Growth of twisted bilayer graphene: Interactions between grains

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Twisted bilayer graphene has a lot of application, like superlattice tunable band gap, hard material. The characteristic is sensitive to the band structure caused by the interlayer twisted angle. We investigate the growth of twisted bilayer graphene through chemical vapor deposition (CVD). Sub-millimeter-sized single crystalline graphene grains with multi-layered adlayer grains formed underneath are identified through optical microscopy. With the spatial mapping of the Raman spectrum, kinds of stacking with different twisted angle are obvious to be distinguished. Through measuring the height, width, and symmetry of the 2D band, the ratio between 2D to G band, the intensity of G band, and the R and R' side-bands, we are able to unambiguously identify the twist angles and categorize them into five types. ARPES offer another way to distinguished the twist angle accurately with van hove singularity. Besides the most abundant AB-stacking (AB-BLG) or large angle (> 15°) twist angle decoupled bilayer graphene (DC-BLG) configurations, there are some bilayer regions that contain specific twist angles (5~8°, 8~13°, and 13.6°) (termed as TBLG). The statistics show that no TBLG is formed for isolated BLG. Furthermore, the formation probability of TBLG is significantly lower when AB-BLG grain merges with a DC-BLG. On the other hand, the probability of TBLG formation is highest when more than two DC-BLG domains merge under high H_2/CH_4 ratio. The growth mechanism of the TBLG is discussed in light of the interactions between the second layer grains