An UHV study of epitaxial growth of graphene on SiC/Si substrates by Si sublimation

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Abstract: - Graphene extraordinary physical and electrical properties, like the high mobility and near ballistic transport at room temperature [1] attracted brilliant researchers to explore its potential application towards sensors, optoelectronics, and nanoelectronic devices. The successful development of graphene based nanoelectronics and sensors depends upon the availability of high quality and large area graphene layers directly grown on a wafer. Epitaxial graphene can be obtained from thermal decomposition of SiC, but the use of SiC wafers is very expensive. Growth of graphene on 3C SiC/Si is more affordable, and at the same time the most efficient route towards the integration of this 2D material in the microelectronic industry based on Si.

The thermal decomposition of SiC in ultra-high vacuum (UHV) gives origin to a contaminant free surface. High temperature annealing of SiC produces the diffusion and sublimation of Si atoms leading to different surface reconstructions depending on the temperature. The challenge in UHV annealing of SiC/Si is to control the Si diffusion. The rate of Si out-diffusionis higher compared to furnace annealing under Ar at atmospheric pressure.

250 nm of 3C SiC was grown on p-doped Si (111) and Si(100) substrate by an alternating supply epitaxy (ASE) process. This process was undertaken in a hot-wall low pressure CVD reactor using the vapour precursors silane (SiH_4) and acetylene (C_2H_2) [2]. Epitaxial Graphene was grown by annealing 3C SiC/Si (111) and 3C SiC (100) substrates in UHV for about 10 mins [3]. We investigated the diffusion of Si in SiC/Si during UHV annealing at temperatures ranging from 1125˚C to 1375˚C in order to produce epitaxial graphene. We have also studied the effect of the exposure to Si flux at 900˚C for various times before the final annealing stage. Chemical composition and surface morphology characterizations were investigated in-situ by using XPS and STM. Ex-situ analysis of the graphene layers was performed by Raman spectroscopy.

Single layer graphene regions have been visualised by Scanning Tunneling Microscopy (STM) in samples annealed at 1250 ˚C. The graphene layers appears to be continuous on the substrate, although wrinkled because of the steps and defects in the underlying SiC/Si(111). STM images of graphene obtained after annealing at 1300 ˚C are shown in Fig. 1. Figure 1a is a 20x20 nm STM image showing a step where a Moiré pattern is visible like a shadow around the center of the image, while in two areas (top and bottom part of the image) it is even possible to observe the honeycomb graphene structure. This difference is due to the presence of multiple/single graphene layers in different areas of the sample. Fig 1b shows the C-rich $(6\sqrt{3}\times6\sqrt{3})$ R30˚ reconstruction caused by the 30˚rotation of the graphene overlayer with respect to the unreconstructed 3C SiC (111) surface. In our case the typical structure due to the Bernal stacking is visible, confirming the presence of more than one graphene layer (Fig 1d).

The number of graphene layers was calculated by using the intensity ratio of XPS graphitic and carbidic peak of SiC. The results show an exponential increase in the number of graphene layers with the increase of temperature, which was fitted to an Arrhenius law (Fig 2). This behaviour is discussed in terms of the Si atom diffusion mechanism through the atomic lattice. The diffusion rate of Si atoms was moderately minimized by depositing Si at 900˚C before the final annealing. Furthermore, STM analysis reveals different types of surface reconstructions during graphene formation. Raman 2D band intensity behaviour confirmed the increase of the graphitic layers leading to a similar Arrhenius law [4]. Further studies are in progress.

References

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

[2] L.Wang et al, Thin Solid films, **519** (2011), 6443-6446

[3] A.Ouerghi, et al. Applied Physics Letters, **96** (2010) 191910-1 – 191910-3.

[4] B Gupta et al. Carbon, **68** (2014) 563-572

Figures

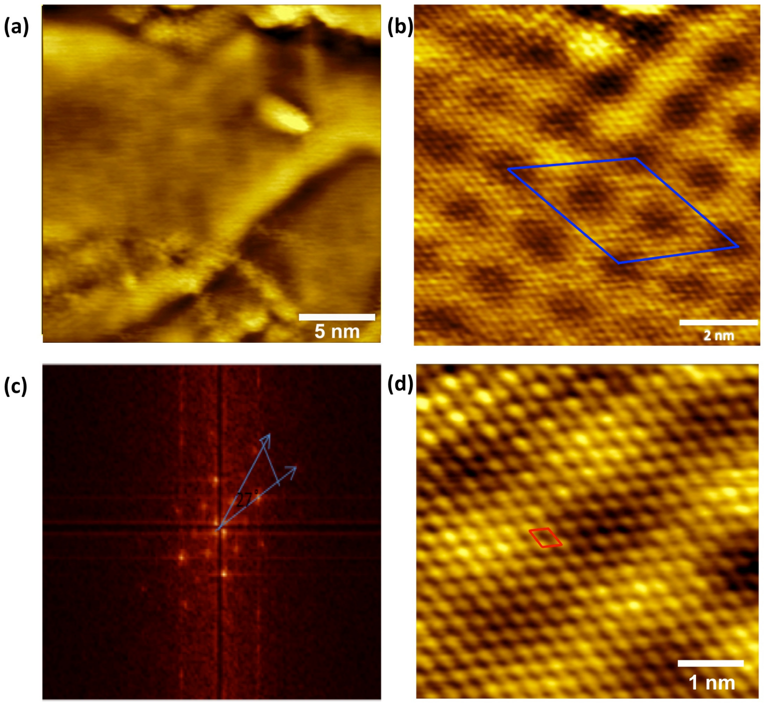

Figure 1 - STM images of graphene obtained by annealing SiC/Si(111) at 1300 ˚C. (a) 20x20 nm^2 area with a step showing a shadow of Moire pattern (V=70 mV, I=0.3 nA), (b) high resolution Moirè pattern with hexagonal symmetry (V=50 mV, I=0.2 nA). A (6√3×6√3) R 30˚ unit cell (blue insert) is also shown, (c) FFT of image (b) showing 27˚ rotation of graphene layer with respect to the buffer layer and (d) high resolution STM image of bi/few layer graphene (V= 50 mV, I= 0.2 nA) with graphene unit cell (red insert) [4].

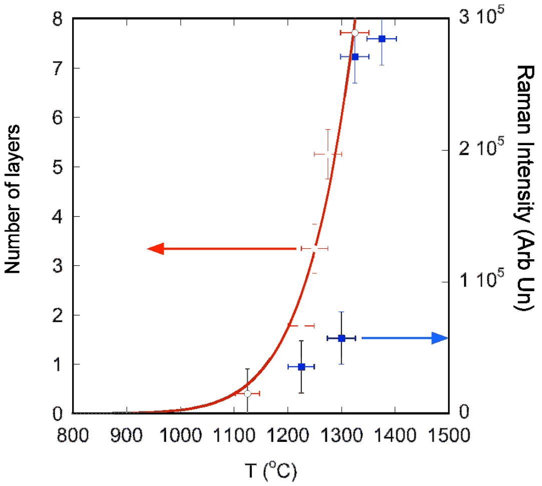

Figure 2 - Number of graphene layers developed in 10’ versus annealing temperature, as obtained from XPS analysis (red circles). The values are fitted to an Arrhenius function (solid line). Blue squares: area of 2D Raman peak versus the annealing temperature.