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Stranski-Krastanov and Volmer-Weber CVD Growth Regimes to Control Stacking Order in Bilayer Graphene

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Aside from unusual properties of mono-layer graphene, bi-layer has been shown to have even more interesting physics, in particular allowing bandgap opening with dual gating for proper interlayer symmetry. Such properties, promising for device applications, ignited significant interest in understanding and controlling the growth of bi-layer graphene. Here we systematically investigate a broad set of flow rates and relative gas ratio of CH_4 to H_2 in atmospheric pressure chemical vapor deposition of multi-layered graphene. Two very different growth windows are identified. For relatively high CH_4 to H_2 ratios, graphene growth is relatively rapid with an initial first full layer forming in seconds upon which new graphene flakes nucleate then grow on top of the first layer. The stacking of these flakes versus the initial graphene layer is mostly turbostratic. This growth mode can be likened to Stranski-Krastanov growth. With relatively low CH_4 to H_2 ratios, growth rates are reduced due to a lower carbon supply rate. In addition bi, tri and few-layer flakes form directly over the Cu substrate as individual islands. Etching studies show that in this growth mode subsequent layers form beneath the first layer presumably through carbon radical intercalation. This growth mode is similar to that found with Volmer-Weber growth and was shown to produce highly oriented AB-stacked materials. These systematic studies provide new insight into bi-layer graphene formation and define the synthetic range where gapped bilayer graphene can be reliably produced.