Supporting information for

Two-dimensional Semiconducting and Single-crystalline Antimony Trioxide Directly-grown on Monolayer Graphene

Qinke Wu[a,†], Taehwan Jeong[a,†], Sanwoo Park[a,†], Sun Jia[b], Hyunmin Kang[a], Tae Geun Yun[c] and Young Jae Song[*,a,d,e,f]

a. *SKKU Advanced Institute of Nanotechnology (SAINT), Sungkyunkwan University (SKKU), Suwon, 440-746, Korea*
b. *Hunan Key Laboratory for Super-Microstructure and Ultrafast Process, School of Physics and Electronics, Central South University, Changsha 410083, Hunan, China*
c. *College of Information and Communication, Sungkyunkwan University (SKKU), Suwon, 440-746, Korea*
d. *Center for Integrated Nanostructure Physics, Institute for Basic Science (IBS), Sungkyunkwan University (SKKU), Suwon, 440-746, Korea*
e. *Department of Physics, Sungkyunkwan University (SKKU), Suwon, 440-746, Korea*
f. *Department of Nano Engineering, Sungkyunkwan University (SKKU), Suwon, 440-746, Korea*

† Qinke Wu, Taehwan Jeong and Sangwoo Park contributed equally in this work.

*Corresponding authors: yjsong@skku.edu (YJS)

Growth process: a) graphene growth: after the copper foil (Alfa, no. 13382) was cleaned by HF (10%) solution, the nitrogen gas was used to dry it. Then the cleaned copper foil was loaded into the growth chamber to grow monolayer high quality graphene at the temperature of 1050 °C, for annealing 2 hours to flat the copper foil surface under H_2 (50 sccm). And then we turned on the gas flow of H_2 (40 sccm) and CH_4 (40 sccm) for 40 min growth, the fully covered monolayer graphene film on copper foil was achieved. b) Antimony trioxide (Sb_2O_3) flake growth: the powder source (Sb_2O_3) was placed 20 centimeter upstream of the graphene/copper (or copper) substrate, and then the carrier gas of Argon (100 sccm) were introduced into the chamber. When the temperature achieved 650 °C, the growth process starts and lasts 10 minutes. Finally, the antimony trioxide (Sb_2O_3)/graphene heterostructure sample (or Sb_2O_3 flake) was successfully grown on copper substrate.

Transferring process: First, the PMMA was spin coated onto the sample surface and then heated on the hot plate for 5 minutes at 90 °C to solid the PMMA. The O_2 plasma (5 sccm) was then used to remove the backside of the sample at 20 W for 1 minute and the sample was then placed into the copper etchant solution for 10 hours to remove the copper substrate. The PMMA/sample flake was rinsed in the DI water and Hcl (5 %) to remove the residuals. After the cleaning, the PMMA/sample was ready for transferring onto the target substrate (like SiO_2, quartz substrate and copper TEM grid). Finally, the PMMA on the sample was removed by Acetone.

DFT calculations: We performed DFT calculations by using the Vienna ab initio Simulation Package (VASP)[1,2]. We used the Perdew–Burke–Ernzerhof exchange-correlation functional with the projector augmented wave approach[3]. The cutoff energy for the plane-wave basis was set as 500 eV and the convergence criterion of atomic force for geometric optimization was set at 0.005 eV/Å. The vacuum region was set as 20 Å to eliminate interactions between

periodic boundaries. A Monkhorst pack 4×4 k-point mesh was used for the optimization of the structures and an 11×11 k-point mesh was used for the calculation of DOS.

Characterizations: X-ray photoelectron spectroscopy (XPS) measurement was carried out on an ESCA Lab2201-XL spectrometer using an Al Kα X-rays as the excitation source. The morphology and crystal characterization were conducted with a transmission electron microscope (TEM 2100F, JEOL). Atomic force microscopy images were acquired in air cantilevers operated in tapping mode.

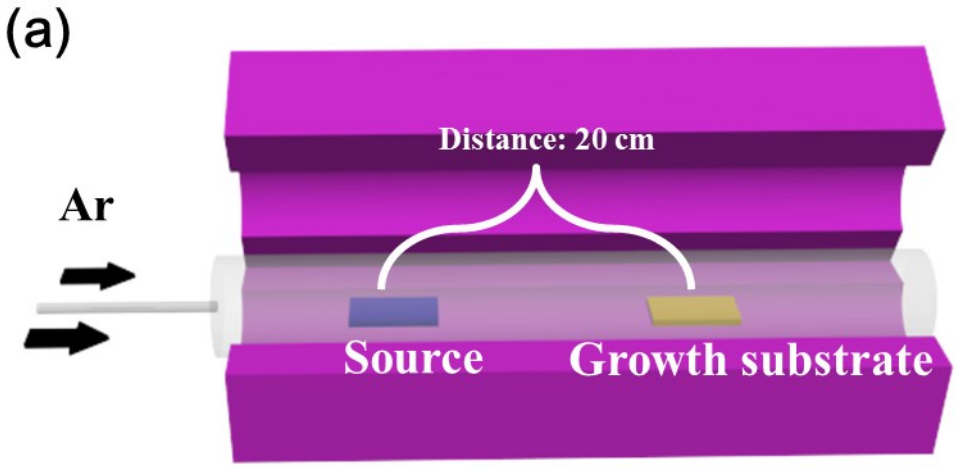

Figure S1. (a) the schematic diagram of the antimony trioxide flakes growth by chemical vapor deposition.

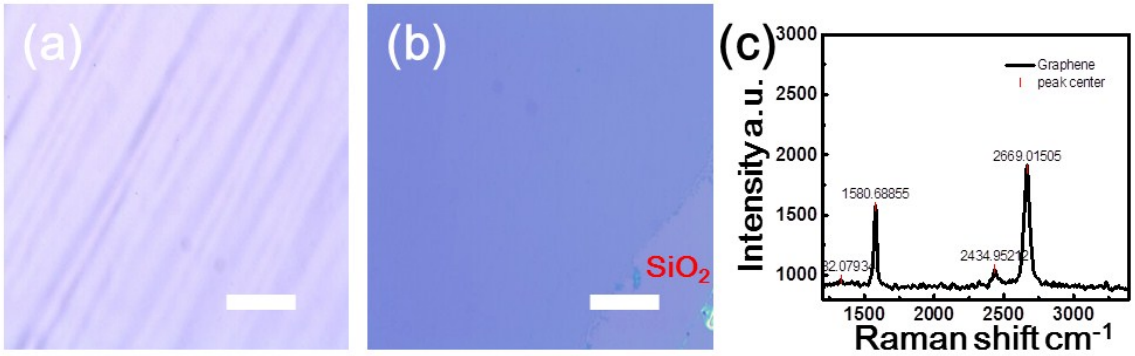

Figure S2. (a) and (b) are the OM images of monolayer graphene on copper and on SiO_2 substrate after transferred. (c) the Raman spectra taken in (b). The scale bars are 20 μm.

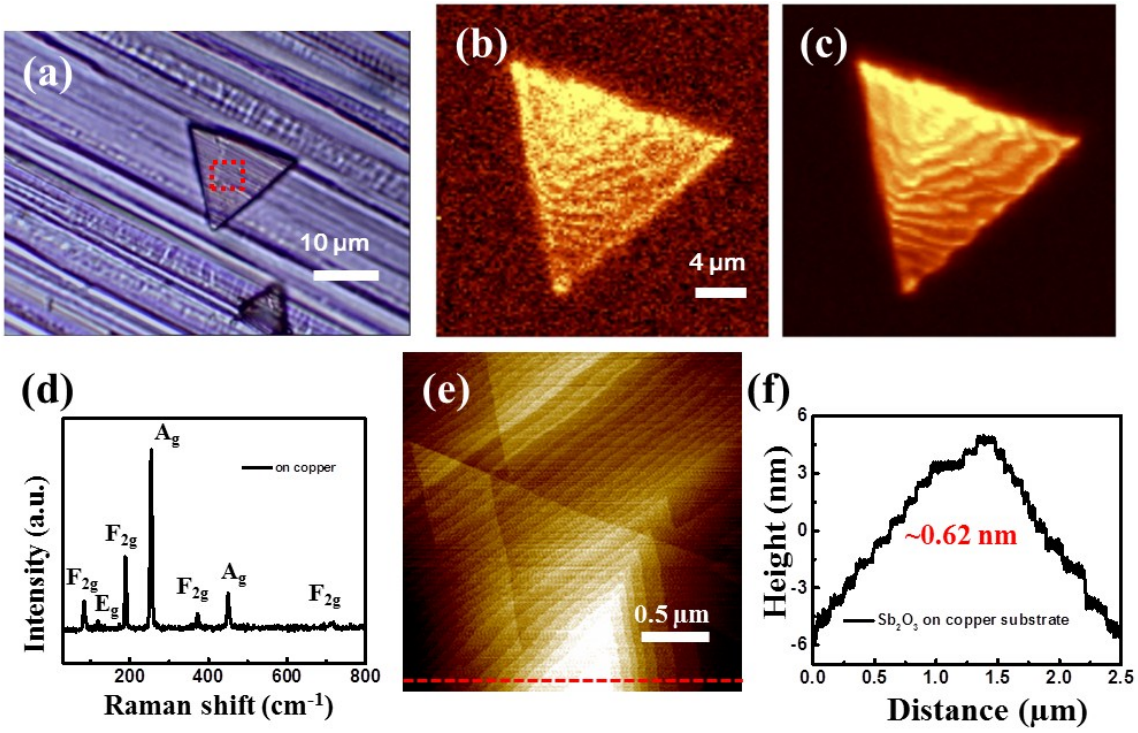

Figure S3. (a) the OM image of the triangle-shape antimony trioxide flake on copper substrate; (b) and (c) are the corresponding Raman mapping of E_g (120 cm^{-1}) and A_g (252 cm^{-1}) peaks of the flake in (a). (d) the Raman spectra measured in (a); (e) the AFM image, measured in the dashed red square in (a). (f) the line profile according the dashed-red line in Figure (e).

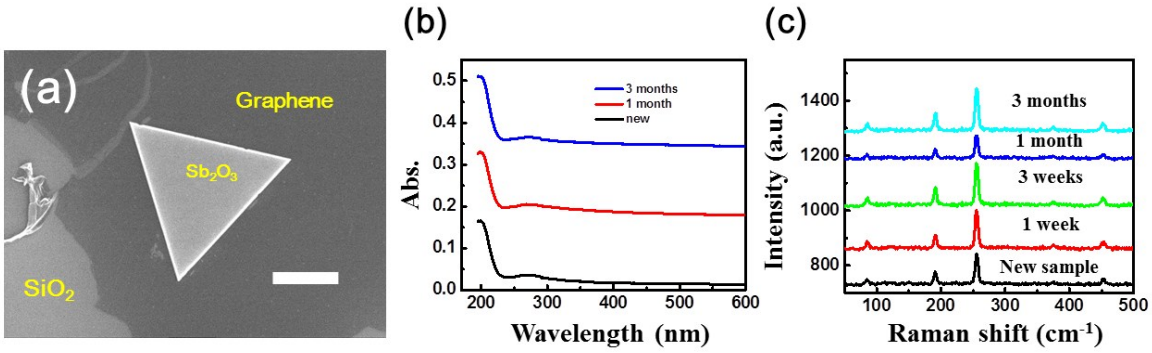

Figure S4. (a) the SEM image of the antimony trioxide/graphene film, after transferred onto SiO_2 substrate. (b) the diagram of the UV-vis spectra measured on the same piece of sample with different exposing time in air. (c) the diagram of the Raman spectra measured on the same piece of sample with different exposing time in air.

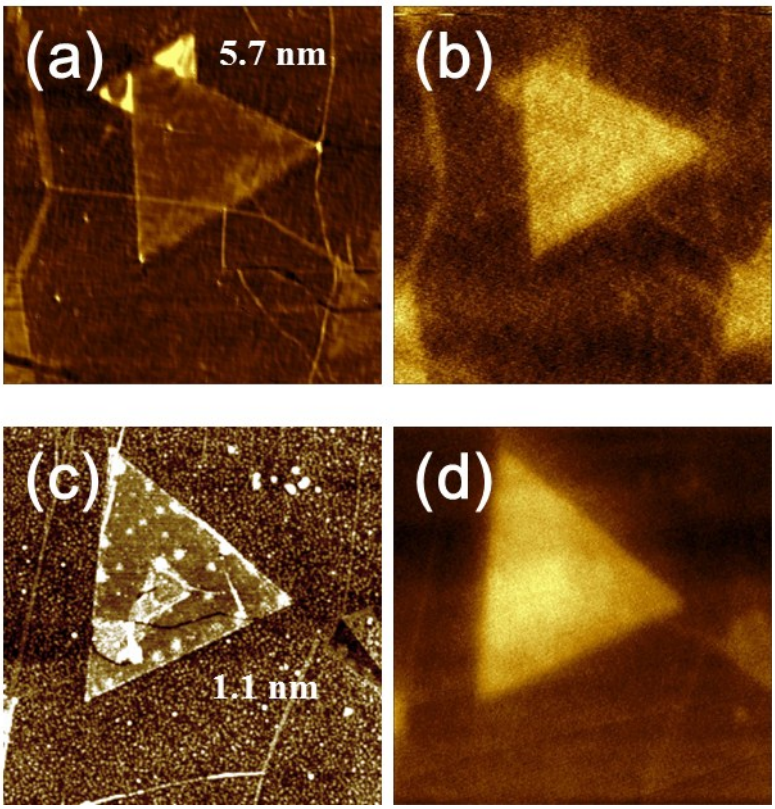

Figure S5. (a) and (c) are the AFM images of the antimony trioxide flake with different thicknesses. (b) and (d) are the corresponding SKPM image.

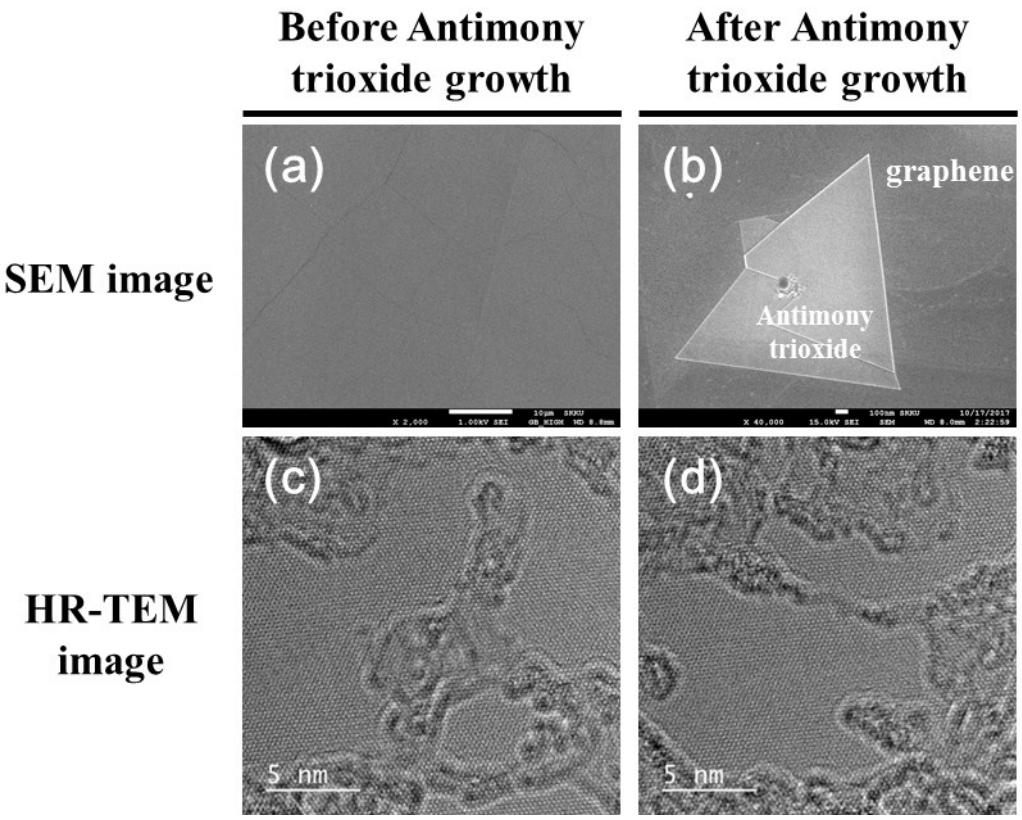

Figure S6. (a) and (b) are the SEM images of the graphene on copper before and after antimony trioxide growth under Ar gas flow. (c) and (d) are the HR-TEM images of the graphene before and after antimony trioxide growth, after transferred onto copper TEM grid.

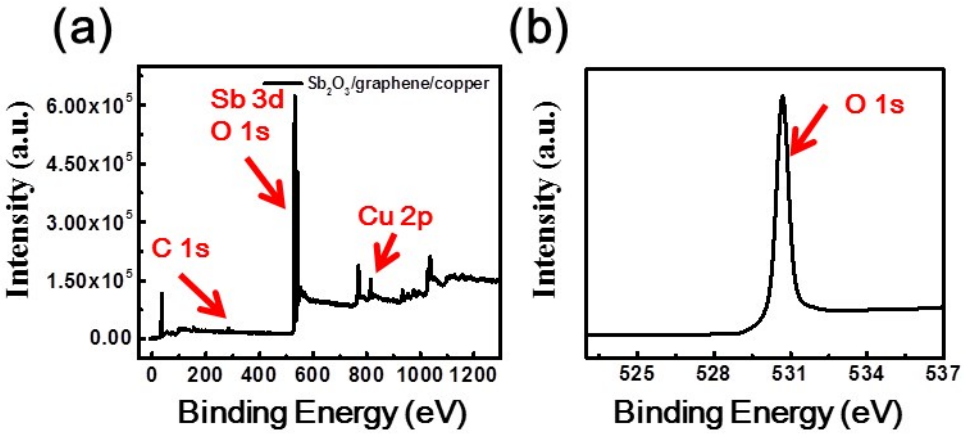

Figure S7. (a) the full width of the XPS spectra of the Sb_2O_3/graphene/copper sample. (b) the XPS spectra of O 1s of the Sb_2O_3/graphene/copper sample.

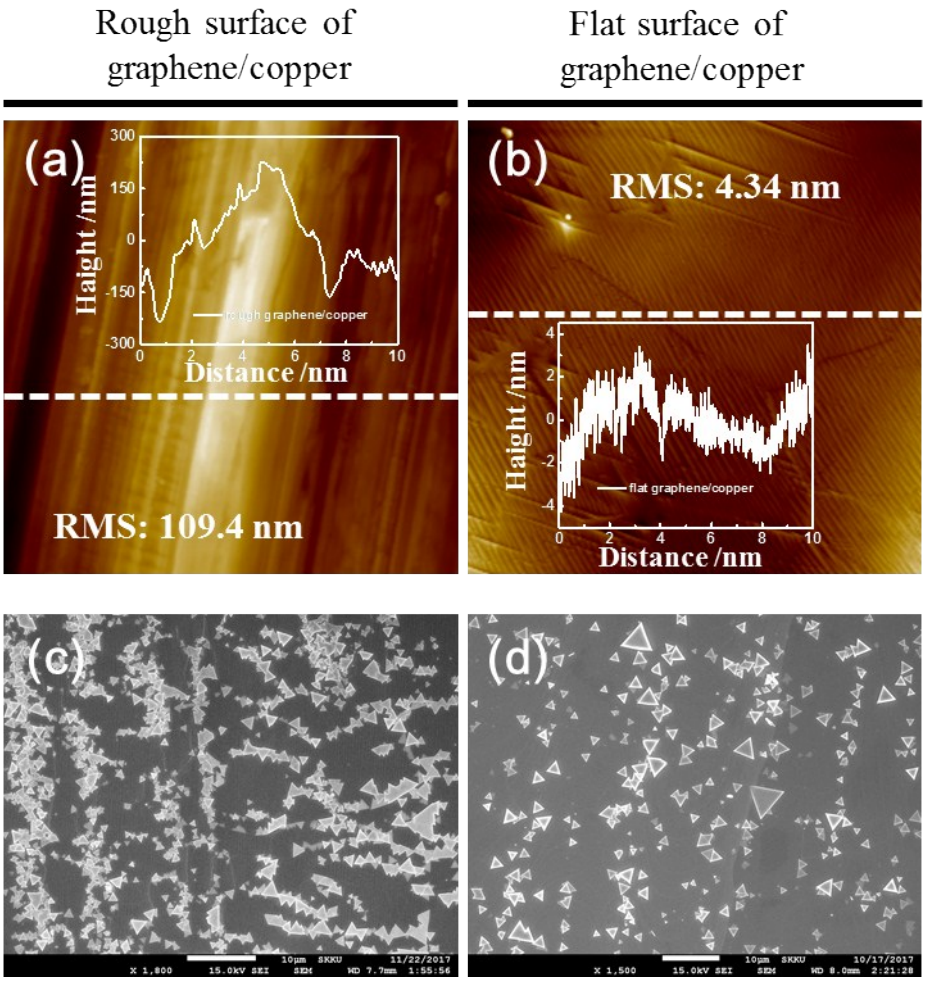

Figure S8. (a) and (b) are the AFM images of the rough and flat graphene/copper substrate surface. (c) and (d) are the SEM images of the rough and flat graphene/copper substrate after antimony trioxide growth under the same growth conditions.

Reference

1. Kresse, G. & Hafner, J. Ab initio molecular dynamics for liquid metals. 4
2. Kresse, G. & Furthmüller, J. Efficient iterative schemes for *ab initio* total-energy calculations using a plane-wave basis set. *Phys. Rev. B* **54,** 11169–11186 (1996).
3. Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized Gradient Approximation Made Simple. *Phys. Rev. Lett.* **77,** 3865–3868 (1996).