Graphene Epitaxy by Chemical Vapor Deposition on SiC

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Graphene, a single sp^2-bonded carbon atomic sheet that is deposited on a SiC substrate has great potential for micro-electronics applications, including conventional components such as high frequency analog devices, and devices in emerging fields such as spintronics, terahertz oscillators, and single-molecule gas sensors. However, a major factor hindering the development of technology for the large-scale production of graphene-based nanoelectronic devices is the lack of access to high-quality uniform graphene layers grown on large SiC substrates. Graphene produced by sublimating Si from SiC heated to high temperatures (1200-2000^{o}C) is sensitive to the surface quality of the SiC substrates. Concurrently, the CVD epitaxial growth of graphene on metal substrates has lately received much attention. Unfortunately, epitaxial growth on metals suffers from the disadvantage that electronic applications require graphene on an insulating substrate, and although wafer-scale transfer is possible, it is a difficult process. A fundamental question thus arises: how can one reduce the dependence of graphene growth on substrate quality and simultaneously improve graphene layer uniformity?

In this paper, we report the CVD of epitaxial graphene (CVD-EG) on SiC substrates using propane gas as the carbon precursor. Graphene layers were grown using a commercially available horizontal CVD hot-wall reactor (Aixtron VP508), which is inductively heated with an rf generator. The epitaxial CVD of graphene relies critically on the creation of dynamic flow conditions in the reactor that simultaneously stop Si sublimation and enable the mass transport of propane to the SiC substrate. Tuning the value of the Reynolds number *Re* enables the formation of optimized Ar boundary layer (BL). Propane that diffuses across the Ar BL to the SiC surface thermally decomposes, and the deposition of epitaxial graphene occurs on the SiC surface. The most critical step is to protect the SiC substrate against Si sublimation at conditions of high temperature ($T \approx 1600^o$C) and low Ar pressure (P). While protecting against Si sublimation, C deposition was enabled with one monolayer resolution by taking advantage of the high efficiency of kinetic processes at high T and low P. Additionally, the formation of FLG is possible on the Si-face SiC(0001) which, in comparison to max 2-3 ML of S-EG, creates greater research opportunities. The proposed method permits the growth rate of graphene on the C-face of SiC(000-1) to be considerably lowered enabling the growth of 1ML, which is extremely difficult in the case of S-EG.

Epitaxial carbon films were deposited predominantly on Si-faces of both semi-insulating and conductive on-axis 4H-SiC substrates. To prove high quality of CVD graphene the ARPES measurements have been performed. The results for layer grown on SiC(0001) are presented below. ARPES data clearly show the expected linear dispersion for relativistic electrons near Dirac point (fig.1). In addition, the samples were characterized by scanning tunneling microscopy (STM) and micro-Raman spectroscopy. The thickness of the graphene films were estimated by ellipsometry. The transport parameters of the graphene samples were measured with the van der Pauw method at room temperature. The electron density in several 1-2 ML graphene films grown in subsequent processes was typically 2-4x10^{12} cm^{-2}, with a macroscopically averaged electron mobility in the range 1200-1800 cm^2/Vs, demonstrating the high electronic quality of the CVD-EG layers on the wafer scale.

The approach proposed here offers numerous potential benefits in comparison to S-EG (sublimated epitaxial graphene), including the application of well-developed commercial epi-systems for SiC epitaxy and the reduction of substrate surface influence on graphene thickness uniformity. Our proposed approach enables precise growth rate control by adjusting the mass transport of the carbon precursor in a similar way to the method used in MOCVD/CVD, as well as the passivation of the SiC substrate by any substances prior to graphene growth. Moreover, one can tune the reactor conditions to grow both CVD-EG and S-EG in the same system. This work is a significant step toward developing a graphene-on-SiC technology that is suitable for industrial scale production in commercial CVD reactors.

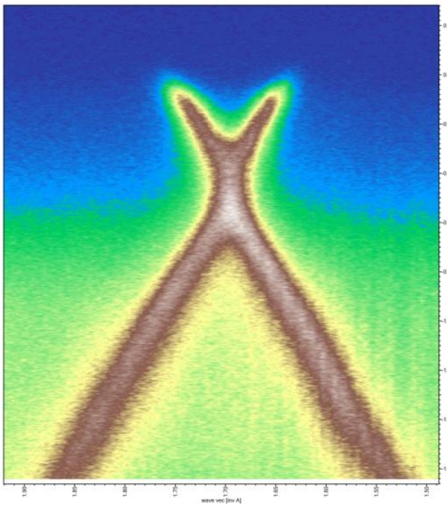

Fig.1 . ARPES spectrum of the CVD-EG grown on an on-axis 4H-SiC(0001) substrate.