

## Supporting Information

# Electrochemical Potential Stabilization of Reconstructed Au(111) Structure by Monolayer Coverage with Graphene

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**EC-STM image of the moiré pattern and its simulated structure of the monolayer graphene grown on Au(111)**

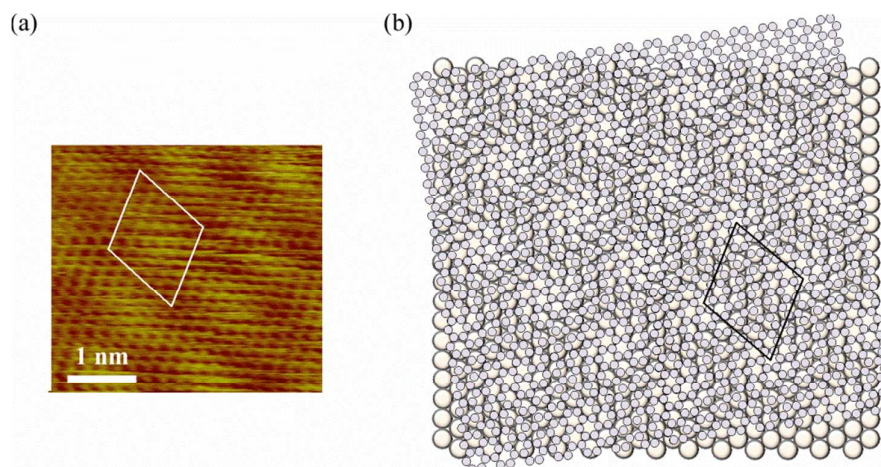


Fig. S1. (a) EC-STM images of graphene grown on Au(111) in 0.1 M H<sub>2</sub>SO<sub>4</sub> aqueous solution and (b) its possible moiré structure. STM image was taken at  $E_{\text{bias}} = -0.02\text{V}$ ,  $E_{\text{sample}} = -0.02\text{V}$  and  $I_t = 2.0$  nA. The angle between the graphene and Au(111) lattices was estimated to be 5 °.

### Estimation of graphene coverage for graphene grown on Au(111) electrode

Graphene coverage for graphene grown on Au(111) electrode was calculated by Au oxidation and reduction method. Briefly, uncovered Au area in graphene/Au electrochemically oxidized, and then the reduction peak of the formed Au oxide was measured. Since the reduction peak is attributed to the reduction of Au oxide on uncovered Au(111) surface<sup>S1</sup>, the ratio of the reduction peak area for graphene grown on Au(111) electrode to that for bare Au(111) electrode corresponds to uncovered Au surface area. Hence, graphene coverage can be calculated by following equation.

$$A (\%) = \left(1 - \frac{Q_{g/Au}}{Q_{Au}}\right) \times 100 \quad \dots (1)$$

where  $A$  is graphene coverage, and  $Q_{g/Au}$  and  $Q_{Au}$  are measured reduction peak areas for graphene grown on Au(111) and bare Au(111) in CV, respectively. Measured  $Q_{g/Au}$  and  $Q_{Au}$  were  $9.949 \times 10^{-8}$  (C) and  $1.841 \times 10^{-5}$  (C), and consequently graphene coverage was estimated to be 99.5 (%).

It is to be noted that the Au oxidation process also causes the oxidation of the graphene as shown Fig. 5(b)IV and produces “pit defects”, result in lowering of the graphene coverage. Hence, the graphene coverage must slightly underestimate. However, we found that the number of formation of the pits is very small if only once oxidation process, and consequently the rough estimation are possible.

### Retaining of reconstructed Au(111) structure covered with graphene for over half a year

Synthesis of graphene on Au(111) : Nov. 6, 2014

STM measurements : August 3, 2015

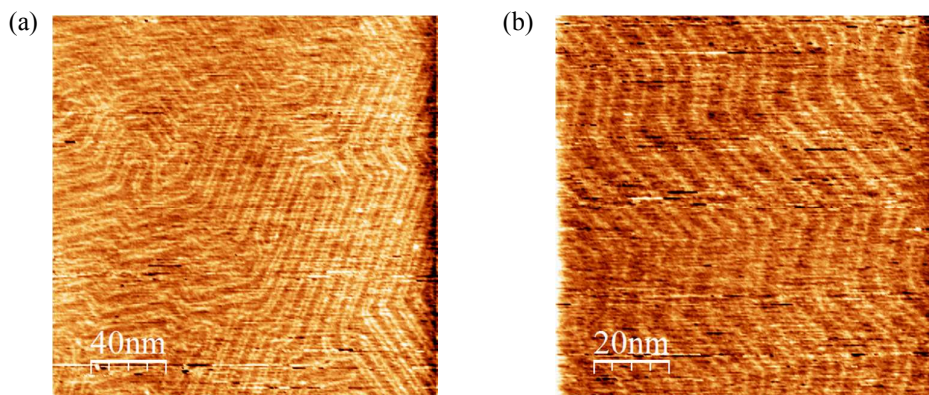


Fig. S2. Representative STM images of graphene grown on Au(111) after half a year passed.

(a)  $E_{\text{bias}} = -15$  mV,  $I_t = 1.15$  nA (b)  $E_{\text{bias}} = -15$  mV,  $I_t = 1.20$  nA

**EC-STM images after the potential was swept negatively from the oxidation range**

(Images was obtained at different location of Fig. 5(b))

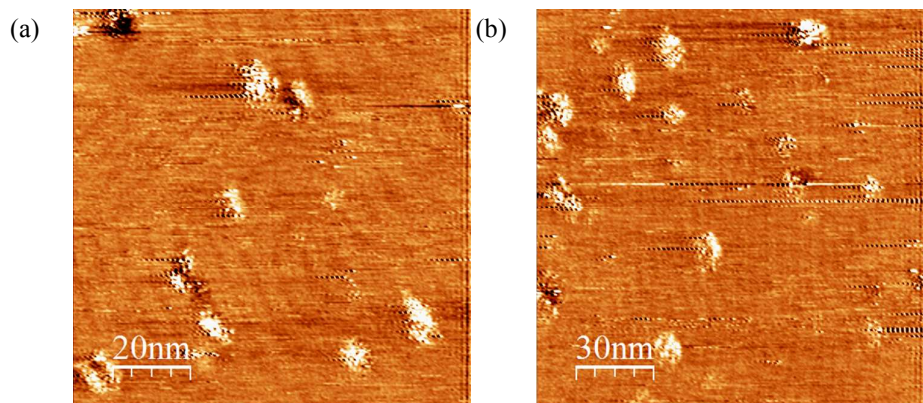


Fig. S3. Representative EC-STM images for the graphene grown on Au(111) in 0.1 M H<sub>2</sub>SO<sub>4</sub> aqueous solution.  $E_{\text{sample}}$  were (a) 0.575 V and (b) 0.705 V. All images were taken at  $I_t = 2.0$  nA and  $E_{\text{bias}} = -0.02$  V.

**Reference**

- (S1) Angerstein-Kozłowska, H.; Conway, B. E.; Hamelin, A.; Stoicoviciu, L. Elementary Steps of Electrochemical Oxidation of Single-Crystal Planes of Au – I. Chemical Basis of Processes Involving Geometry of Anions and the Electrode Surfaces *Electrochim. Acta.* **1986**, 31, 1051-1061.