Catalytic-Free Synthesis of High Mobility Graphene on Sapphire

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Recent success of graphene transistor operation in the GHz frequency range demonstrates the potential of this material for high speed electronics.[1,2,3] Methods such as spin deposition of reduced graphene oxide,[4] chemical vapor deposition (CVD) on metal substrates,[5,6] plasma assisted deposition,[7] and sublimation of Si from the surface of single crystal SiC semiconductor wafers[8] have all shown potential for large area synthesis routes. Of these techniques, the CVD process is likely the most attractive alternative to the "well established" silicon sublimation from SiC due to the inherent control over process chemistry and the flexibility in choosing precursors. To date, CVD of graphene has been limited to catalytic growth on Ni or Cu at temperatures near 1000°C.[9,10] However, the presence of a metal substrate induces challenges for the production of semiconductor devices. We present a novel CVD process for the growth of high quality monolayer graphene on sapphire *without* the presence of a metal catalyst.

We will discuss catalytic-free synthesis of graphene on standard 50 - 100 mm diameter c-plane sapphire wafers. Graphene films are synthesized via the decomposition of methane (CH_4) in hydrogen (H_2) using an argon (Ar) carrier gas. Growths are performed at 1425°C - 1700°C at pressures of 50 - 600 Torr. Highly uniform growth is achieved via this technique, with variation in sheet resistance across a 50mm wafer of <2% (Figure 1) – comparable to industry standards. Raman spectroscopy confirms the formation of monolayer and bilayer graphene, with improved structural quality as deposition temperature increases. Using the D/G ratio, we find that the domain size increases from an average low of 32 nm at 1425-1450°C (D/G = 0.42) to greater than 270 nm (D/G = 0.05) at 1575°C (Figure 2a and 2b). Additionally, we find that in all cases, the 2D/G ratio is greater than 1.5 with the 2D peak being fit to either one or four Lorentzians, indicating deposition of mono- or bi-layer graphene (Figure 2b and 2c) is achieved. In addition to high structural quality, CVD graphene on sapphire yields strain relieved films with minimal graphene/sapphire interaction, as indicated by a 2D peak position that ranges from 2685 – 2705cm^{-1} (Figure 2d), indicating minimal interaction between the graphene and sapphire. Additionally, according to XPS (Figure 2e), there is no evidence of an interfacial (buffer) layer between graphene and sapphire.

Importantly, we will also present data indicating that catalytic-free synthesis on sapphire produces graphene with superior transport properties compared to graphene on SiC(0001) and comparable to high quality catalytic-CVD graphene. Evident in Figure 4, the carrier mobility dramatically improves from <1000 cm^2/Vs to an average of 3200 cm^2/Vs as the growth temperature is increased to >1500°C. Additionally, we note that water vapor and other environmental conditions can significantly effect the charge carrier concentration and thus mobility. This is evident in Figure 4, where we find a mild anneal at 5x10^{-5} Torr for 3 hrs results in a 1.5x increase in carrier mobility, presumably the result of water desorption from the graphene. Finally, transport properties vary little with magnetic field and temperature (Fig.5).

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Figures

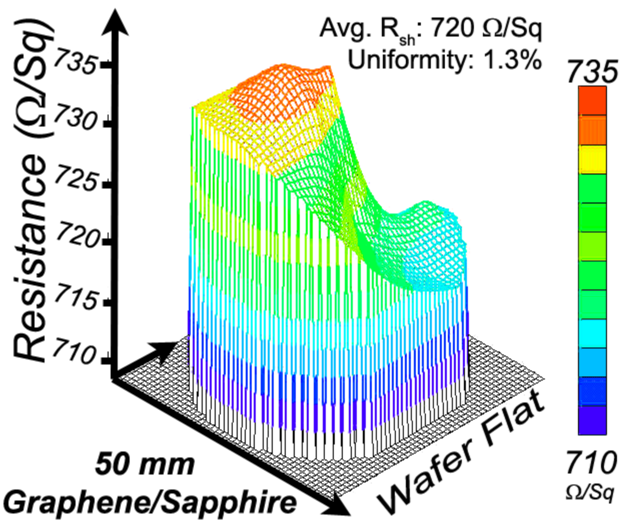

Figure 1: Sheet resistance map of a 50mm wafer of graphene on sapphire demonstrating a high level of uniformity.

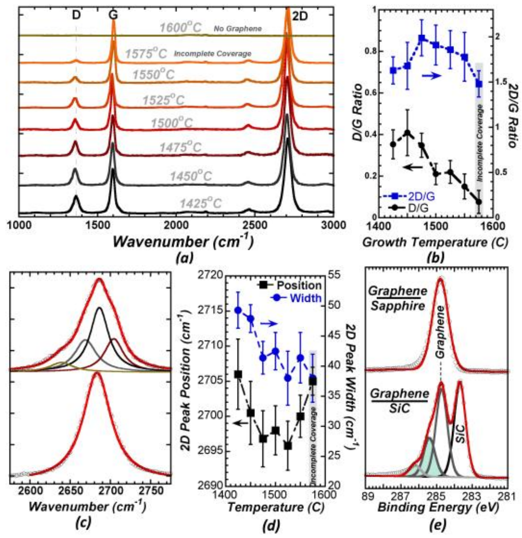

Figure 2: Raman spectroscopy (a-d) of graphene grown on sapphire demonstrating improved structural quality (a,b) with growth temperature, mono and bi-layer graphene (c), and XPS (e) indicating the lack of a buffer layer compared to graphene on SiC.

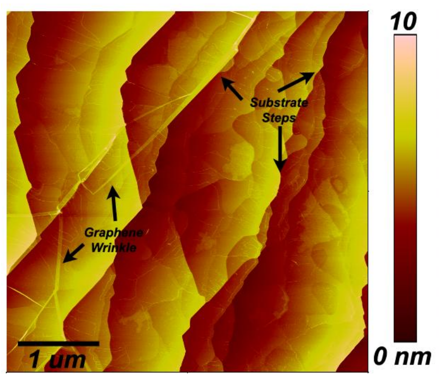

Figure 3: Atomic force micrograph of graphene on sapphire grown at 1500℃. Note the presence of steps in the sapphire substrate and small wrinkles (<1nm high) in the graphene film.

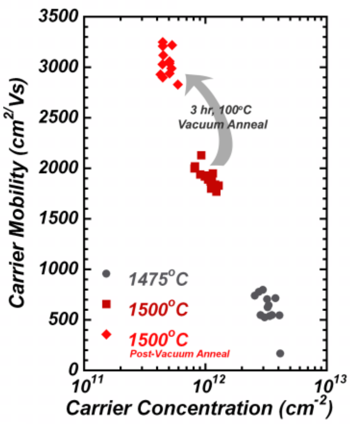

Figure 4: Carrier mobility and concentration of graphene/ sapphire as a function of temperature and measurement conditions.

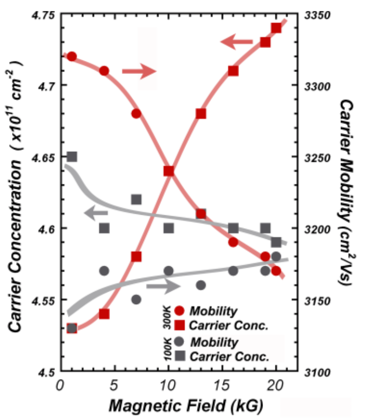

Figure 5: Carrier mobility and concentration of graphene / sapphire grown at 1500℃ as a function of magnetic field at 300K and 100K.