Seamless stitching of graphene domains on polished copper (111) foil

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Abstract
Graphene grain boundaries (GGBs) are inevitably formed via stitching of graphene flakes, consequently limiting the graphene quality. There have been numerous reports that GGBs are a primary carrier scattering source, thus degrading the related device performance. Therefore, it is always desired to obtain large-area graphene without forming GGBs. Two approaches exit for to synthesizing monocrystalline graphene. One approach is to reduce the number of nucleation seeds. However, large-area growth is terminated by an unknown self-limiting growth factor and requires long growth time. Another approach entails the alignment of graphene domains while leading them to stitch together to form uniform single monocrystalline graphene. Thus, seamless stitching of graphene domains during chemical vapor deposition (CVD) is an ideal concept to realize large-area monocrystalline graphene. Cu(111) substrate maintains hexagonal symmetry with minimum lattice mismatch with graphene and has been tried to grow graphene but no evidence for seamless stitching has been provided. Use of copper substrate is technologically relevant, since monolayer graphene is easily tailored due to the limited carbon solubility in copper, and moreover the surface morphology can be controlled at large area with low cost. In this work, we prove the concept of seamless stitching without forming GGBs by preparing a polished Cu(111) foil for CVD. The seamless stitching was realized by merging hexagonal graphene domains in the same orientation and verified by not only at atomic scale by scanning tunneling microscopy (STM) and transmission electron microscopy (TEM) but also at macro-scale by optical microscopy after UV-treatment[1]. This was markedly distinct from the clear GGBs formed by the similar hexagonal domains in different orientations, in congruent with our density-functional calculations. This concept was extended to synthesize monocrystalline graphene of 3x6 cm² size by CVD for an hour by merging multiple hexagonal graphene domains in the same orientation on Cu(111) foil. The monocrystallinity of the large-area sample was confirmed by new observation method achieved by correlating confocal Raman mapping on overlapped graphene bilayers to polarized optical microscopy (POM) on spin-casted nematic liquid crystal (NLC) layer, combined with transport measurements at the stitched region.

References

Figures