Guowen Yuan
Libo Gao
Nanjing University, Hankou road 22, Nanjing, China

gwyuannju@outlook.com

Chemical vapor deposition (CVD) growth Mechanism of graphene monocrystalline

Chemical vapor deposition (CVD) growth of graphene[1-4] becomes the most promising among all the graphene preparation methods for its future high quality applications. Growing higher quality graphene films, including large grain size[5-6], without transfer defect, wrinkle free, etc., still cannot meet the high-performance demands. In the meanwhile, there are plenty of theoretical results to predict or explain the growth process in atomic level[7-9]. Most of their results cannot be easily realized by experimental works. Here, we investigate their mechanism experimentally by comparing growing graphene on different substrates, including rough commercial polycrystalline Cu foils and single crystalline Cu films which sputtered on sapphire substrates. When fast growing individual graphene grains on Cu foils, we found that there are always some smaller grains around the existent larger grains. The larger grains usually show no hexagonal shape, but dendrites combined with smaller grains. Considering the metal surface perform quasi-liquid state at the growth condition (more than 1000 °C), the grown graphene grains are nearly floating on the surface, so the larger grains should come from the combination of plenty of smaller grains. It is worth noting that most grains show the nearly the same orientation before their joint, and the deviation should origin from the rough surfaces. The growth on single crystalline Cu(111) films show the similar growth behavior, but the smooth surface result in much higher ordered individual grains before joint, and nearly no dendritic grains are formed. Therefore, we suppose nearly all shape of formed graphene domains should be combined with plenty of tiny hexagonal grains, not from the carbon atoms decomposed from in CH4 directly. These smoother substrate surfaces performed much promising growth results in large scale single crystalline graphene films. This result should provide a guideline to the fast growth of wrinkle free graphene single crystalline films.

References

[1] Reina A et al. 2009 Nano Lett. 9 30
[2] Kim K S et al. 2009 Nature 457 706
[3] Li X S et al. 2009 Science 324 1312
[4] S. Bae et al. 2010 Nat. Nanotechnol 5 574
[5] L Li et al.2016 Acs Nano 10 2922
[6] Wu T et al. 2015 Nat. Mater.15 43
[7] Yuan Q, Yakobson B I, Ding F. 2014 J. Phys. Chem. Lett. 5 3093
[8] Shu H, Chen X, Tao X, et al. 2012 Acs Nano, 6 3243
[9] ZhangX, Li H, Ding F. 2014 Adv. Mater., 26 5488

Figures

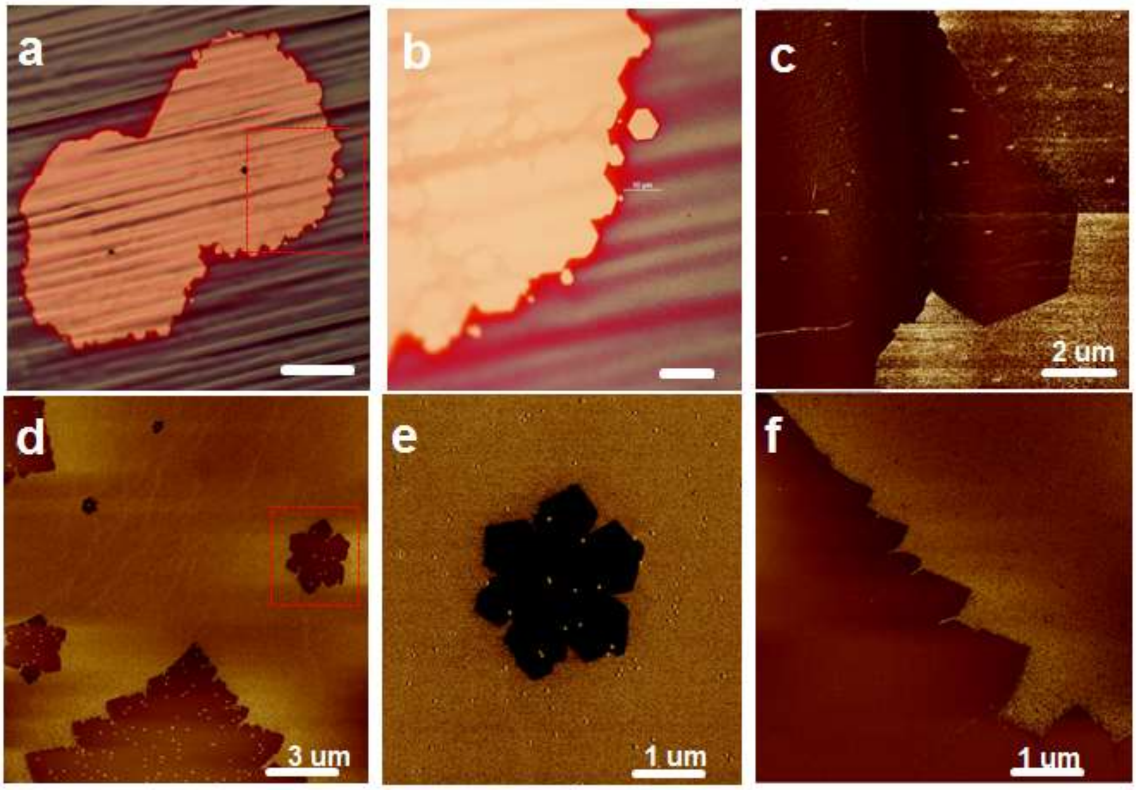

Figure : Graphene grain grown on copper foils and oxidized Cu/Ni substrates (a-b) Optical image of graphene grain on copper foils , which were oxidized at 5 min under 200 ºC. (c) AFM phase image of the grown grain in (b). (d-f) AFM phase images of graphene grain ,which grown on oxidized single crystalline Cu/Ni film

.