

## *Supporting Information*

### **Facile Growth of Centimeter-Size Single-Crystal Graphene on Copper Foil at Atmospheric Pressure**

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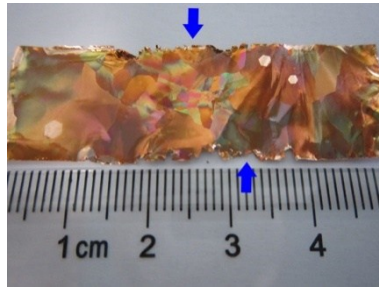


Figure S1. Optical photograph of Cu foil, which was grown for 3h without hydrogen reducing the Cu oxide before growth. Blue arrows mark the over-oxidized and etched Cu foil edge.

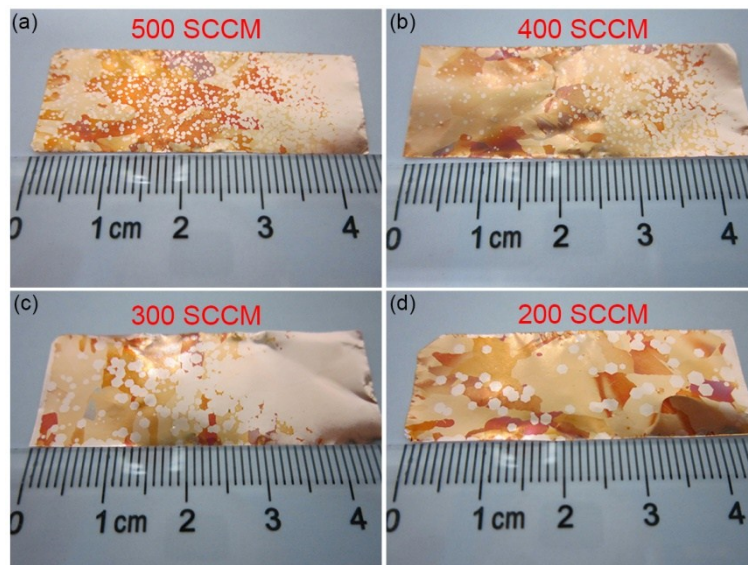


Figure S2. Optical photograph of graphene (bright color) on Cu with respect to total gas flow rate marked in each images, respectively.

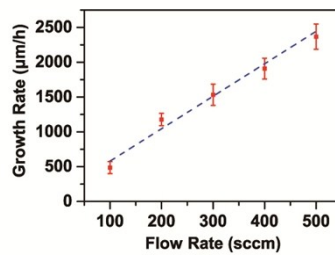


Figure S3. Plot of graphene growth rate relative to the total gas flow rate. The collected data are plotted as red squares, while the linear fitting curve is indicated by blue dashed line.

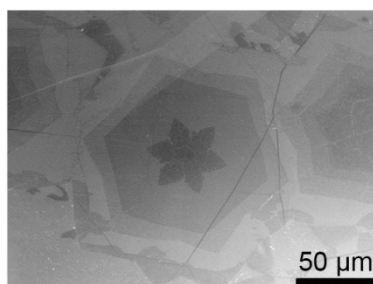


Figure S4. The SEM image of the typical multi-layer graphene structure on Cu substrate.

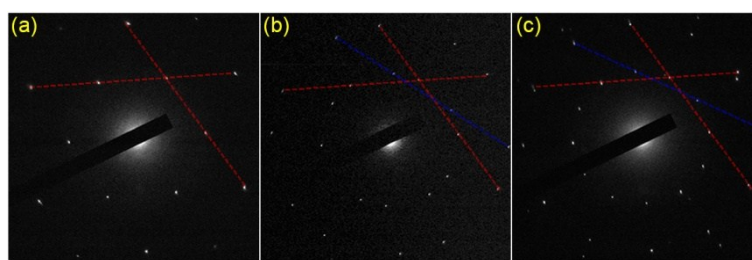


Figure S5. SAED pattern of the multi-layer graphene structure. Even though the multi-layer orientation (as blue dashed line shows) is uncontrollable, the monolayer graphene orientation (as red dashed line shows) is relatively uniform.

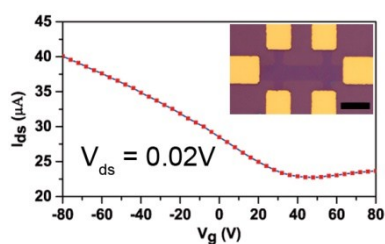


Figure S6. Plot of drain current ( $I_{ds}$ ) versus gate voltage ( $V_g$ ) of graphene based FET in air. Insert is the optical microscope image of the back-gate graphene FET, scale bar is 10  $\mu m$ .

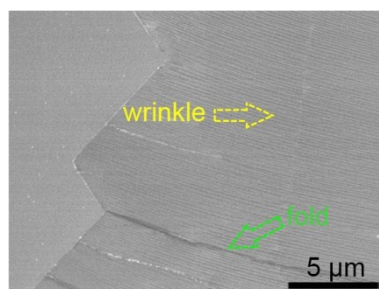


Figure S7. SEM image of the graphene wrinkle and fold on Cu surface.

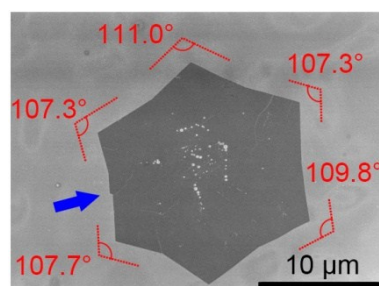


Figure S8. The SEM image of a 10 $\mu m$ -size graphene flake on Cu substrate, the degree of GC is labelled, and the newly formed GC on graphene edge is indicated by the blue arrow.

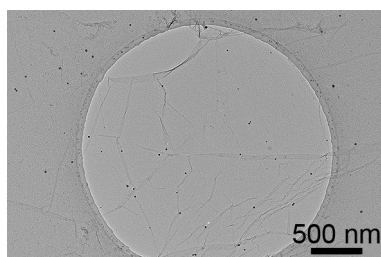


Figure S9. High resolution TEM image of graphene as marked by blue spot in Figure 4c.

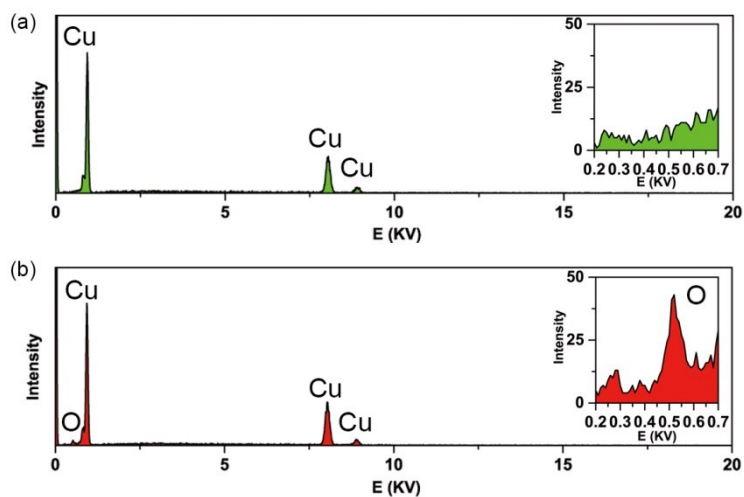


Figure S10. The EDS spectra of the Cu crystal surface (as the green cross marks in Figure 5a) and Cu crystal boundary clusters (as the red cross marks in Figure 5a), respectively.

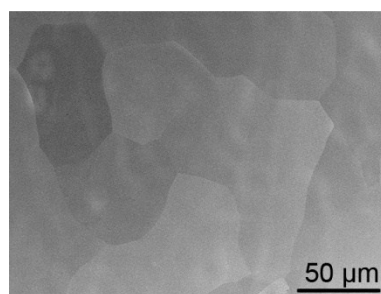


Figure S11. The SEM image of the Cu surface which was taken out from the furnace right after heated in Argon gas for 15 minute and then reduced in H<sub>2</sub> for about 1 minute, respectively.

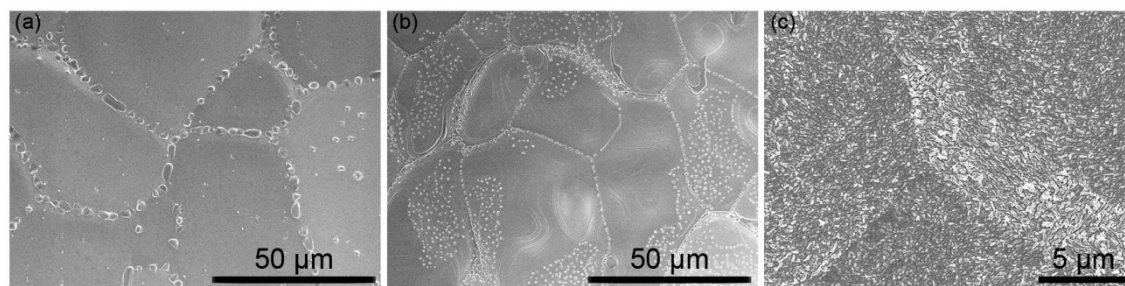


Figure S12. The SEM image of Cu surface after heated in high purity Argon (a), high purity Argon from room temperature to 1050 °C in 15 min and 1050 °C min for another 15min (b), high purity Argon from room temperature to 1050°C in 15 min and 500 ppm O<sub>2</sub> at 1050 °C for another 12 h (c), respectively.