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# Epitaxial growth of large-area single-layer graphene over Cu(111)/sapphire by atmospheric pressure CVD

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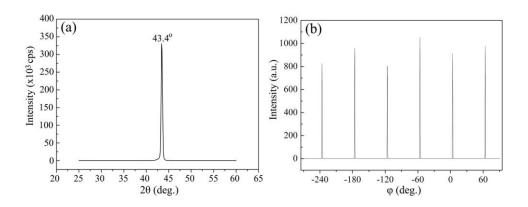
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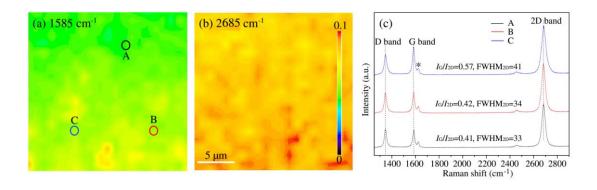
# **Supplementary Material**

Epitaxial growth of large-area single-layer graphene over Cu(111)/sapphire by atmospheric pressure CVD

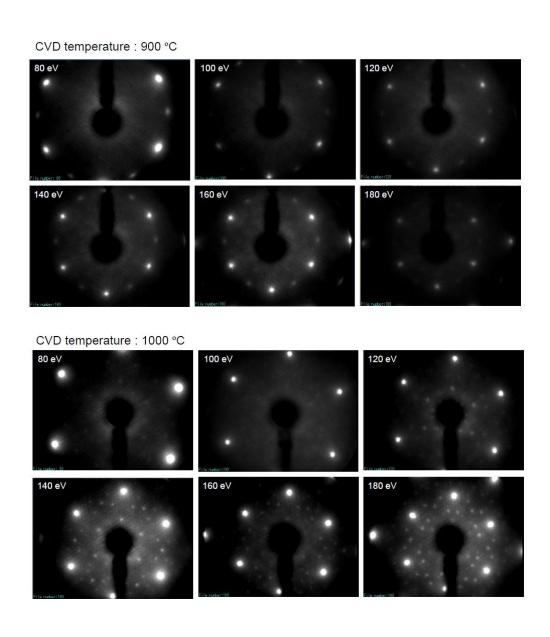
Baoshan Hu, Hiroki Ago,\* Yoshito Ito, Masaharu Tsuji, Eisuke Magome, Kazushi Sumitani, Noriaki Mizuta, Ken-ichi Ikeda, and Seigi Mizuno



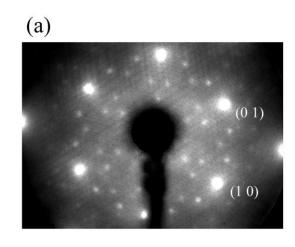
**Fig. S-1** (a)  $\theta$ -2 $\theta$  profile, and (b)  $\phi$  scan of XRD for as-grown graphene/Cu/c-plane Al<sub>2</sub>O<sub>3</sub> at 1000 °C CVD.

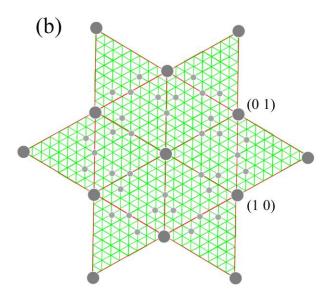


**Fig. S-2** Raman mapping images of (a) G band (~1585 cm<sup>-1</sup>) and (b) 2D band (~2685 cm<sup>-1</sup>) intensities of transferred graphene which was grown at 900 °C. (c) Raman spectra of 3 highlighted points with circles in (a). The symbol "\*" at around 1620 cm<sup>-1</sup> in (c) indicates the defect-related D' peak.

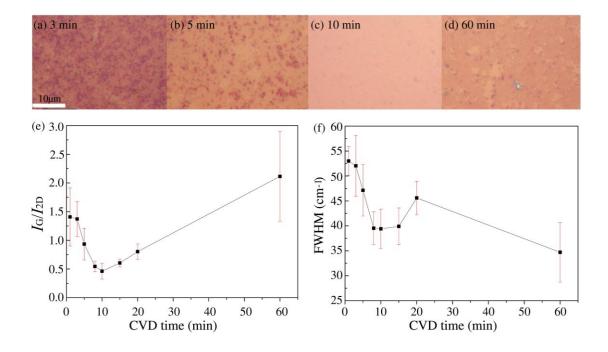


**Fig. S-3** LEED patterns of the graphene/Cu/c-plane sapphire samples measured at different electron energies.

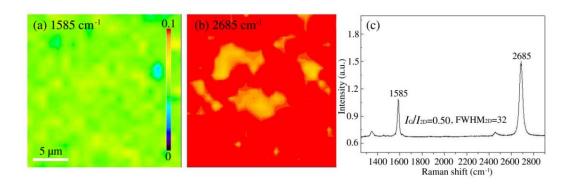




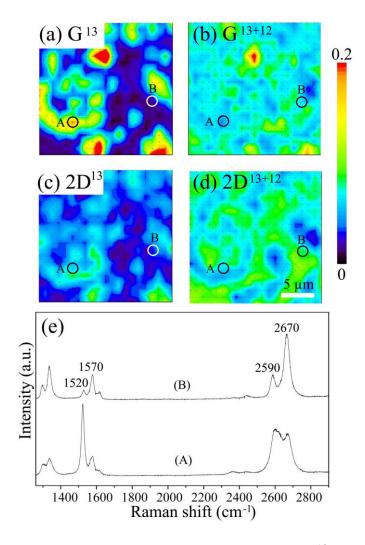
**Fig. S-4** (a) LEED pattern of single-layer graphene on Cu/c-plane sapphire grown at °C (beam energy is 190 eV). (b) Calculated diffraction spots for image (a). Red and green lines correspond to  $(1\times1)$  and  $(8\times8)$  structures, respectively.



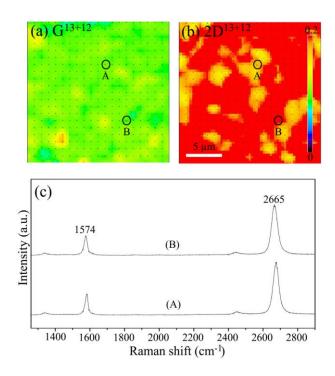
**Fig. S-5** Optical micrographs (a-d) of transferred graphene sheets grown with different CVD times at 900 °C. (e)  $I_{\rm G}/I_{\rm 2D}$  and (f) FWHM<sub>2D</sub> of transferred graphene as a function of CVD time, calculated from the Raman mapping data.



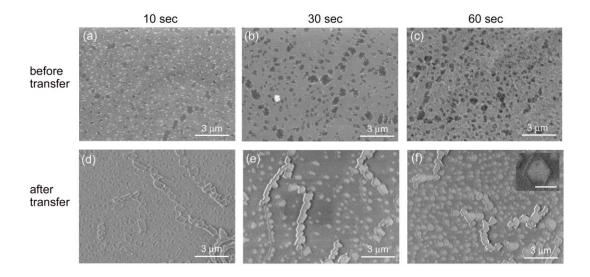
**Fig. S-6** Raman mapping images of G (a) and 2D bands (b), and representative Raman spectrum of the transferred graphene on  $SiO_2/Si$  substrate. The graphene was grown on Cu/c-plane sapphire by 3 min CVD at 1000 °C.



**Fig. S-7** Raman measurement of transferred graphene film with <sup>13</sup>CH<sub>4</sub> (0-8 min) and <sup>12</sup>CH<sub>4</sub> (8-10 min) grown from 900 °C CVD. Raman mapping images of G bands at 1520 cm<sup>-1</sup> (a) and 1570 cm<sup>-1</sup> (b). Raman mapping images of 2D bands at 2590 cm<sup>-1</sup> (c) and 2670 cm<sup>-1</sup> (d). (e) Raman spectra of 2 points highlighted in (a-d).



**Fig. S-8** Raman data of transferred graphene with  $^{13}$ CH<sub>4</sub> (0-8 min) and  $^{12}$ CH<sub>4</sub> (8-10 min) grown from 1000 °C CVD. Raman mapping images of G bands at 1570 cm<sup>-1</sup> (a) and 2D bands at 2655 cm<sup>-1</sup> (b). (c) Raman spectra measured at 2 points marked with A and B in the same region as (a) and (b).



**Fig. S-9** SEM images of graphene domains measured before (a-c) and after (d-f) transfer onto SiO<sub>2</sub>/Si. The CH<sub>4</sub> supply time is 10 sec (a,d), 30 sec (b,e), and 60 sec (c,f). White dots seen in (a) are supposed to be originated in Cu nanoparticles. Inset of (f) shows a hexagonal graphene domain (scale bar: 500 nm).