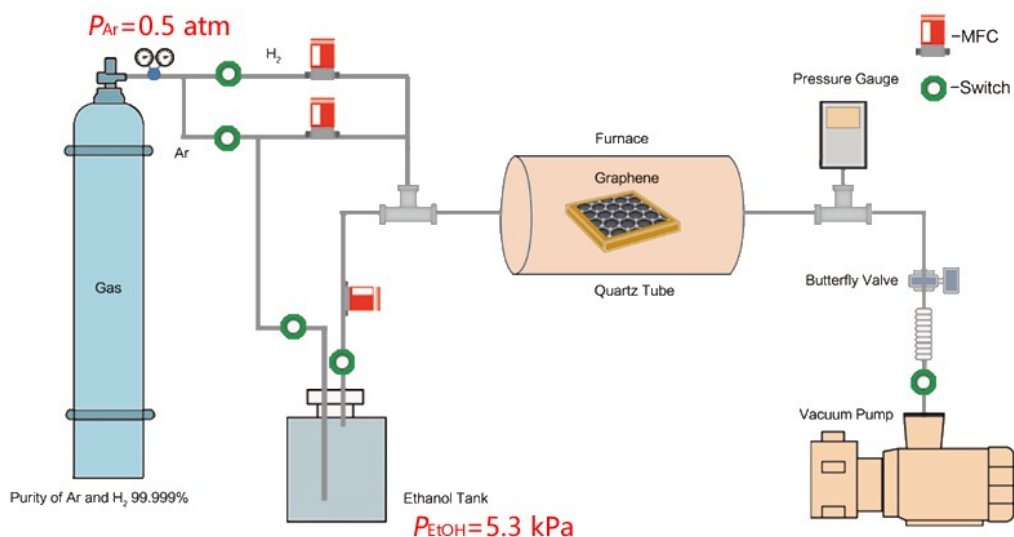


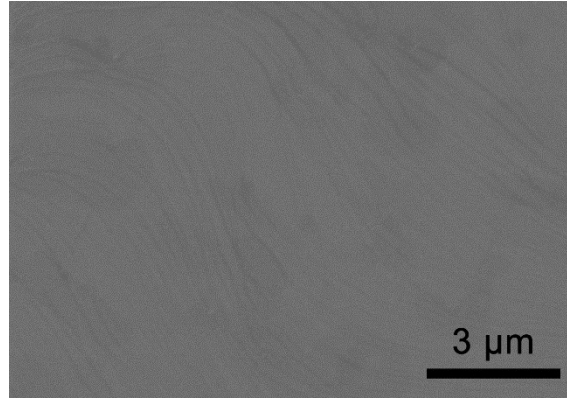
## Electronic Supplementary Information

### Epitaxial nucleation of CVD bilayer graphene on copper

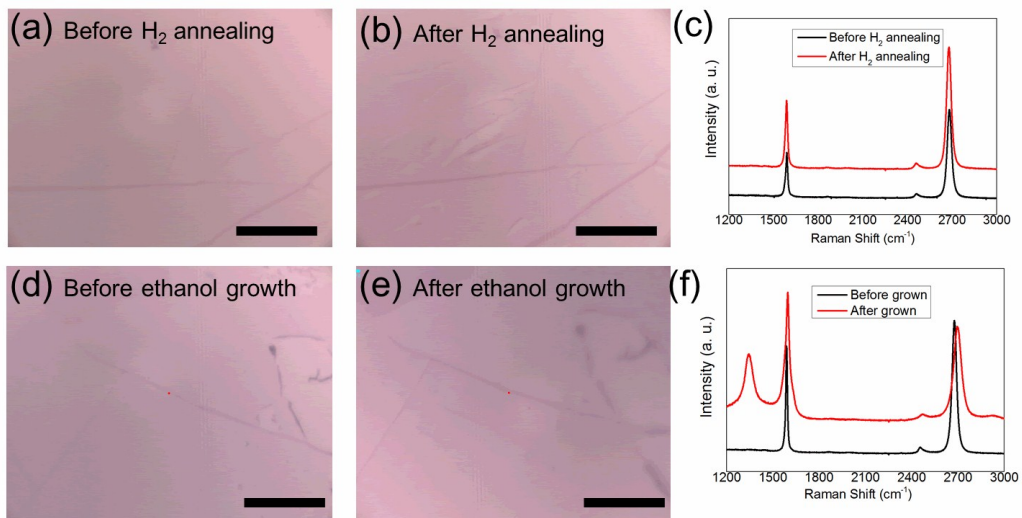
*Yenan Song, Jianing Zhuang, Meng Song, Shaoqian Yin, Yu Cheng, Xuwei Zhang, Miao Wang, Rong Xiang, Yang Xia, Shigeo Maruyama, Pei Zhao, Feng Ding, Hongtao Wang*



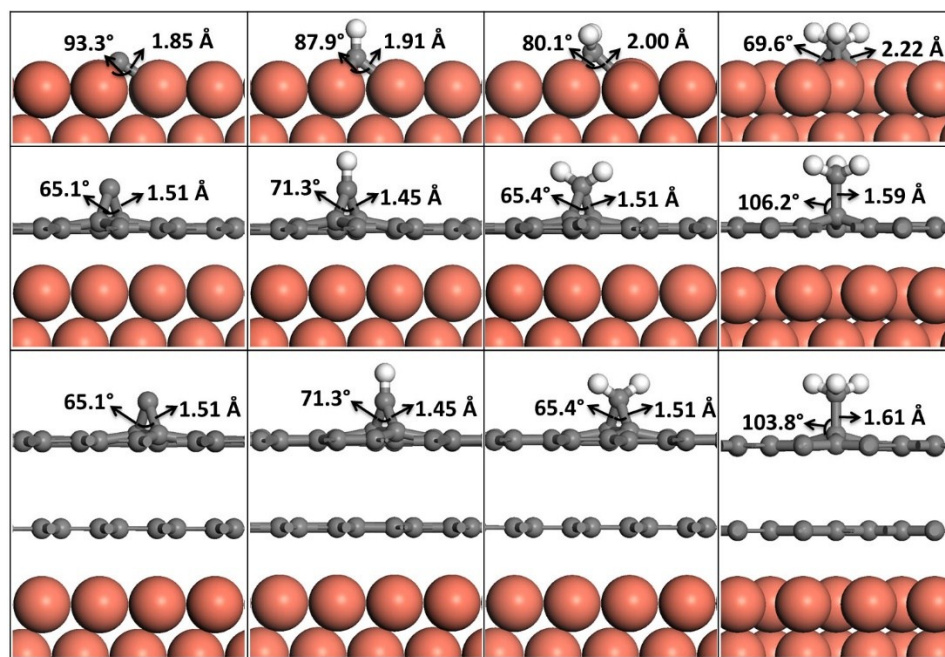
**Figure S1.** An ethanol-based epitaxial CVD system for BLG growth. The pressure of Ar flow was controlled by the release valve to be 0.5 atm, and the ethanol temperature was maintained at 19 °C, whose saturated vapor pressure is 5.3 kPa. Therefore, the flow rate ratio between Ar and ethanol in a bubbled ethanol flow is approximately 10:1.



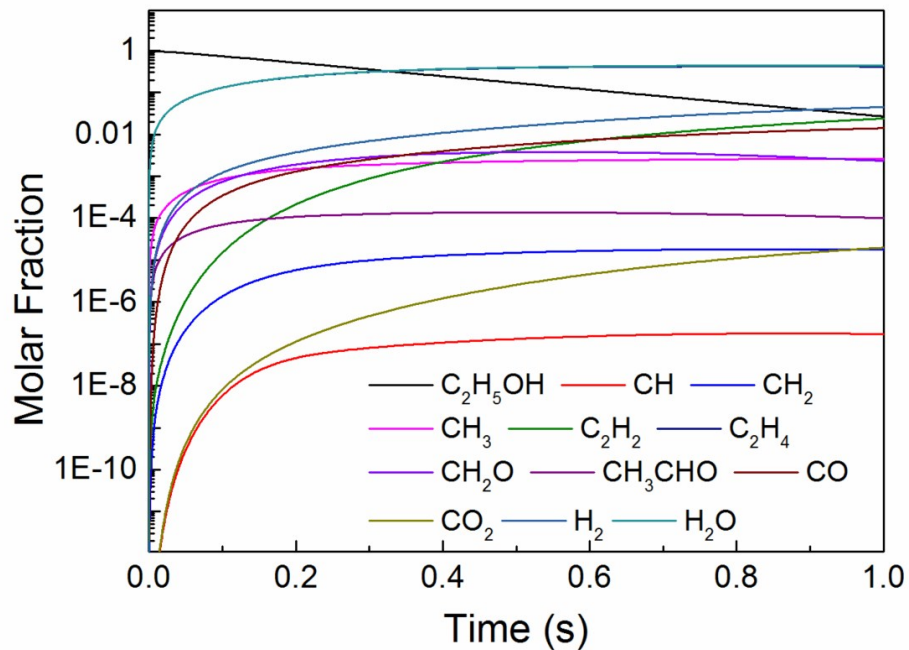
**Figure S2.** CVD growth after 5 hours still obtains a uniform BLG film, which indicates an equilibrium state for BLG growth using ethanol as the precursor.



**Figure S3.** CVD growth at an equivalent condition using ethanol as the precursor but graphene transferred onto a Si/SiO<sub>2</sub> as the substrate instead of copper. No new graphene layer was obtained even consider the etching effect from H<sub>2</sub>.



**Figure S4.** Geometries of different  $\text{CH}_i$  ( $i=0, 1, 2, 3$ ) radicals on a Cu(111)/graphene substrate.



**Figure S5.** Decomposition products of from  $\text{C}_2\text{H}_5\text{OH}$  at high temperature calculated by CHEMKIN-II.

**Table S1.** Total energy data calculated for the radical/graphene/Cu(111) system.

Radical	C	CH	CH <sub>2</sub>	CH <sub>3</sub>
Total energy (eV)	-1.23966895	-6.05787351	-12.11872387	-18.21134477
Substrate	Cu(111)	Cu(111)/MLG	Cu(111)/BLG	
Total energy (eV)	-140.81099300	-308.66524756	-475.86328245	
	C	CH	CH <sub>2</sub>	CH <sub>3</sub>
Cu(111)	-147.39233313	-152.24136847	-156.34241493	-160.82963338
Cu(111)/MLG	-311.70228781	-316.63018221	-322.55594031	-328.01285288
Cu(111)/BLG	-479.17043701	-484.47056304	-490.43820732	-494.42125405

**Table S2.** Binding energy data for the CH<sub>i</sub> radical on a graphene/Cu(111) substrate.

	C	CH	CH <sub>2</sub>	CH <sub>3</sub>
Cu	5.3417	5.3725	3.4127	1.8073
1LG/Cu	1.7303	1.8400	1.7049	1.0692
2LG/Cu	2.0637	2.5456	2.4524	0.3429