The growth of multi-layered graphene flakes in compact millimeter-size single layered graphene grain on copper

Full Title

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Abstract (Century Gothic 11)

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Chemical vapor deposition (CVD) is the major method to produce large scale high quality graphene. Graphene grown on copper through CVD is predominatly single-layered (SLG) because solid solubility of carbon in copper is low. Nevertheless, multi-layered graphene (MLG) can still be found in some cases. Several previous reports demonstrated controllability of coverage of the MLG regions, and paved a new way for band-gap graphene based device application [xxx]. Nevertheless, in those works, the model for MLG formation mechanisms in one study can sometimes be incommensurate with the other studies [xxx]. The above issues cause difficulty for real industrial adaption of the CVD method. It is now a consensus that the grown MLG is an inverted wedding cake structure, in which add-layers are grown underneath an existing SLG grain. However, there are still debates on the source of carbon, nucleation mechanisms, and growth pathways for the underneath MLG flakes. Furthermore, the role of hydrogen in the MLG formation is still not a settled issue. In this work we have found that the nucleation density and coverage areas of MLG underneath a compact milimeter-sized SLG increases as hydrogen partial pressure (P_{H2}) increases or growth temperature (T_{growth}) decreases. Moreover, the distribution of the MLG flakes in a SLG become more uniform when P_{H2} increases or T_{growth} decreases. Our finding is contrary to previous work by xxx et al., in which the SLG grains are fractal like, and the underneath MLG is thought to form via carbon sources diffuse through fratctal grain edges. Instead, our experimental results can be well explained by XXX et al., wherein hydrogen termination at SLG edges determines the MLG growth, and the carbon sources for MLG formation diffuse from the outermost SLG grain edges.

References

[1] M.-C. Chuang, W.-Y. Woon , Carbon., 103(2016), 384
[2] Xiuyun Zhang et al., J. Am. Chem. Soc., 136(2014), 3040

Figures

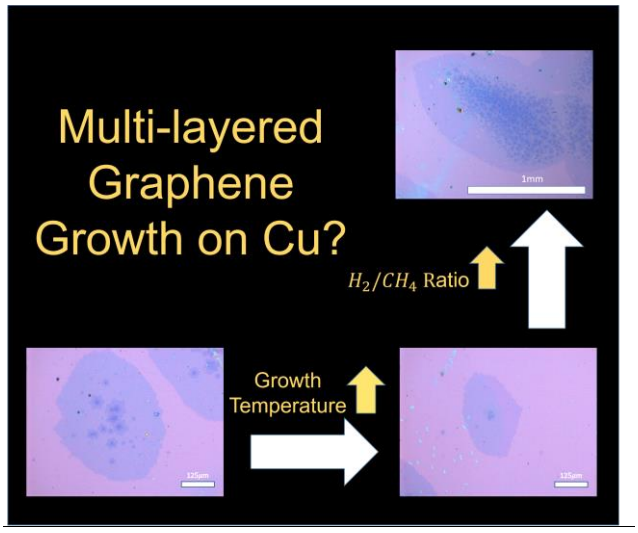
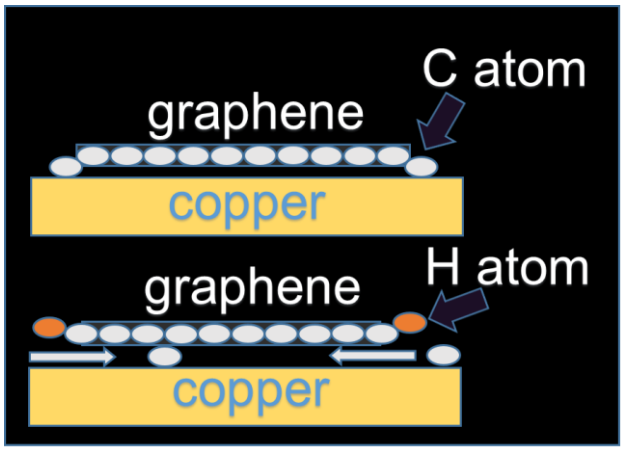


Figure 1: Multilayer region growth with different condition: the multilayer coverage is proportional to the hydrogen/methane ratio

and inversely proportional to the growth temperature.



- **Figure 2:** Active C diffuse from the naked Cu surface to the graphene covered area laterally: Graphene with C-terminated edges which keep the absorption would make grains bigger. With H-terminated edges, graphene gets lifted from the Cu surface and it would be possible to form underlying few-layer regions