On the Fundamental Mechanisms that underpin CVD Technology for Atomically Thin 2D Films

Stephan Hofmann
Department of Engineering, University of Cambridge, United Kingdom

Driven by the industrial demand for "electronic-grade" 2D material films, we focus on developing scalable process technology, and in this talk I will review our recent progress in scalable (MO)CVD and device integration approaches for highly crystalline graphene, hexagonal boron nitride (h-BN) and transition metal dichalcogenide (WS_2) films [1]. The systematic use of in-situ metrology, ranging from high-pressure XPS to environmental electron microscopy, allows us to reveal some of the mechanisms that dictate crystal phase, micro-structure, defects, and heterogeneous integration control at industrially relevant conditions.

While oxidation is widely used to remove impurities in metal catalysts and to control the nucleation density of graphene, we show that minute concentrations of residual bulk oxygen can significantly deteriorate the quality of as-grown graphene highlighted by an increased Raman D/G ratio, increased propensity to post-growth etching and increased fraction of multi-layer graphene nucleation [2]. Starting from commercial Cu foils, we show that a simple hydrogen annealing step after the initial oxidation allows us to lower the residual oxygen level as measured by time-of-flight secondary ion mass spectrometry to produce graphene of significantly higher quality. We devised a new process to achieve wafer-scale high-quality single crystal graphene by combing single-crystal Cu(111) epitaxy on sapphire and graphene growth within a single process flow in a standard commercial reactor. Crucially, we can match this with improved approaches to scalable transfer. We show that such CVD graphene can sustain mobilities of 70000 cm^2/Vs at RT even when initially wet-transferred [3].

We employ a "deconstructed" MOCVD process for WS_2, based on a simple sequential exposure pattern, as model system to systematically explore each growth aspect, including different substrates and their pre-treatment. Using Au support, we find not only allows a significant reduction of the carbon contamination but also a self-limiting behaviour to mono-layer WS_2 with reasonable crystallinity, and full coverage within 10min exposure time. We also report that WS_2 post-growth treatment with oleic acid can greatly enhance the PL yield and mobility [4].

I will also report on our efforts to find new methods for high-throughput quality monitoring of 2D layers, incl. the use of ellipsometric contrast micrography [5] and super-resolution imaging [6]. The latter is particularly interesting to study emissive defect sites in h-BN films. We explore a range of CVD growth, transfer and encapsulation methods, towards controlled emitting-monolayer interfacing and the ability to vertically place emitters with high accuracy. We report on detailed emitter statistics depended on process conditions and how surface interactions heavily influence the photodynamics. We compare this optical characterisation with a range of other h-BN characterisation techniques to develop a holistic understanding.

References

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