Growth of Millimeter-Size CVD Graphene Single Crystals Using Height-Confined Reaction Cavity and High Performance Graphene Field Effect Transistors

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In conventional chemical vapour deposited (CVD) polycrystalline graphene, adsorptions on grain boundaries introduce severe electron/hole scattering, which degrades the physical properties of the material. Through nucleation density control in a graphite reaction cavity, high-quality graphene single crystals can be grown over large areas. The graphite reaction cavity mimics the Cu-enclosed configuration [1] where sublimated Cu atoms are effectively trapped, thus increasing the Cu vapour pressure inside the cavity. In this way the roughness of the growth substrate can be effectively reduced by Cu re-deposition. Contrary to the Cu-enclosed configuration, our approach does not damage the catalyst substrate and allows upscaling graphene growth on flat Cu foils of arbitrary size. We used a secondary height-controlled sapphire cavity to accommodate the substrate inside the primary graphite cavity and release in situ trace amounts of oxygen that oxidize the Cu substrate and reduce the nucleation density [2]. The oxygen concentration at the Cu surface was further controlled by ex-situ pre-oxidation of the substrate. Using e-beam lithography multiple graphene transistors are patterned and measured sequentially within a single mm-sized crystal. A detailed map of the electronic properties of the single crystal is thus obtained with sub-micrometre resolution. The quality and structure of graphene single crystals are further elucidated by atomic resolution TEM and Raman analysis.

References

[1] X.S. Li, C.W. Magnuson, A. Venugopal et al., J. Am. Chem. Soc., 133 (2011) 2816–2819
[2] Y.F. Hao, M.S. Bharathi, L. Wang et al., Science, 6159 (2013) 720–723

Figures

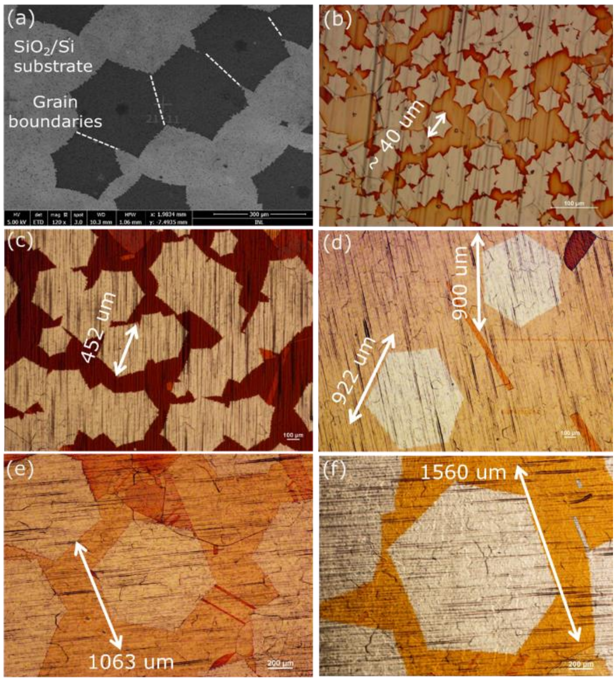

Figure 1: (a) SEM image shows graphene grain boundaries before coalescence. (b) The conventional grain size of CVD-grown graphene is less than 50 μm. (c) The Cu growth substrate is treated by HCl/$FeCl_3$ solution, which helps to reduce defect sites of Cu foil. The reduced nucleation density around 600 nuclei/cm^2 can be achieved. (d) Graphene single crystals with lateral dimension of ~900 μm can be obtained using SiO_2 wafer as a confined slit. (e) and (f) shows the height of the gap affects the nucleation density. The lateral size of graphene single crystal can reach to up to 1500 μm, using Al_2O_3 confined slit with gap height of ~500 μm.