gate dielectric over-layer on conduction within the graphene channel is of fundamental interest in designing and producing graphene based FETs. Atomic layer deposition (ALD) has proven to be an excellent technique toward the integration of dielectrics with graphene and provides a means to produce high quality films for gate dielectrics at temperatures below 300C, but requires the use of a thin nucleation layer to promote complete coverage and to protect the graphene.

We present results on graphene FETs utilizing various gate dielectrics and various nucleation layers. Graphene was grown epitaxially on 100 mm SiC wafers and processed using standard photolithographic techniques. Al₂O₃ and HfO₂ gate dielectrics were investigated using SiO₂, TiO₂, and Al₂O₃ nucleation layers in various combinations. We show that choice of gate dielectric and nucleation layer can have a dramatic effect on transistor performance and charge carrier mobility. Saturation current. transconductance, and device hysteresis were examined in the fabricated FETs while charge carrier mobility and charge carrier density within the epitaxial graphene were evaluated using Van der Pauw structures. Graphene FETs utilizing Al₂O₃ and SiO₂ seeded dielectrics exhibit the best performance while TiO2 seeded and unseeded devices exhibit large gate leakage currents resulting in non-functioning FETs. Additionally we provide evidence that the choice of dielectric and seed can significantly impact the Dirac point (minimum conduction), amount of hysteresis, and on/off ratio of the graphene FETs. Trends in saturation current, and transconductance appear be independent of nucleation layer and gate dielectric choice, indicating that conduction through the channel may be limited by mechanisms independent of the nucleation layer and gate dielectric.

In addition to the aforementioned performance metrics, FET performance after continued application of high electric fields across the channel will be reported. Finally, we examine how choice of channel length and width, along with transistor design, effect performance.

8:20am **GR+EM+MS+TF+MI-WeM2** Graphene Oxide on Patterned Self-Assembly Monolayer for Cancer Marker Detection, *D.-J. Kim, C.Y. Jung, O.J. Yoon, N.-E. Lee,* Sungkyunkwan University, Republic of Korea, *J.-S. Park*, Korea Electronics Technology Institute, Republic of Korea

Graphene, two-dimensional sheet of sp²-hybridized carbon atom, has received much attention in recent years for many applications such as energy storage and conversion, batteries, fuel cells, optoelectronic device, field-effect transistors, electromechanical resonators, chemical sensors, and biotechnologies. Oxidized form of graphene, graphene oxide, which has functional groups of epoxide, carbonyls, hydroxide, and phenol groups at both sides can also provide various functionalities for biomedical applications because the high densities of defective sites can provide many active sites for electron transfer to biological species, similarly to graphene. In this study, we demonstrate the graphene oxide based immunosensor for cancer marker detection. Graphene oxide pattern was formed selectively on self-assembly monolayer (SAM) pattern formed by using inkjet printing. The hydroxyl terminated ITO electrode was first selectively functionalized by the amine (-NH₂) group by inkjet printing of SAM, and the graphene oxide flakes were adsorbed selectively on the SAM pattern. After immobilization of monoclonal antibody to PSA (prostate specific antigen) which is the biomarker in prostate cancer, the electrochemical performance of immunosensor was evaluated by varying the concentration of PSA-ACT (prostate specific antigen/α1-antichymotrypsin) complex. And the PSA polyclonal antibody conjugated with gold nanoparticles was applied to amplification of electrochemical signal because the attachment of nanoparticles onto electrodes drastically enhances the conductivity and electron transfer from the redox analytes. The results showed that graphene oxide based immunosensor can be possibly applied to the delicate diagnosis of various disease markers as well as PSA at the extremely low level and with ultra-high sensitivity.

8:40am GR+EM+MS+TF+MI-WeM3 Spintronics in Graphene Based Nanostructures, J. Fernández-Rossier, Universidad de Alicante, Spain INVITED

Spintronics in graphene has received attention from two complementary points of view. On one side, the small spin orbit coupling and nuclear spin density suggest that graphene should have very long spin relaxation and decoherence times, which would make it suitable for long distance spin transport and, eventually, for spin-based quantum computing. Although small, spin orbit interaction turns graphene into a spin hall insulator, at very small temperatures, so that the edges of graphene should carry spontaneous spin currents. Independently of the above, the zigzag edges of graphene nanostructures, like graphene ribbons and islands, are predicted to present ferromagnetic spin order due to electron electron coupling. In this talk I will discuss the properties of graphene nanostructures when both the spin-orbit coupling and the Coulomb interactions are included. I will show how the combined action of spontaneous spin order and spin orbit coupling results in persistent charge currents flow in the edges of a variety of graphene nanostructures.

9:20am **GR+EM+MS+TF+MI-WeM5 CVD** Gate Dielectrics and **Bandgap Engineering of Graphene Layers**, *W.J. Zhu*, *D. Neumayer*, *V. Perebeinos*, *P. Avouris*, IBM T.J. Watson Research Center

Graphene is very promising for electronic devices, particularly for analogue high frequency devices due to its high intrinsic mobility. One challenge in graphene devices is the formation of gate dielectrics on graphene due to its hydrophobic nature. We found a new CVD gate insulator that provides uniform coverage on graphene while preserving the channel mobility, thus making it a very promising gate dielectric for graphene devices. We also studied the channel resistance and the modification of the band structure as a function of the vertical electric field near the Dirac (neutrality) points in the fabricated top-gated graphene devices. We found that as the vertical field increases, in bi-and tri-layer graphenes, the band-gap increases, while in multi-layer graphene, the band-overlap increases. The values of bandgap/overlap in bi-, tri- and multi-layer graphenes were estimated using a simple model which takes into account the variations of the surface electrostatic potential at the Dirac/neutrality point.

9:40am GR+EM+MS+TF+MI-WeM6 Characterization of Devices Fabricated from Electrostatically Transferred Graphene: Comparison with Epitaxial based Devices, S.W. Howell, L.B. Biedermann, T. Ohta, T.E. Beechem, W. Pan, A.J. Ross, D.C. Trotter, Sandia National Laboratories

In this paper, we present our recent advancements in electrostatically transferring epitaxial graphene (EG) from SiC(0001) and SiC(000-1) to arbitrary glass substrates (including Pyrex). We will compare the electronic properties of electrostatically transferred EG and nominally-equivalent asgrown EG on SiC. These properties are measured using magnetoresistive, four-probe, and field effect transistor geometries. We feel this is a potential enabling technology for integration of graphene with structured and electronically-active substrates, such as MEMS and CMOS.

CVD-grown graphene on Cu has attracted wide interest since it can be readily transferred to arbitrary substrates. However, CVD-grown graphene has been shown to have lower mobilities and smaller domain sizes than EG. EG is difficult to transfer to arbitrary substrates. Currently two techniques exist to transfer EG – a gold/polymer film handle and thermal tape exfoliation [1,2]. While transfer is possible with these techniques, problems exist including contamination and damage, as measured by Raman spectroscopy.

To overcome the issues with the above mentioned transfer techniques, we have developed a technique capable of electrostatically transferring both patterned and chip-scale regions of EG to arbitrary glass substrates. We start with high-quality graphene (mobility 14,000 cm²/Vs and domains >100 um²) grown using an Ar-mediated approach [3,4]. In electrostatic graphene transfer, a large electric field is applied between the donor graphene sample (anode) and the acceptor insulating substrate (cathode). This strong electrostatic force deposits graphene on the insulating surface. Electrostatic transfer of EG is a clean technique which, unlike other EG transfer methods, does not contaminate the graphene from SiC(000-1) and monolayer graphene from SiC(0001) have been transferred using this technique.

Our initial attempts at EG transfer have been extensively characterized with Raman spectroscopy and atomic force microscopy. Raman spectroscopy shows that the inherent strain in EG has been partially relaxed. Furthermore, a defect peak (D peak) is frequently not seen in the transferred graphene indicating that the procedure does not significantly damage the graphene film.

Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy under contract DE-AC04-94AL85000.

[1] D. Lee et al. Nano Lett. 8, 4320 (2008)

[2] S. Unarunotai et al. APL 95, 202101 (2009)

[3] W. Pan et al. submitted to APL (2010)

[4] K. Emtsev et al. Nature Mat. 8, 203 (2009)

10:40am GR+EM+MS+TF+MI-WeM9 Electron Transport in Dual-Gated Mono- and Bilayer Graphene Devices with High-k Dielectrics, E. Tutuc, S. Kim, B. Fallah, K. Lee, J. Nah, S.K. Banerjee, The University of INVITED Texas at Austin, L. Colombo, Texas Instruments, Inc. A key issue for graphene-based devices is the deposition of thin high-k dielectric layers combined with a minimum electron mobility degradation. Here we examine the carrier transport in mono and bilayer graphene fieldeffect transistors with top Al₂O₃ and HfO₂ dielectrics. The high-k dielectric films are grown by atomic layer deposition (ALD), and using a 1nm-thick Al interfacial layer in order to create intentional nucleation centers for the ALD. We show that this deposition technique allows the realization of high mobility graphene devices with aggressively scaled top dielectrics. We investigate the carrier mobility dependence on dielectric thickness and temperature in mono-layer graphene field-effect transistors with high-k dielectrics. The electron transport in dual-gated graphene bilayers with Al₂O₃ top dielectric reveals an interesting band-gap energy dependence on transverse electric field and perpendicular magnetic field in this system.

11:20am GR+EM+MS+TF+MI-WeM11 In-situ X-ray Photoelectron Spectroscopy Studies of Ozone-based ALD Al₂O₃ Dielectrics on Graphite and Graphene, A. Pirkle, S. McDonnell, University of Texas at Dallas, L. Colombo, Texas Instruments, Inc., R.M. Wallace, University of Texas at Dallas

We present a study of ozone-based atomic layer deposition (ALD) of Al₂O₃ films on graphene and bulk graphite. Uniform deposition of scalable devicequality high-k dielectrics on graphene is a substantial hurdle for the implementation of conventional FET devices as well as novel device structures exploiting the unique transport properties of graphene. Trimethylaluminum (TMA) / O₃ processes are found to result in uniform Al₂O₃ depositions on graphite and graphene surfaces (1), in contrast to common TMA / H₂O-based processes which result in nonuniform nucleation at defects and step edges.

In order to further examine the nature of interactions between TMA / O_3 and graphene, we utilize *in-situ* x-ray photoelectron spectroscopy (XPS) coupled via a UHV transfer line to an ALD reactor. Morphology of deposited films is also examined *ex-situ* using atomic force microscopy (AFM). We examine the impact of several parameters on Al₂O₃ deposition. Choice of deposition temperature is critical, as etching of graphene by O₃ is observed at elevated temperatures (2) but dielectric quality is degraded at low temperature (3). We also examine the impact of surface condition on Al₂O₃ composition particularly with regard to partially reacted TMA precursor molecules; various surface treatments are employed to approximate realistic device processing conditions. Finally, the effect of variations in purge time between ALD precursor pulses is studied; a reduction in deposition with increased purge time indicates that weakly bonded precursor molecules (TMA and O₃) are easily desorbed from the graphene surface.

This work is supported by the NRI SWAN and MIND centers.

1: B. Lee, et. al., Appl. Phys. Lett. 92 (20), 203102 (2008)

2: G. Lee, et. al., J. Phys. Chem. C 113 (32), 14225 (2009)

3: S. K. Kim, et. al., J. Electrochem. Soc. 153 (5), F69 (2006)

Authors Index

Bold page numbers indicate the presenter

— A —

Avouris, P.: GR+EM+MS+TF+MI-WeM5, 1 — **B** —

Banerjee, S.K.: GR+EM+MS+TF+MI-WeM9, 2 Beechem, T.E.: GR+EM+MS+TF+MI-WeM6, 1 Biedermann, L.B.: GR+EM+MS+TF+MI-WeM6,

¹ — **C** — Cavalero, R.: GR+EM+MS+TF+MI-WeM1, 1 Colombo, L.: GR+EM+MS+TF+MI-WeM11, 2;

GR+EM+MS+TF+MI-WeM9, 2

— F —

Fallah, B.: GR+EM+MS+TF+MI-WeM9, 2 Fernández-Rossier, J.: GR+EM+MS+TF+MI-WeM3, **1**

— н —

Hollander, M.J.: GR+EM+MS+TF+MI-WeM1, **1** Howell, S.W.: GR+EM+MS+TF+MI-WeM6, **1** Hughes, Z.: GR+EM+MS+TF+MI-WeM1, 1 — **J** — Jung, C.Y.: GR+EM+MS+TF+MI-WeM2, 1 — **K** —

Kim, D.-J.: GR+EM+MS+TF+MI-WeM2, 1 Kim, S.: GR+EM+MS+TF+MI-WeM9, 2 — L —

LaBella, M.: GR+EM+MS+TF+MI-WeM1, 1 Lee, K.: GR+EM+MS+TF+MI-WeM9, 2 Lee, N.-E.: GR+EM+MS+TF+MI-WeM2, 1 — M —

McDonnell, S.: GR+EM+MS+TF+MI-WeM11, 2

Nah, J.: GR+EM+MS+TF+MI-WeM9, 2 Neumayer, D.: GR+EM+MS+TF+MI-WeM5, 1

-0-

Ohta, T.: GR+EM+MS+TF+MI-WeM6, 1

Pan, W.: GR+EM+MS+TF+MI-WeM6, 1

Park, J.-S.: GR+EM+MS+TF+MI-WeM2, 1 Perebeinos, V.: GR+EM+MS+TF+MI-WeM5, 1 Pirkle, A.: GR+EM+MS+TF+MI-WeM11, 2

Robinson, J.: GR+EM+MS+TF+MI-WeM1, 1 Ross, A.J.: GR+EM+MS+TF+MI-WeM6, 1 — S —

Snyder, D.: GR+EM+MS+TF+MI-WeM1, 1

Trotter, D.C.: GR+EM+MS+TF+MI-WeM6, 1 Trumbull, K.: GR+EM+MS+TF+MI-WeM1, 1 Tutuc, E.: GR+EM+MS+TF+MI-WeM9, 2

— W — Wallace, R.M.: GR+EM+MS+TF+MI-WeM11, 2 — Y —

Yoon, O.J.: GR+EM+MS+TF+MI-WeM2, 1

Zhu, W.J.: GR+EM+MS+TF+MI-WeM5, 1