

# Supporting Information

## Influence of SiO<sub>2</sub> or h-BN substrate on the room-temperature electronic transport in chemically derived single layer graphene

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### Experimental Section

#### 1. Synthesis of oxo-G

Oxo-G was prepared by the optimized oxidation of graphite at low temperature (< 4 °C), using potassium permanganate as oxidant in sulfuric acid, as reported before by our group.<sup>1</sup>

#### 2. Langmuir-Blodgett films of oxo-G

Flakes of oxo-G were deposited onto the Si/SiO<sub>2</sub> substrate by Langmuir–Blodgett technique<sup>2</sup> (LB, Kibron  $\mu$ trough, 3 mN m<sup>-1</sup> with the surface tension of water as reference value of 72.8 mN m<sup>-1</sup>). Reduction was performed by vapor of hydriodic acid and trifluoroacetic acid (1/1 mixture by volume) at 80 °C (10 min). Subsequently, the surface of graphene was cleaned with doubly distilled water (Carl Roth) to remove iodine species, which are adsorbed on <sup>0.5%</sup>G flakes.

#### 3. Fabrication of h-BN/<sup>0.5%</sup>G heterostructures device

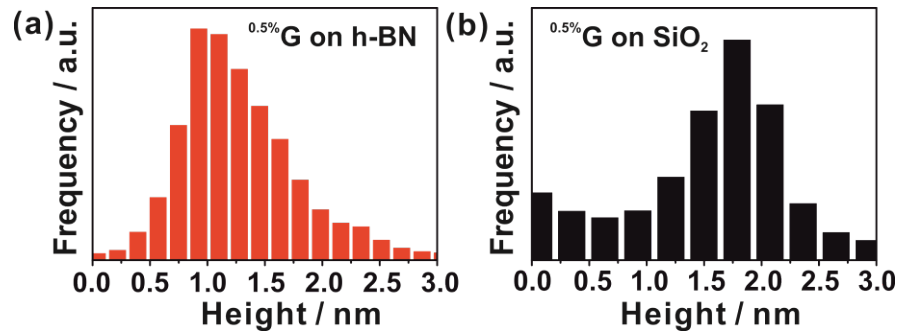
In order to make h-BN/<sup>0.5%</sup>G heterostructures, a h-BN flake (obtained from Taniguchi group, Japan) was mechanically exfoliated onto the Si/SiO<sub>2</sub> substrate by scotch tape method.<sup>3</sup> The <sup>0.5%</sup>G flake was transferred onto Si/SiO<sub>2</sub>/h-BN surfaces with Polydimethylsiloxane (PDMS, Gel-pack) as a carrier.<sup>4</sup> Then, the h-BN/<sup>0.5%</sup>G heterostructure with the same monolayer <sup>0.5%</sup>G flake partially covered on SiO<sub>2</sub> and h-BN surfaces was

finished. Patterning of the electrode structure was achieved by standard electron beam lithography processing (Raith PIONEER TWO). The 5/70 nm Cr/Au electrodes were deposited by thermal evaporation (Kurt J. Lesker NANO 36).

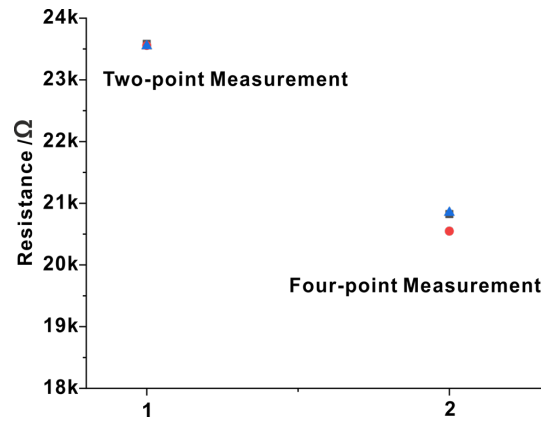
#### 4. Measurements

Optical imaging was performed using a Nikon LV100ND. Scanning tunnelling microscopy (STM) and Scanning tunnelling spectroscopy (STS) were performed with an ultra-high vacuum ( $< 10^{-10}$  mbar) STM (Omicron-STM1). AFM characterization was performed using a JPK Nanowizard 4 atomic force microscope in tapping mode at room temperature. Statistical Raman spectroscopy was recorded using a confocal Raman microscope (Horiba Explorer, 532 nm excitation wavelength). Electrical measurements were carried out at ambient conditions using a probe station with micromanipulated probes and two source-measurement units (Keithley 2450). In addition, comparing experiments were carried out at vacuum (HI-CUBE) using Lakeshore probe station.

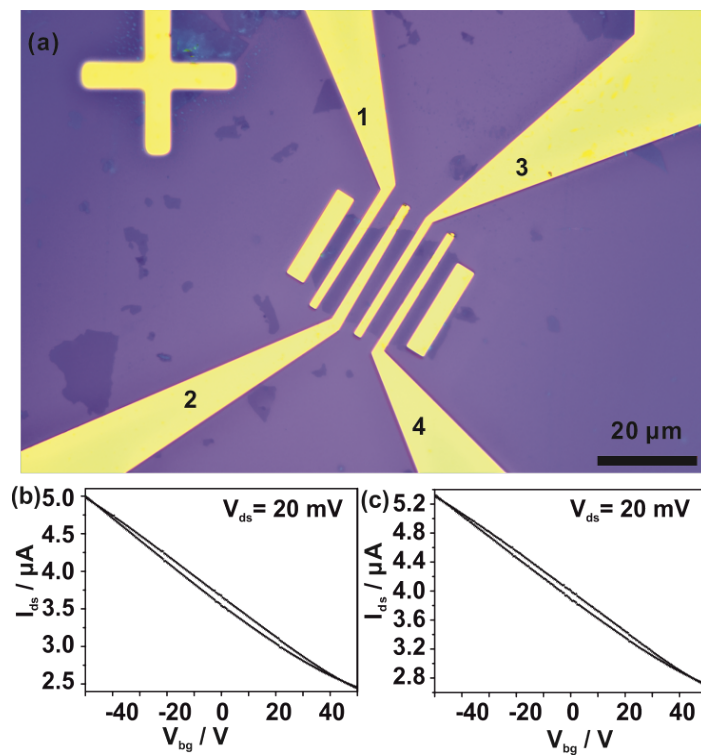
#### Results



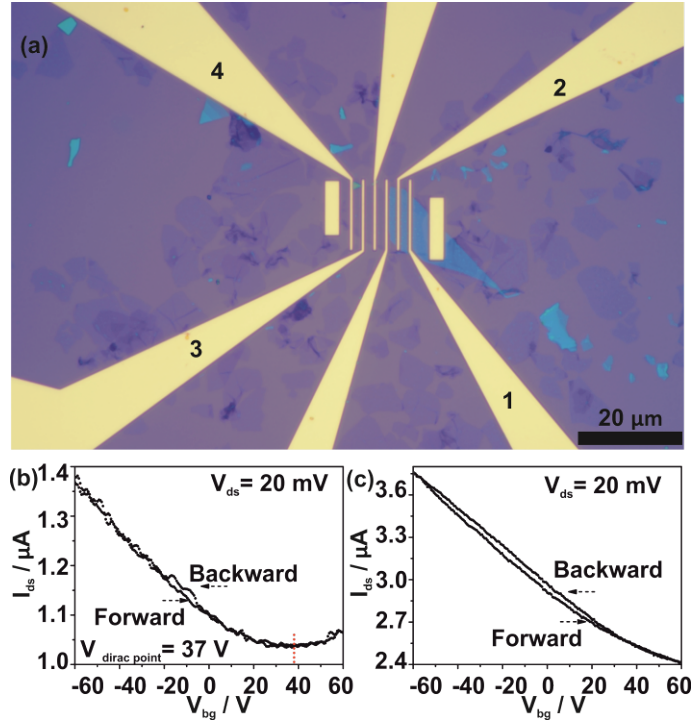
**Fig. S1.** (a) and (b) Histogram of the height distribution measured by AFM for the 0.5%G on h-BN and SiO<sub>2</sub>.



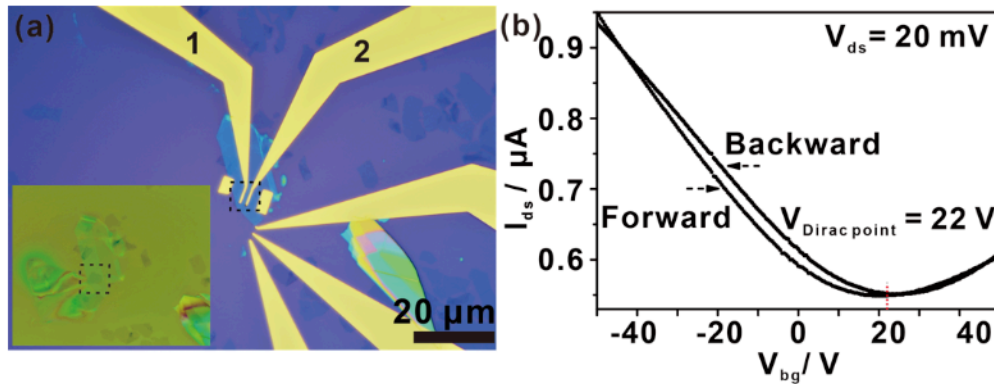
**Fig. S2.** Measurements of resistance of monolayer  $^{0.5\%}\text{G}$  in two-point and four-point configuration, respectively.



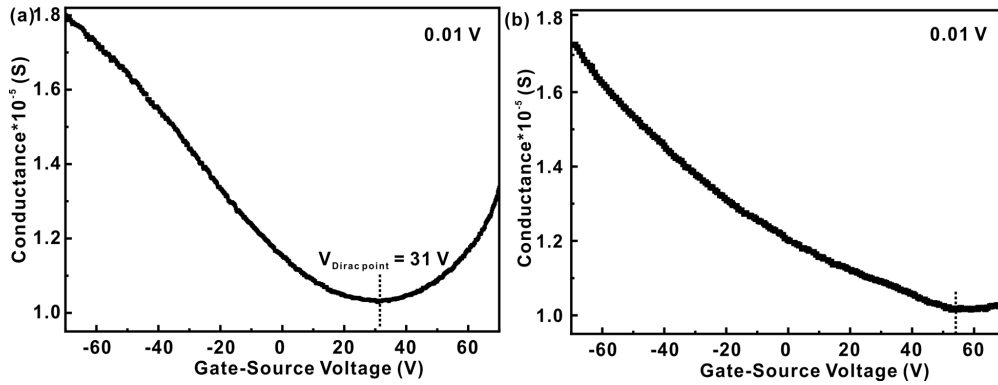
**Fig. S3.** (a) Optical image of the fabricated monolayer  $^{0.5\%}\text{G}$  device with back-gate. (b) Electrodes 1 and 2 were measured for transfer characteristics for the  $^{0.5\%}\text{G}$  on  $\text{SiO}_2$ . Channel length and width between 1 and 2 is 3  $\mu\text{m}$  and 24  $\mu\text{m}$ , respectively. (c) Electrodes 2 and 3 were measured for transfer characteristics for the  $^{0.5\%}\text{G}$  on  $\text{SiO}_2$ . Channel length and width between 2 and 3 is 3  $\mu\text{m}$  and 24  $\mu\text{m}$ , respectively.



**Fig. S4.** (a) Optical image of the fabricated h-BN/<sup>0.5%</sup>G and SiO<sub>2</sub>/<sup>0.5%</sup>G heterostructures device with back-gate. (b) Electrodes 1 and 2 were measured for the <sup>0.5%</sup>G on h-BN. Channel length and width between 1 and 2 is 2 μm and 15 μm, respectively. Electrodes 3 and 4 are measured for the <sup>0.5%</sup>G on SiO<sub>2</sub>. Channel length and width between 3 and 4 is 2 μm and 15 μm, respectively.



**Fig. S5.** (a) Optical image of the fabricated h-BN/<sup>0.5%</sup>G (the <sup>0.5%</sup>G is completely deposited onto the h-BN) heterostructures device. Channel length and width is 1.5 μm and 5 μm, respectively. The inset shows that the <sup>0.5%</sup>G flake is completely deposited on h-BN, which can fully avoid the effect of SiO<sub>2</sub> substrate on the electrical transport properties of the <sup>0.5%</sup>G. (b) Transfer characteristics for the <sup>0.5%</sup>G on h-BN.



**Fig. S6.** Transfer curves of the reduced oxo-G with 0.8% defects in vacuum and air by two-probe configuration.

### Reference

1. S. Eigler, M. Enzelberger-Heim, S. Grimm, P. Hofmann, W. Kroener, A. Geworski, C. Dotzer, M. Rockert, J. Xiao, C. Papp, O. Lytken, H. P. Steinrück, P. Müller and A. Hirsch, *Adv. Mater.*, 2013, **25**, 3583-3587.
2. L. J. Cote, F. Kim and J. Huang, *J. Am. Chem. Soc.*, 2009, **131**, 1043-1049.
3. R. V. Noorden, *Nature*, 2012, **483**, S32-S33.
4. X. Ma, Q. Liu, D. Xu, Y. Zhu, S. Kim, Y. Cui, L. Zhong and M. Liu, *Nano Lett.*, 2017, **17**, 6961-6967.