

Direct Growth of Large Area Graphene on Si/SiO₂ substrate from Sputtered Carbon/Nickel Films

Genhua Pan¹, Mark Heath^{1,2}, Bing Li¹, David Horsell², M. Lesley Wears², Shakil Awan¹, Laith Al-Taani¹

¹Wolfson Nanomaterials & Devices Laboratory, Faculty of Science and Technology,
University of Plymouth, Devon, PL4 8AA, UK

²College of Engineering, Mathematics and Physical Sciences, University of Exeter, Exeter,
EX4 4QF, UK

G.Pan@plymouth.ac.uk

Abstract

Graphene is a highly promising material for numerous potential applications, ranging from electronics and photonics to sensors and composites [1-2]. To exploit these applications at an industrial level requires large scale growth of high quality graphene on device compatible substrates. To date, this has mainly been achieved via chemical vapour deposition (CVD) [3] and single crystal SiC epitaxial [4] growth routes. CVD graphene has been synthesised on various metal substrates such as nickel [5, 6] and copper [3], but the need to transfer it to different substrates for device fabrication has so far hindered its up-scaling to roll-to-roll production methods. Epitaxially grown graphene has been demonstrated to be a viable route for the production of electronic devices, such as field effect transistors[7]; however, SiC wafers are expensive and, unless SiC is required in the device, again graphene needs to be transferred. Graphene has also been synthesised via the rapid thermal processing (RTP) route with Ni Layer atop of single crystal SiC substrate at a temperatures ranging from 1100 °C [8] down to 750 °C [9]. Large area single or few layer graphene were also grown via the RTP route using amorphous SiC or carbon films and metal layers deposited by evaporation [10] or sputtering [11]. However, in all those growth techniques, transfer of graphene from metal substrate to insulator is required for device applications.

Here we show the direct growth of large area graphene on Si/SiO₂ substrates from sputtered amorphous carbon or SiC films either atop or underneath a Ni layer using in-situ or ex-situ RTP in the temperature range from 650 to 1000 °C. We have found that for samples with thick Ni layers (>100nm), graphene grew on the top surface of the stack, in close contact with the Ni or Ni-silicide and suspended in the liquid solution after the etching of the Ni-Silicide in HCl. However, when thinner Ni and carbon layers were used, the graphene remained on the original substrate (Si/SiO₂) after the Ni-Silicide is etched away. Figure 1 is a summary of the typical properties of the graphene. Raman spectra typical of high quality exfoliated monolayer graphene [12] were obtained for samples under optimised conditions. A fast cooling rate was found to be essential to the formation of monolayer graphene. Samples with Ni atop SiC or carbon produced the best monolayer graphene spectra with ~40% surface area coverage as shown by Raman mapping, whereas samples with Ni below SiC or carbon produced poorer quality graphene but with 99% coverage. The results present a potential route for the production of large area graphene directly on Si/SiO₂ insulating wafers, which could be a critical step forward for the integration of graphene into modern semiconductor device process flows. Growth mechanism and detailed characterisation of graphene by Raman, optical, AFM, and electrical transport measurement will be presented at the conference.

References

- [1] Geim A. K., "Graphene: Status and Prospects", *Science*, **324** (2009),1530-1534.
- [2] Geim A. K, Novoselov K. S., "The rise of graphene", *Nat Mater.*, **6** (2007), 183-191.
- [3] Li XS, Cai WW, An JH, Kim S, Nah J, Yang DX, et al. "Large-Area Synthesis of High-Quality and Uniform Graphene Films on Copper Foils", *Science*, **324** (2009),1312-1314.
- [4] Berger C, Song ZM, Li TB, Li XB, Ogbazghi AY, Feng R, et al. "Ultrathin epitaxial graphite: 2D electron gas properties and a route toward graphene-based nanoelectronics", *J Phys Chem B*. **108** (2004),19912-19916.
- [5] Yu Q, Lian J, Siriponglert S, Li H, Chen YP, Pei S-S., "Graphene segregated on Ni surfaces and transferred to insulators", *Appl. Phys Lett.*, **93** (2008),113103-3.

- [6] Kim KS, Zhao Y, Jang H, Lee SY, Kim JM, Kim KS, et al., "Large-scale pattern growth of graphene films for stretchable transparent electrodes", *Nature* **457** (2009), 706-710.
- [7] Moon JS, Curtis D, Hu M, Wong D, McGuire C, Campbell PM, et al., "Epitaxial-Graphene RF Field-Effect Transistors on Si-Face 6H-SiC Substrates", *IEEE Electron Device Letters*, **30** (2009), 650-652.
- [8] Hofrichter J, Szafranek BuN, Otto M, Echtermeyer TJ, Baus M, Majerus A, et al., "Synthesis of Graphene on Silicon Dioxide by a Solid Carbon Source", *Nano Lett.* **10** (2009), 36-42.
- [9] Juang Z-Y, Wu C-Y, Lo C-W, Chen W-Y, Huang C-F, Hwang J-C, et al., "Synthesis of graphene on silicon carbide substrates at low temperature", *Carbon*, **47** (2009), 2026-2031.
- [10] Zheng M, Takei K, Hsia B, Fang H, Zhang X, et al., "Metal-catalyzed crystallization of amorphous carbon to graphene", *APPLIED PHYSICS LETTERS* **96** (2010), 063110.
- [11] Orofeo CM., Ago H, Hu B, and Tsuji M, "Synthesis of Large Area, Homogeneous, Single Layer Graphene Films by Annealing Amorphous Carbon on Co and Ni", *Nano Res* **4** (2011), 531-540.
- [12] Ferrari AC, Meyer JC, Scardaci V, Casiraghi C, Lazzeri M, Mauri F, et al. Raman Spectrum of Graphene and Graphene Layers. *Physical Review Letters*. **97** (2006):187401.

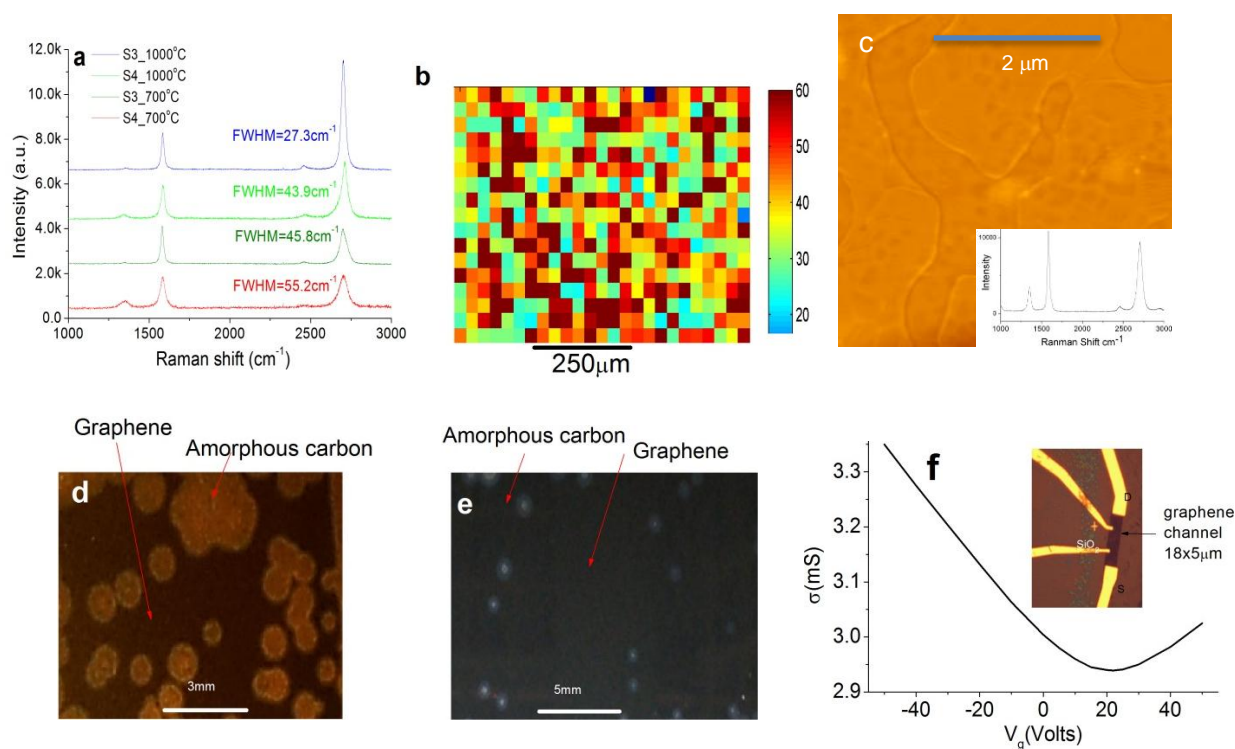


Figure 1. Summary of properties of graphene. (a) Raman spectra of samples processed with different RTP process conditions. (b) Raman mapping of FWHM of the 2D band for a sample area of 750µm x 500µm. (c) AFM image of graphene on Si/SiO₂ substrate after etching of Ni-Silicide in HCl, inset is a Raman spectrum of the sample. (d) and (e), Camera shots showing distinctive areas with and without graphene after RTP. (f) Electrical characteristics of a back-gated graphene field effect transistor, inset is an optical microscope picture of the device.