

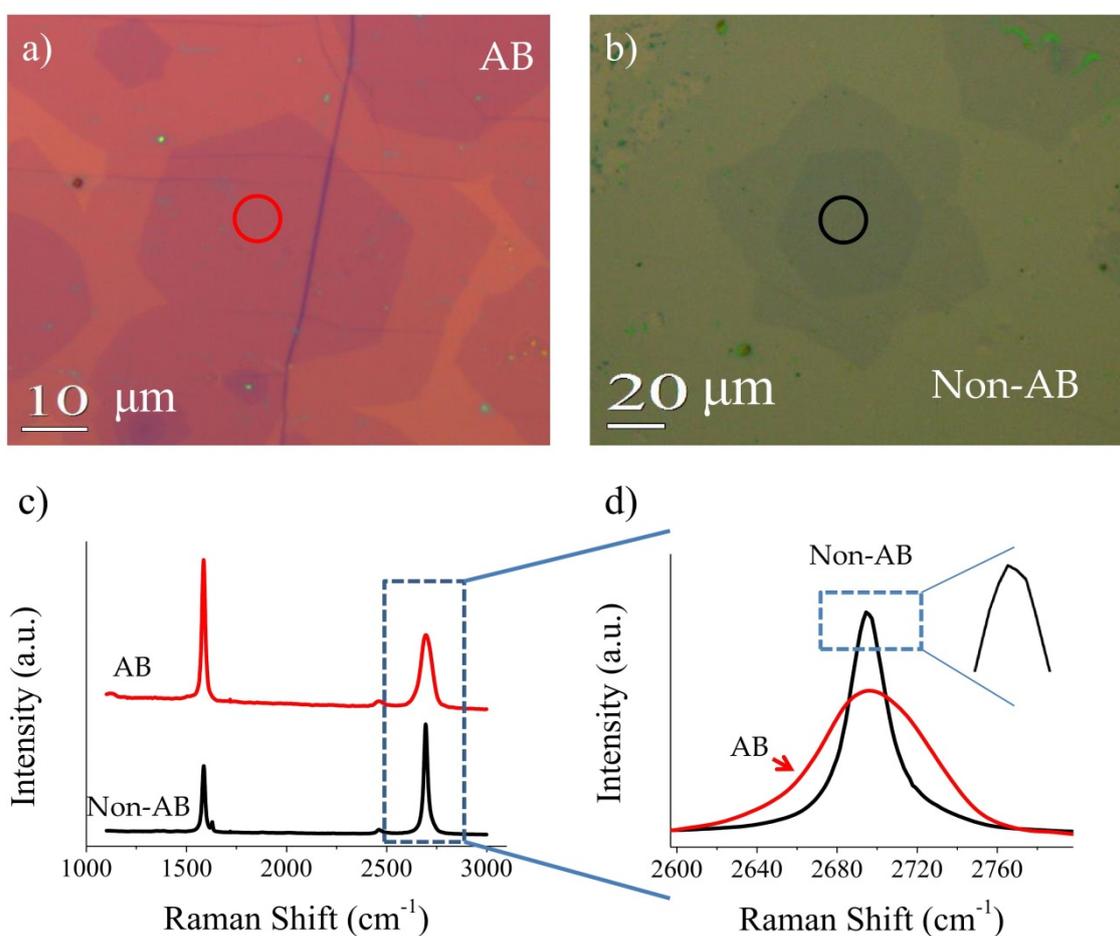
## Electronic Supplementary Material

### Breakdown of Self-limiting Growth on Oxidized Copper Substrate: a Facile Way for Large-size High-quality Bi- and Trilayer graphene synthesis

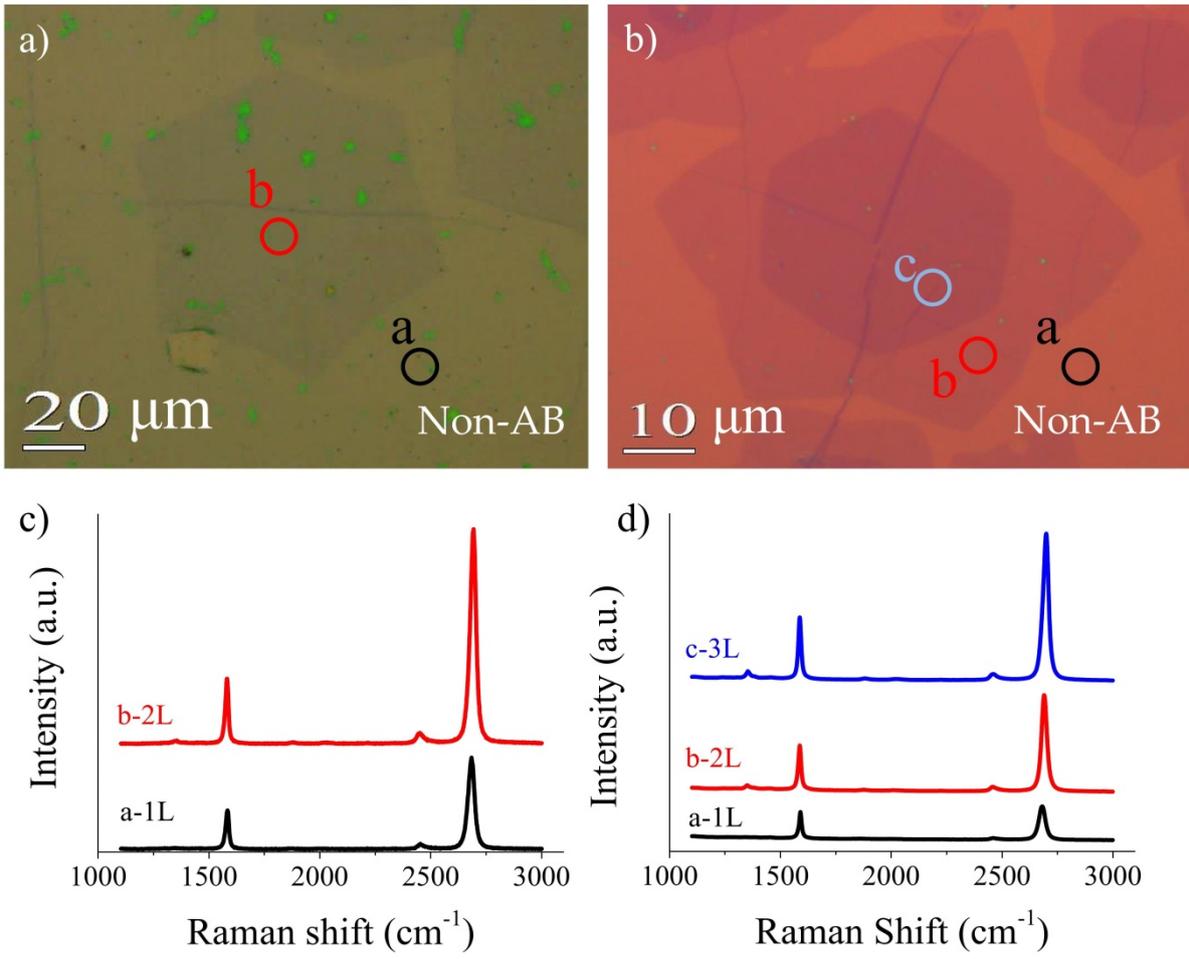
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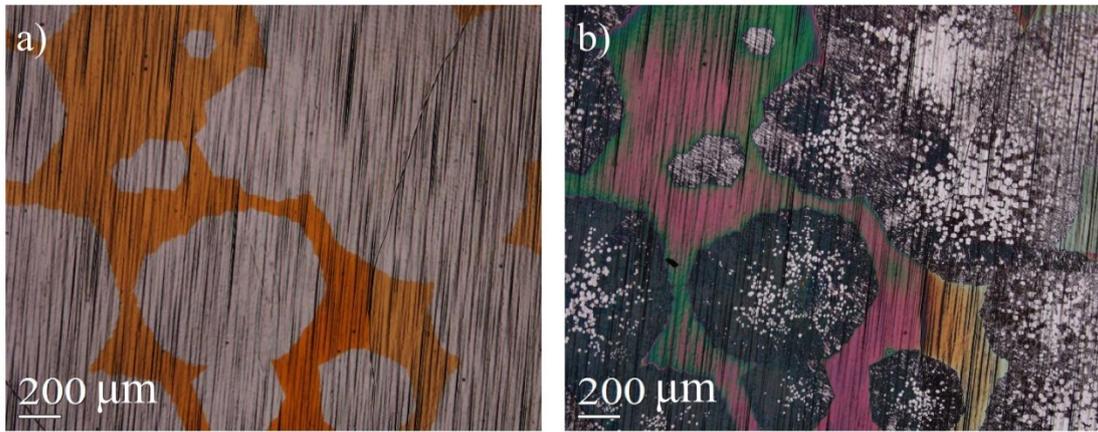
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**Figure S1.** The comparison of Raman spectrum for AB and non-AB stacking trilayer graphene grains. (a,b) optical images for AB and non-AB stacking trilayer graphene, respectively. (c) Comparison of the 3th layer Raman spectrum in AB and non-AB stacking grains. (d) Comparison of the shape of 2D peak in AB and non-AB stacking grains.



**Figure S2.** Optical and Raman spectra for non-AB stacking bilayer and trilayer graphene grains.



**Figure S3.** (a) Optical image for monolayer grains grown on oxidized copper substrate, the whole copper was heated in air to visualize the grains. (b) Optical image for the same sample undergone an improved visualization method to show the nucleuses in monolayer grains.

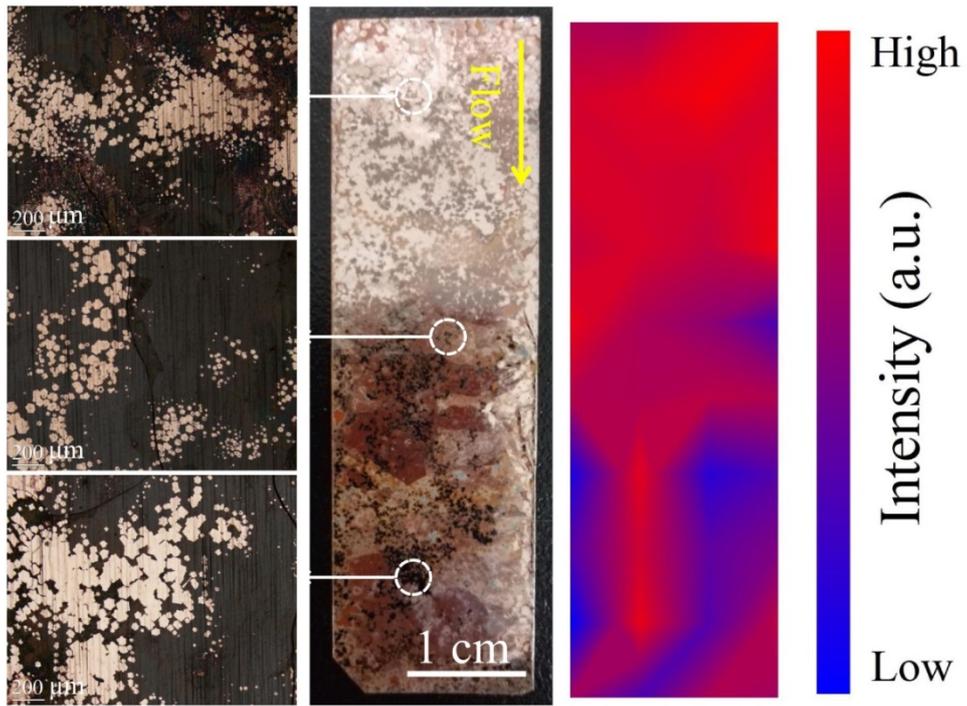
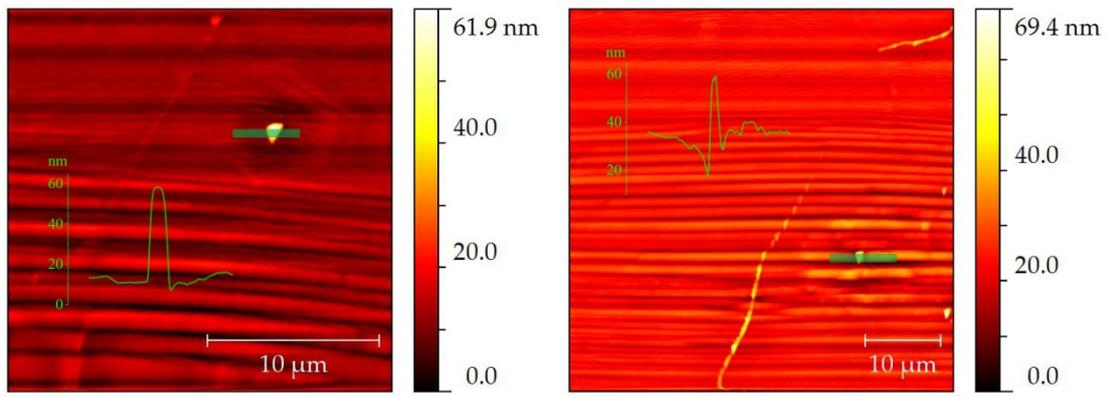
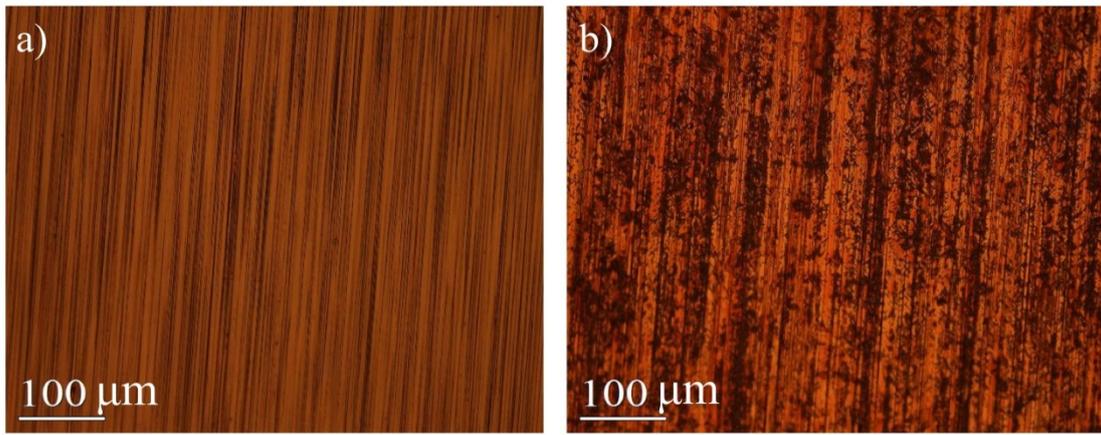


Figure S4. Statistics for the nucleuses on oxidized copper substrate.



**Figure S5.** AFM image for the nucleuses of graphene grains on copper.



**Figure S6.** (a) Optical image for as-received copper substrate. (b) Optical image for polished copper substrate.