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Supporting Information

Chemical vapor deposition of bilayer graphene with layer-resolved growth through dynamic pressure control

Birong Luo[a], Bingyan Chen[b], Anle Wang[c], Dechao Geng[a], Jie Xu[a], Huaping Wang[a], Zhiyong Zhang[b], Lianmao Peng[b], Zhiping Xu[c] and Gui Yu*[ad]*

[a] Beijing National Laboratory for Molecular Sciences, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, P. R. China

[b] Key Laboratory for the Physics and Chemistry of Nanodevices and Department of Electronics, Peking University, Beijing 100871, P. R. China

[c]Applied Mechanics Laboratory, Department of Engineering Mechanics and Center for Nano and Micro Mechanics, Tsinghua University, Beijing 100084 , P. R. China

[d] University of Chinese Academy of Sciences, Beijing 100049, China

*Corresponding Author

E-mail: yugui@iccas.ac.cn; xuzp@tsinghua.edu.cn

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S1. Experiment conditions of VPCVD growth

Table S1. The experiment parameters of as-grown graphene by VPCVD method.

Experiment number (#)	The flow rate ratio of CH_4 to H_2 ( )	Growth time (min)	Variation rate of pressure (KPa/min)	Growth temperature (^oC)
1	1:200	5.5	18.36	1020
2	1:150	7.5	13.47	
3	1:100	10	10.1	
4	1:75	15	6.73	
5	1:50	21	4.81	
6	1:40	27	3.74	
7	1:30	37	2.73	
8	1:20	45	2.24	

S2. Supplementary SEM, optical, and Raman microscope characterizations

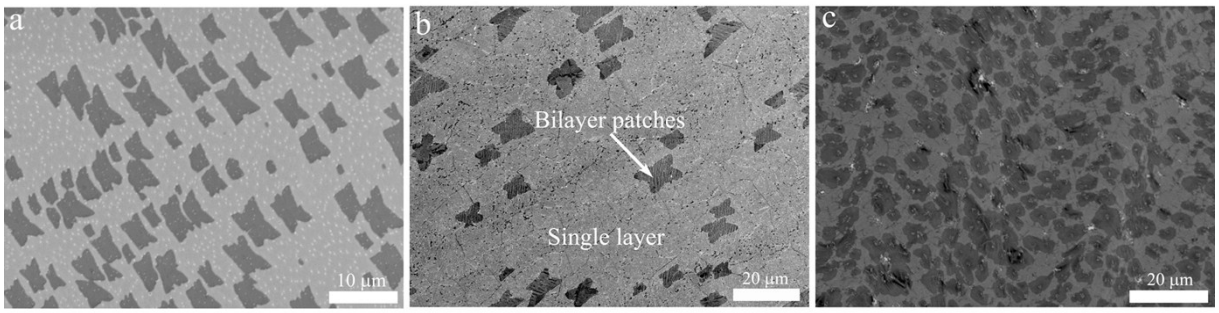

Fig. S1 SEM images of as-grown graphene on copper foils by VPCVD with 1 sccm CH_4 and different H_2 flow rates [a) 150, b) 75, and c) 30 sccm, also see Table S1].

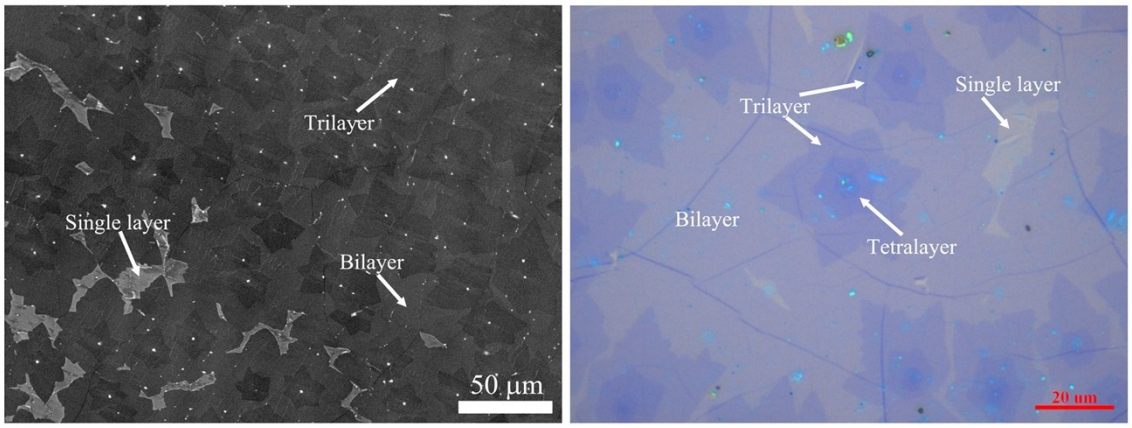

Fig. S2 SEM and optical images of as-grown graphene by VPCVD with 1:10 flow rate ratio of CH_4/H_2, showing the formation of tri- even tetra-layer graphene.

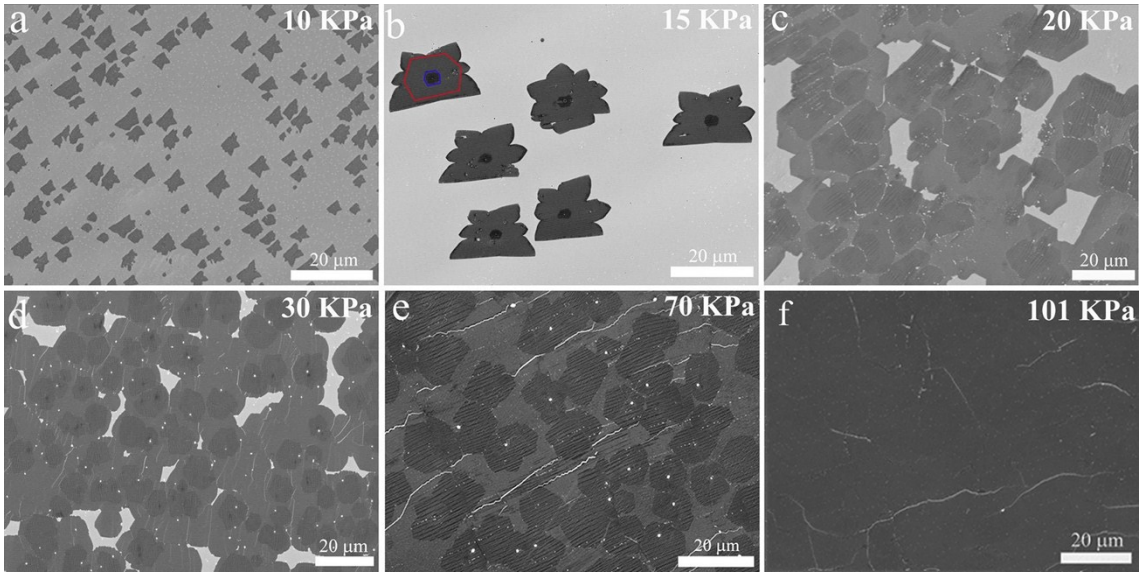

Fig. S3 SEM images of as-grown graphene at different intermediate states of end pressure (10, 15, 20, 30, 70, 101 KPa) in the VPCVD using 1:20 flow rate ratio of CH_4/H_2 gas flow.

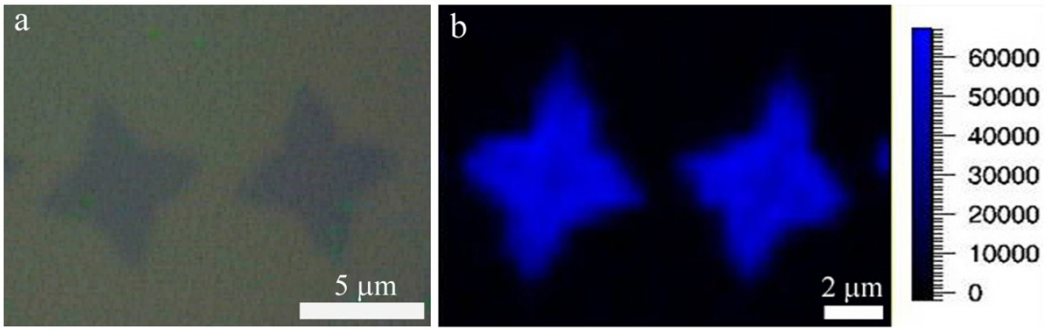

Fig. S4 a) Optical image of single-layer graphene grains. b) The corresponding intensity mapping of G Raman band for the single-layer graphene grains.

S3. AFM characterization of bilayer graphene patches

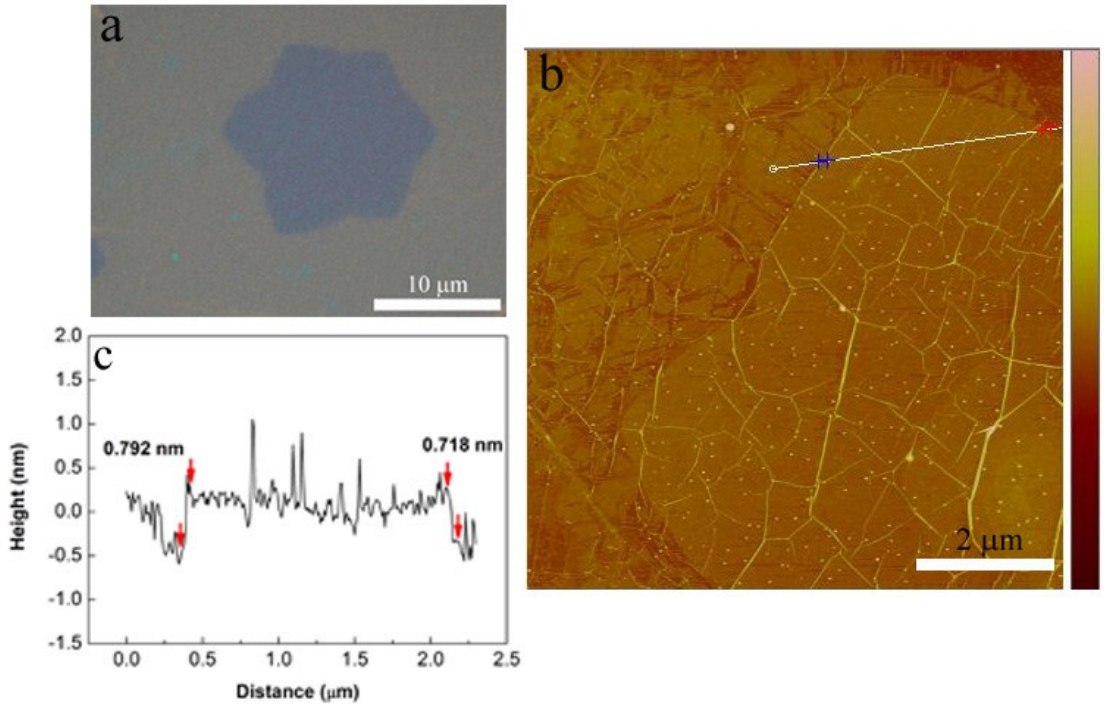

Fig. S5 a) Optical image of a bilayer graphene patch transferred onto SiO_2/Si substrate. b) The AFM height image of lobes of bilayer graphene patch transferred onto SiO_2/Si substrate. c) The corresponding step height profile between the second layer and the first layer graphene along the line in (b).

S4. TEM characterizations of the bilayer graphene with disoriented order

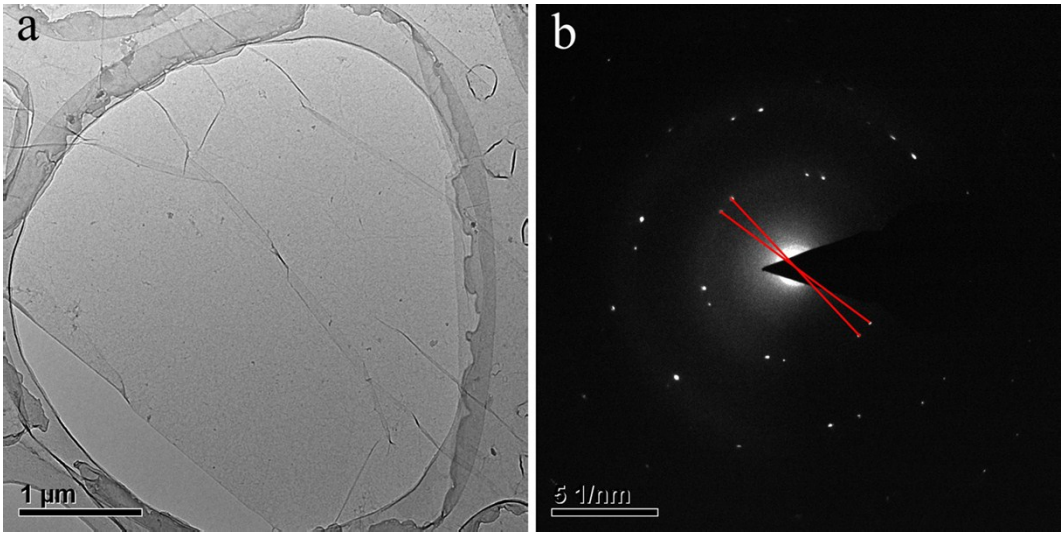

Fig. S6 a) TEM image of bilayer graphene with disoriented stacking order produced during the sample fabrications. b) Its corresponding diffraction patterns.

S5. **Method for the theoretical calculation**

We perform molecular dynamics (MD) simulations using the large-scale atomic/molecular massively parallel simulator (LAMMPS) package[1]. The adaptive intermolecular reactive empirical bond-order (AIREBO) potential function is employed to calculate interaction between carbon atoms in graphene[2]. Copper substrates are modeled using the embedded atom method (EAM), where the graphene and Cu (111) surface are aligned in the *top-fcc* configuration[3]. In additional to the van der Waals interactions between graphene layers, a pairwise interaction in the Lennard-Jones form $V(r) = 4\varepsilon[(\sigma/r)^{12} - (\sigma/r)^6]$ is introduced for the carbon-copper interactions, where the parameters $\varepsilon = 0.02578$ eV and $\sigma = 0.30825$ are obtained by mixing the parameters for carbon (in graphite form) and copper (in face-centered crystal) using the Lorentz-Berthelot rule[3]. The stacked graphene-metal hybrids with different mis-orienated angles are constructed and relaxed afterwards using the conjugated-gradient (CG) algorithm to obtain the total energies.

References

1. S. Plimpton, *J. Comp. Phys.* 1995, **117 (1)**, 1-19.
2. D. W.Brenner, O. A.Shenderova, J. A. Harrison, S. J. Stuart, B. Ni and S. B. Sinnott, *J. Phys.: Condens. Matter.,* 2002, **14 (4)**, 783-802.
3. Z. Xu and M. J. Buehler, *ACS Nano,* 2009, **3 (9)**, 2767-2775.

S6. Supplementary electronic properties of the dual-gate devices

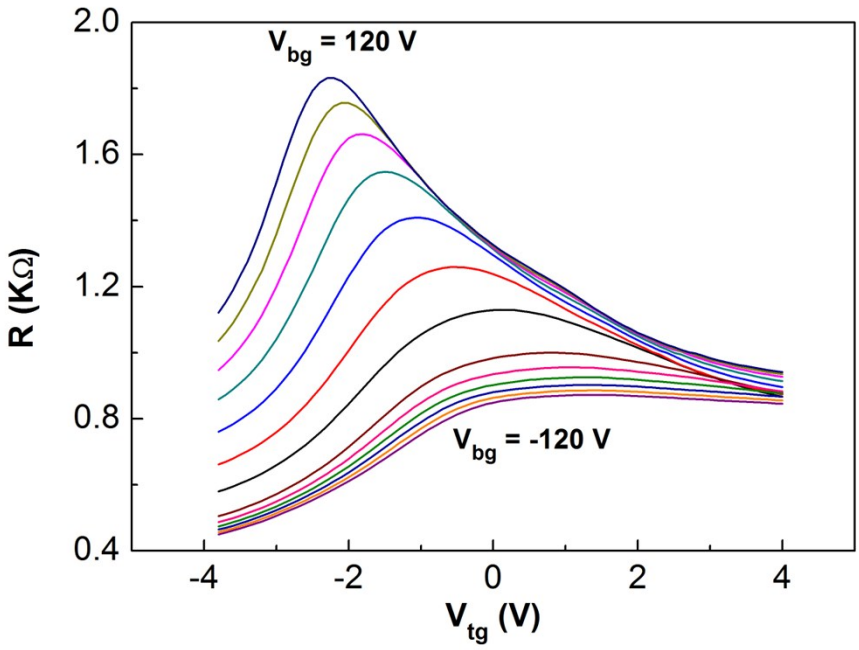

Fig. S7 R *vs.* top-gate voltage (V_{tg}) at different back-gate voltage (V_{bg}) ranging from 120 V to -120 V with 20 V steps for the dual-gate device measured in the air atmosphere.