# Supporting Information

# **Directional Copper Dewetting to Grow Graphene Ribbons Arrays**

Xiaogang Li,<sup>a,#</sup> Xuyao Xiong,<sup>a,#</sup> Congcong Ning,<sup>a,#</sup> Qian Yang,<sup>a,b</sup> Dongling Li,<sup>c</sup>

Zegao Wang,<sup>d</sup> Yan Jin,<sup>a</sup> Wenbin Zhao,<sup>a</sup> Baoshan Hu \*,a,c

<sup>a</sup>School of Chemistry and Chemical Engineering, Chongqing University, Chongqing, 401331, China

<sup>b</sup>School of Metallurgy and Materials Engineering, Chongqing University of Science and Technology, Chongqing, 401331, China

<sup>c</sup>Defense Key Disciplines Lab of Novel Micro-nano Devices and System Technology, Chongqing University, Chongqing, 400044, China

<sup>d</sup>College of Materials Science and Engineering, Sichuan University, Chengdu, 610064, Sichuan Province, China

<sup>#</sup>The authors contributed equally.

\*To whom correspondence should be addressed: hubaoshan@cqu.edu.cn (B.S. Hu)

#### **Experimental Materials and Methods**

#### 1 Fabrication of V-grooved Substrates

The V-grooved structures on Si(100) were fabricated by lithography patterning and anisotropic etching followed by thermal oxidation. And the Cu film in the grooves was prepared by a conventional lift-off process with UV lithography (MA6/BA6, Suss).

#### 2 Synthesis of Graphene Ribbons

A substrate was placed onto a quartz holder and loaded outside the heating zone of tube furnace. Then the furnace was heated to desired temperatures with a mixed gas flow of argon (300 sccm) and hydrogen (10 sccm) under ambient pressure. The substrate was immediately loaded into the center of the heating zone to perform rapid heating. After annealing for several minutes, 1 sccm  $CH_4$  was introduced into the CVD system for GNRs growth and the growth time was typically 5 min. Following that, the substrate was moved from the centre to the outside of the heating zone for rapid cooling in a gas flow of  $H_2$  (10 sccm)/Ar (300 sccm).

## 3 Transfer of Graphene Ribbons

The as-grown graphene ribbons on the Cu bars were spin-coated with polymethyl methacrylate (PMMA, Sigma Aldrich) and then baked at 150 °C for 5 min, followed by the etching away of the SiO<sub>2</sub> layer in 10% HF, resulting in a PMMA/ graphene ribbons/Cu stack floating on the surface of the etchant solution. Subsequently, the PMMA/graphene ribbons/Cu stack was floated on 1 M FeCl<sub>3</sub> solution in 10% HCl to etch the Cu, and then washed with DI water repeatedly. The stack of PMMA/graphene ribbons was transferred a SiO<sub>2</sub>/Si wafer and heated to 150 °C for 5 min. The PMMA is dissolved in acetone, leaving graphene ribbons on the SiO<sub>2</sub>/Si wafer.

## 4 Characterizations

The morphology and structure of Cu bars was characterized using scanning electron microscopy (SEM) (JSM-7800F, JEOL). Transmission electron microscopy (TEM) images and selected area electron diffraction (SAED) patterns of the transferred graphene ribbons on a carbon support film grid were measured with a Talos F200S (FEI). Raman mappings of graphene ribbons were measured with a LabRAM HR Evolution (HORIBA Scientic) using a green laser of 532 nm, spot size of 0.7  $\mu$ m and step of 0.1  $\mu$ m. Photoluminescence (PL) was measured in a home-built optical UHV chamber with the same Raman system using 532 and 325 nm laser at 300 K. A joint Atomic force microscopy (AFM)-Raman apparatus was employed to reflect the morphological and spectroscopic properties from same one graphene ribbon over a NanoXplora platform with an AFM tip of APPNano and 532 nm laser with a spot size of 0.7  $\mu$ m.



Fig. S1. The cross-sectional SEM images of V-grooves after depositing Cu films for different sputtering time: (a) 5 min, (b) 6 min, (c) 7 min, (d) 8 min, (e) 9 min, (f) 10 min and (g) 15 min. (h) The fitting linear relationship of film thickness as a function of sputtering time.



Fig. S2. Heating rates of three heat modes, (a) slow heating rate at around 0.25 °C/s from the room temperature in line with the heating zone of CVD chamber, (b) moderate heating rate from room temperature directly to 700 °C at around 33.24 °C/s and then to 1090 °C at around 0.25 °C/s, and (c) rapid heating rate at around 76.42 °C/s from room temperature directly to 1090 °C. The heating rate is measured with a calibrator thermometer (OMEGA, CL3515R).



Fig. S3. Effect of heating rate of Cu film on the dewetting behavior. (a) The as-received Cu film before loading into the CVD chamber, (b) The Cu film is heated from the room temperature in line with the heating zone of CVD chamber, (c) The Cu film is heated while the heating zone of CVD chamber reaches 700 °C, (d) The Cu film is heated while the heating zone of CVD chamber reaches 1090 °C.



Fig. S4. Topview SEM images of annealed substrates loaded with different angles against the horizontal direction, (a) 30°, (b) 90° and (c) 180°.



Fig. S5. SEM images of the substrates with Cu sputtering time of 8 min after annealing at different temperature (a) 1050 °C and (b) 1090 °C.



Fig. S6. (a) PL of graphene ribbons on Cu (black curve) and on  $SiO_2/Si$  (red curve) excited by 532 nm laser at 300 K. (b) PL of graphene ribbons on Cu (black curve) and on  $SiO_2/Si$  (red curve) excited by 325 nm laser at 300 K.