Large Area Plasma-Enhanced Chemical Vapor Deposition of Nanocrystalline Graphite on Insulator for Electronic Device Application

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This paper reports on large area plasma-enhanced chemical vapor deposition (PECVD) of nanocrystalline graphite (NCG) on thermally grown SiO_2 wafer, quartz and sapphire substrates. Grown films are evaluated using Raman spectroscopy, ellipsometry, scanning electron microscopy (SEM) and atomic force microscopy (AFM). Electrical characterization and optical transmission measurements indicate promising properties of this material for use as transparent electrodes and for electronic device application. A plasma-based etch process for NCG has been developed.

Recently, growth of NCG on sapphire by molecular beam epitaxy has been reported [1]. This process, however, requires ultra high vacuum compared to PECVD. Our contribution is a step towards large area direct growth of few layer graphene on insulating substrates by PECVD, thus eliminating the required transfer of graphene grown on metal catalysts by CVD [2].

The NC-graphite film is deposited using an Oxford Instruments Nanofab 1000 Agile tool, which allows processing of wafers up to 8" in size. The table temperature is measured by a thermocouple below the susceptor on which the substrate sits; the actual temperature of the samples is lower. In this work a thermally oxidized 150 mm diameter silicon wafer was used for deposition. Quartz and sapphire samples of 11x11 mm^2 in size were used in a separate run for comparison. The process chamber is first conditioned with hydrogen at 1000 mT and a flow rate of 100 sccm. This is followed by a heat-up procedure from loading temperature of 700°C to the processing temperature of 900°C. The deposition is based on introduction of methane and hydrogen at 60 sccm and 75 sccm, respectively, at a pressure of 1500 mT and RF power of 100 W.

As-deposited NCG films are analyzed using a Rennishaw InVia Raman spectrometer with 532 nm laser excitation wavelength. Typical Raman spectra of as-deposited NCG on SiO_2, quartz and sapphire are shown in Fig 1. All spectra exhibit distinct D, G and 2D peaks at around 1350, 1600 and 2700 cm^{-1}, respectively. The G peak position is unaffected by the excitation wavelength (532, 633 and 785 nm), and the intensity ratio of the D to G peak is ~2.3. These observations are in accordance with previous reports on Raman spectra of NCG [3]. The in-plane correlation length L_a = 21 Å is estimated from the D/G peak intensity ratio [4]. Figure 2 shows an SEM micrograph of the surface topology of a 35 nm thick NCG film on SiO_2. The roughness RMS of 1.17 nm for this film was obtained from a 1x1 μm^2 large AFM scan. This is an increase compared to 0.1 ± 0.02 nm for the underlying SiO_2. The thickness uniformity of the NCG film grown on a 150 nm wafer (shown in Fig. 3) was measured by ellipsometry, with results shown in Fig. 4. The central 7x7 cm^2 area of the wafer (see dotted line in Fig. 4) was mapped using Raman spectroscopy (532 nm, 5 mm data point spacing). The obtained distribution of the D to G peak intensity ratio is shown in Fig. 5.

Four-point hall measurement on 35 nm thick NCG on SiO_2 was used to obtain the sheet resistance R_S = 3728 Ω/sq, mobility μ = 2.49 cm^2/Vs and carrier concentration N = 1.8x10^{20} cm^{-3}. Additionally, simple electrical structures were fabricated on various samples for resistance measurements and film thickness measurements by AFM. First, we pattern 4 and 50 µm wide lines with various lengths using photolithography. This is followed by reactive ion etching of the NCG film in 20 sccm O_2, 20 mT pressure and 20 W RF power with an etch rate of 5 nm/min. After resist strip using N-Methyl-2-pyrrolidone (NMP), Ti/Au contacts are realized by photolithography and subsequent lift-off in NMP, as shown in Fig. 6. For these processed films, bulk resistivity of ρ = 0.029 Ωcm was obtained from two-probe measurement. The optical transmission of an as-deposited 6.6 nm thick NCG on quartz glass was measured and is above 80% across the visible spectrum.

Current work involves optimization of NCG film thickness uniformity and process tuning for thinner films with larger crystalline domains.

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References

[1] S. Jerng, D. Yu, Y. Kim, J. Ryou, S. Hong, C. Kim, S. Yoon, et al. J. Phys. Chem. C, 2011, **115 (11)**, pp 4491–4494

[2] A.K. Geim, Science, **324(5934)** (2009), 1530-1534.

[3] A. C. Ferrari and J. Robertson, Phil. Trans. R. Soc. A **362** (2004), pp 2477–2512.

[4] A. C. Ferrari and J. Robertson, Physical Review B, **61(20)** (2000), 14095.

Figures

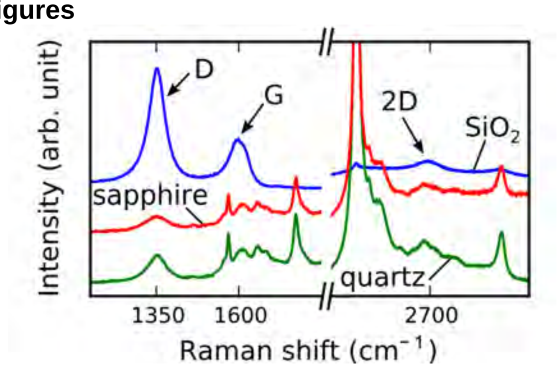

Figure 1: Raman spectra of NCG on thermally grown SiO_2, sapphire and quartz glass.

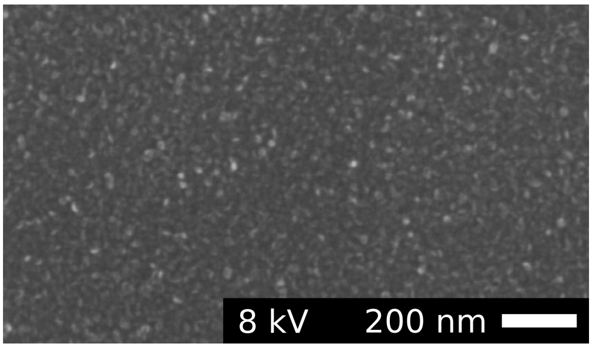

Figure 2: SEM close-up of as-deposited NCG film on thermal oxide.

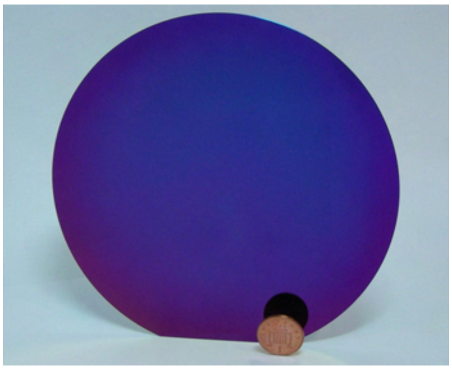

Figure 3: Photograph of 150 mm wafer with NCG film. British penny shown for size comparison.

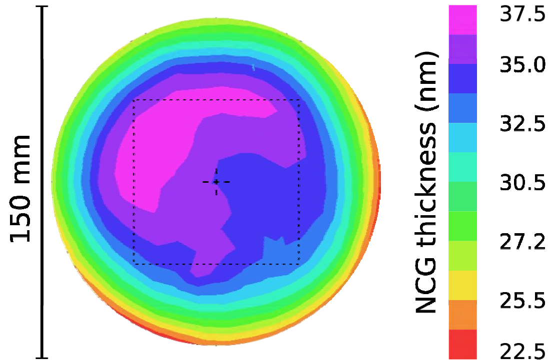

Figure 4: Ellipsometer thickness mapping of NCG film grown on 150 mm wafer.

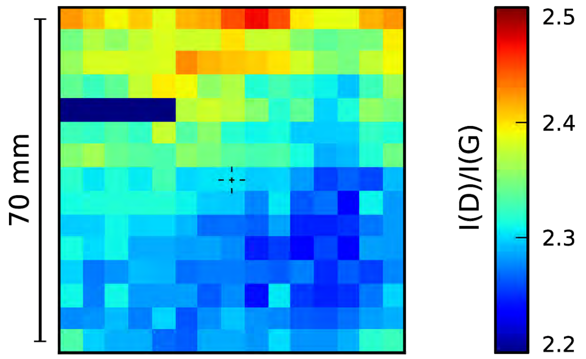

Figure 5: Distribution of Raman D to G peak intensity of central part of 150 mm wafer. Mapping area indicated in Fig 4.

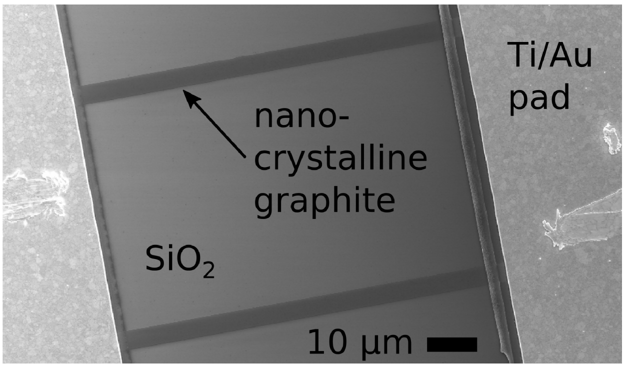

Figure 6: SEM micrograph of NCG film patterned into two 4 µm wide ribbons and contacted by Ti/Au.