## **Controlled Removal of Monolayers for Bilayer Graphene**

## **Preparation and Visualization**

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Figure S1. The evolution of grains and nucleus on copper during the high temperature heating in air.



**Figure S2. Comparison of visualized nucleus in grains by long-time natural oxidation and high temperature heating in air.** (a, b) Visualized nucleus in grains on copper by long-time natural oxidation and enlarged image in nucleus sites. Broken sites were concentrated in the bulk of nucleus. (c, d) Visualized nucleus in grains on copper by high temperature heating in air and enlarged image in nucleus sites. Broken sites were concentrated along the edges of nucleus.



**Figure S3. Comparison of pristine CVD-grown grains on copper and on oxidized copper surface**. (a) Grains on copper. Rippers can be easily observed in most large sized graphene grains. (b) Profile along the red line in Fig. S5a. (c) Grains on oxidized copper. Large-sized copper oxide can be easily observed on grain surface. (d) Profile along the yellow line in Fig. S5c.



**Figure S4. Visualization of nucleus in grains on copper**. (a, b) SEM images for grains on copper and enlarged image for nucleus in center. (c, d) Optical images for the same grains on mild oxidized copper and enlarged image for center. (e, f) Optical images for the same grains on heavily oxidized copper and enlarged image for nucleus.



Figure S5. Optical images for individual grains with more than one nucleus.



Figure S6. XPS spectrum of oxidized copper surface (core-level Cu 2P<sub>3/2</sub>).