Controlling orientation, edge geometry and thickness of CVD graphene

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The industrial exploitation of chemical vapour deposition (CVD) graphene crucially depends on the ability to generate large-area sheets with selected thicknesses, predefined domain orientations, edge geometries, and grain boundaries which govern the electronic structure (Yazyev, O.V. et al., 2010, Yu, Q. et al., 2011), chemical activity (Sun, Z. et al., 2011), and mechanical strength (Huang, P.Y. et al., 2011). While attention has been directed at understanding the CVD growth mechanism on polycrystalline Cu (Wofford, J.M. et al., 2010), the precise control of CVD graphene remains unsolved (Sun, Z., et al., 2011). Here we show, both experimentally and theoretically, the implementation of structurally defined Cu substrates in conjunction with low-pressure CVD to generate high-quality, tailored graphene. SEM, EBSD mapping and spatially resolved Raman spectroscopy demonstrated that the shape, orientation, edge geometry, thickness, and quality of graphene domains were dictated by the crystallographic orientation of the polycrystalline Cu substrate. Domain edges were observed to preferentially orient to the <110> direction(s) of Cu crystallographic orientations, which was corroborated by computational modelling. The substrate's crystallographic orientation can therefore be exploited to preferentially orient domain edges parallel to the <110> direction while generating high-quality single layer graphene on Cu(111) and bilayer graphene on Cu(001). This research provides a new avenue towards the controlled production of high-quality large-area graphene sheets for implementation in transparent conducting electrodes, sensors, and nanoelectronic device applications.

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