Yuting Hou
Longlong Zhan, Bojun Wang, Xiaobin Niu, Fangzhu Qing, Xuesong Li
University of Electronic Science and Technology of China, No.4,Section 2,North Jianshe Road,Chengdu,P.R.China
lxs@uestc.edu.cn

Oriented Graphene Domain Growth on Non-Cu(111) Surface

Since graphene was successfully separated from the bulk in 2004, it has attracted significant scientific and industrial interests because of its excellent properties and many promising applications. Up to date high quality graphene films are commonly synthesized on Cu substrate by chemical vapor deposition (CVD) method[1]. Mostly, the graphene films prepared in this way are polycrystalline and the grain boundaries are one of the major defects. Thus to eliminate grain boundaries is prerequisite to achieve high quality graphene, which can be realized with two strategies. One is to reduce the nucleation density so as to grow large-area single crystals from individual nucleation sites, the so-called the single-core method[2-4]. Various strategies have been developed to reduce the nucleation density and increase growth rate such as by using smooth substrate surface, introducing oxygen, carefully controlling the feedstock[3, 5]. However, practically it usually requires the film size in wafer scale or even larger while it is difficult to define just one nucleation site in such a large scale and the growth rate is also quite limited.

The other is to simultaneously grow multiple oriented graphene domains, which finally seamless stitch together to form a grain boundary free film, the so-called multi-core method[6]. Compared with the single-core method, the multi-core method is more efficient. The key is how to control the orientation of graphene domains. It has been reported that oriented graphene domains can be achieved on some specific facets, i.e., Ge (110)[7] and Cu (111)[6]. On other facets such as Cu (110) and Cu (100), the graphene domains prefer to multiple optimal orientations[8]. Restriction to specific crystal facets is both cost and difficult. Even on the preferred facets such as Cu (111), mis-oriented graphene domains have also been observed while the criteria to control the orientation is still mysterious.

Here, we report that oriented graphene domains can be grown on almost any Cu facets except for Cu (111). We have investigated the criteria to control the orientation of graphene domains and explained the mechanisms by density functional theory. This discovery not only provides a new way for the preparation of graphene single crystals, but also plays an important role in the development of other two-dimensional materials (such as h-BN) single crystal preparation.

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Figure

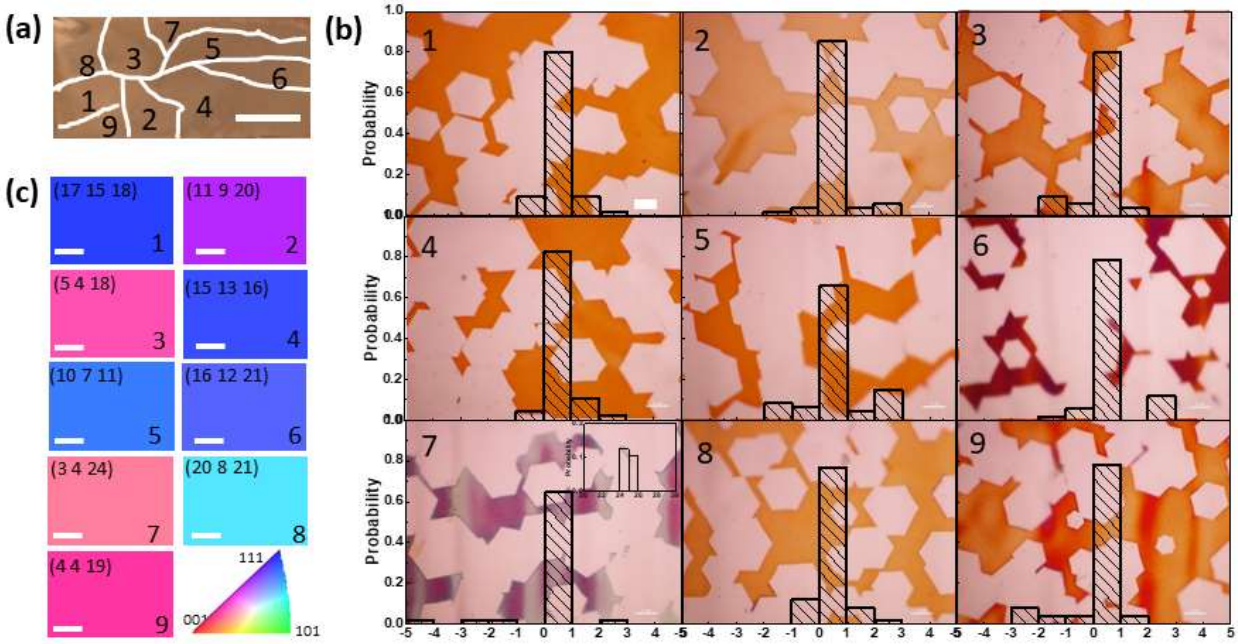

Figure 1: (a) The photo of a piece of Cu with 9 grains. The Cu grain boundaries are illustrated with the white lines. (b) Optical Microscopy images of the oriented graphene domains on corresponding numbered Cu grains. The substrate was slightly heated in air to oxidize the exposed Cu so as to enhance the optical contrast between graphene domains (the white regions) and the exposed Cu (the colored regions). The insets show the statistics of the rotational angles of graphene domains. (c) EBSD results show the surface indices of corresponding numbered Cu grains. The scale bars in (a)-(c) are 1 cm, 20 μm, and 100 μm, respectively.