Controlling Growth of 2D Materials on Metals: Defects, Strain, Orientation

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Abstract:
The discoveries of the novel electronic, vibrational, mechanical and optical properties of atomically thin graphene triggered an explosion of interest in this and other two-dimensional (2D) materials, including hexagonal boron nitride (h-BN). Growth of these materials by chemical vapor deposition (CVD) on metal surfaces underpins current capabilities to fabricate epitaxial layers over large areas with atomic layer control and transferability to arbitrary substrates. However, the formation of imperfections such as point and line defects, i.e. grain boundaries, is prevalent during CVD growth that may undermine the exceptional properties of 2D materials that are vital to their applications. Alternatively, defects may exhibit distinct properties of their own that might be harnessed to tailor material properties or alter its functionality.

We have studied the growth/defect structure of graphene and h-BN on metals using low energy electron microscopy (LEEM) and two-dimensional scanning micro-low energy electron diffraction (2D μLEED) measurements. Spot profile analysis of diffraction peaks at every point in the very fine 2D μLEED scan provides detailed quantitative structural information with high spatial resolution. These investigations reveal evidence either of in-plane mosaicity in graphene due to the proliferation of point defects or incommensurability and polymorphism in point defect-free graphene grown on Ru(0001), depending upon growth conditions. Comparative studies of graphene grown on Ir(111), Rh(111) and Ru(0001) indicate coupling of lattice strain and small angle lattice rotations that correlate with the strength of interaction of graphene with the different substrates.

Large-scale single crystal graphene can be produced if the formation of grain boundaries can be avoided, either by increasing the size of individual grains or by the coalescence of aligned grains. We demonstrate that high pressure CVD growth of graphene on Cu(111) without hydrogen produces a single grain orientation, which is favorable for eliminating grain boundaries upon the coalescence of grains. Furthermore, the presence of oxygen on Cu can also produce large individual grains because it greatly reduces the graphene nucleation density and enhances the graphene growth rate. Motivated by these observations, we have explored oxidation of Cu(111) using surface science methods as a foundation for controlling graphene growth. This work revealed a new sub-monolayer oxygen-induced surface structure, with as yet undetermined atomic arrangement, that exhibits exceptional pattern formation and dynamical behaviour. This and other oxygen-induced surface modification of Cu may offer opportunities for the control of graphene grain growth and patterning that is desired.

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Contribution: Invited