Probing residual strain in epitaxial graphene layers grown on 4H-silicon carbide (0001) with Raman Spectroscopy

A.J Strudwick, G.L Creeth, N.A.B Johansson & C.H Marrows

School of physics and Astronomy, University of Leeds, Leeds, UK, LS2 9JT phy4ajs@leeds.ac.uk

Thermal decomposition of silicon carbide (SiC) in either UHV or inert gas atmospheres has shown promise as a process to allow the early exciting electrical properties [1,2] of graphene to be integrated into large scale electronics manufacturing. In this process, SiC is heated to temperatures ~1200°C or higher, at these high temperatures the Si preferentially evaporates away from the surface, thus leaving behind a carbon-rich surface that reconstructs to form a sp² bonded epitaxial graphene (EG) layer.[3]

Work here has concentrated on the carbon-terminated $(000\overline{1})$ face of 4H-SiC, EG grown on this face has been shown to provide higher carrier mobilities when compared to the silicon-terminated (0001) face [4]. EG grown on the $(000\overline{1})$ face has been proven to grow with rotational disorder between layers, rather than Bernal stacking, causing the graphene layers to be decoupled from each other and allowing multilayer films to maintain the electronic properties associated with single-layer graphene [5].

A series of techniques have been used to characterize our epitaxial graphene samples. In situ LEED measurements show the expected ring pattern associated with EG grown on SiC(0001) at an onset temperature of around 1300°C with a full graphene film seen for 30 an anneal time of 30 minutes at this temperature. AFM shows a domain like structure with typical lateral grain size of 1µm for samples annealed at 1400°C which is an order of magnitude larger than the grain size observed in samples annealed at 1300°C. LEEM I-V measurements show areas with coherent response signals that are 2-3 µm in lateral size, suggesting a grouping together of the smaller domains seen in AFM micrographs. LEEM I-V measurements also show the samples to be around 2-3 monolayers thick, this measurement is consistent with recent preliminary TEM measurements.

The main focus of this work is the use of Raman microscopy to probe strains present in EG sheets on SiC (0001) [6] formed at different anneal times (10-50 minutes) at a temperature of 1400°C in UHV (10¹⁰ mbar). The strains where calculated by tracking the average 2D peak position deviation from a reference position measured in free-standing exfoliated graphene [7], then using a similar process to previous work on EG (0001) [8,9] strain values can be extracted by linking the Raman shifts to the strains via the Grüneisen parameter of graphene . A compressive strain is seen to increase with anneal time to a maximum value of -0.5% which is less than the -0.9% which would be expected if the strain was caused by differential contraction between the EG and SiC, a model which has fit well to work on EG on SiC(0001) [8,9]. This discrepancy has been attributed to the rotational disorder present during EG growth on SiC(0001) which allows greater decoupling between graphene layers and therefore strain relief.[6] The understanding of the strains present within these epitaxial graphene films is off importance as they have been shown to affect electronic properties such as band gaps [10,11] and carrier mobilities.[8]

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Fig. 1. Examples of Raman spectra of EG on SiC. Spectra are shown for the blank SiC (black squares) and for EG graphene after a 28 minute anneal at 1400°C (red circles). The results of subtracting the background are shown using the blue triangle data markers. The D,G, and 2D band peaks are marked. Only the latter is not obscured by features in the SiC spectrum. Inset: Example of a Lorentzian fit to determine the width and position of a 2D peak [6]



Fig. 2. Variation of 2D peak position shift from that for free-standing exfoliated graphene of 2642 cm⁻¹ (squares) and calculated resulting strain values (triangles) as functions of UHV anneal times for an annealing temperature of 1400°C. The dotted lines are the zeroes of the two ordinate axes. The straight lines are linear fits to the data, and have a reduced R^2 value of 0.928.[6]