

Supporting Information for

Wafer Scale Homogeneous Bilayer Graphene Films by Chemical Vapor Deposition

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Bilayer graphene growth & transfer

25 μ m thick copper foil (99.8%, Alfa Aesar) was loaded into an inner quartz tube inside a 3 inch horizontal tube furnace of a commercial CVD system (First Nano EasyTube 3000). The system was purged with argon gas and evacuated to a vacuum of 0.1 Torr. The sample was then heated to 1000°C in H₂ (100 sccm) environment with vacuum level of 0.35 Torr. When 1000°C is reached, 70 sccm of CH₄ is flowed for 15 minutes at vacuum level of 0.45 Torr. The sample is then cooled slowly to room temperature with a feed back loop to control the cooling rate. The vacuum level is maintained at 0.5 Torr with 100 sccm of argon flowing. The time plot of the entire growth process is shown in Fig. S1.

Two different methods were used to transfer bilayer graphene from copper foil to SiO₂ substrates. The first method utilize thermal release tape (Nitto Denko) to transfer bilayer graphene from the copper foil.¹ The tape was attached to the copper surface and a force of 6.25 N/cm² was applied to the copper/bilayer graphene/tape stack for 10 minutes with EVG EV520IS wafer bonder. The other side of the copper is exposed to O₂ plasma for 30 seconds to remove the graphene on that side. The copper was etched away using iron (III) nitrate (Sigma Aldrich) solution (0.05g/ml) for 12 hours. A 4 inch silicon wafer with thermally grown SiO₂ was precleaned with nP12 nanoPREP using plasma power of 500W for 40 seconds to modify the surface energy and produce a hydrophilic surface. The tape and bilayer graphene stack was transferred to the precleaned SiO₂ wafer and a force of 12.5N/cm² was applied for

10 minutes. The substrate was then heated to 120 °C to eliminate the adhesion strength of the thermal release tape. The tape was then peeled off and the adhesive residue was removed with warm acetone.

Polymethyl methacrylate (PMMA) can also be used instead of thermal release tape to transfer bilayer graphene.² This method is easier as it does not require a bonding tool but the edge part of the graphene is usually rough due to uneven thickness of spin coated PMMA at the edge. In this method, one side of the sample is coated with 950PMMA A6 (Microchem) resist and cured at 180°C for 5 minutes. The other side of the sample is exposed to O₂ plasma for 30 seconds to remove the graphene on that side. The sample is then left in iron (III) nitrate (Sigma Aldrich) solution (0.05g/ml) for at least 12 hours to completely dissolve away the copper layer. The sample is transferred on to a silicon substrate with thermal oxide. The PMMA coating is removed with acetone and the substrate is rinsed several times. After all transfers, Raman spectroscopy as well as optical microscope were used to characterize the graphene film. Electrical transport measurement was done with samples prepared with PMMA method.

Bilayer graphene device fabrication

After the bilayer graphene is transferred onto SiO₂/Si substrate, Ti/Au (5nm/100nm) is deposited to form source/drain electrodes. Bilayer graphene films are then patterned into 1 μm×1μm, 1 μm×2μm, and 2 μm×2μm pieces using conventional lithography and oxygen plasma etching. After 40nm of Al₂O₃ is deposited as the top gate dielectric by Atomic Layer Deposition (ALD), top gates are patterned and metals are evaporated (Ti/Au: 5nm/100nm). We note that the adhesion between metal electrodes and graphene is not perfect, resulting in metal peeling off. We also observed gate leakage/breakdown at high field for some devices. These devices are regarded as fabrication defects, as shown in Fig. 5a.

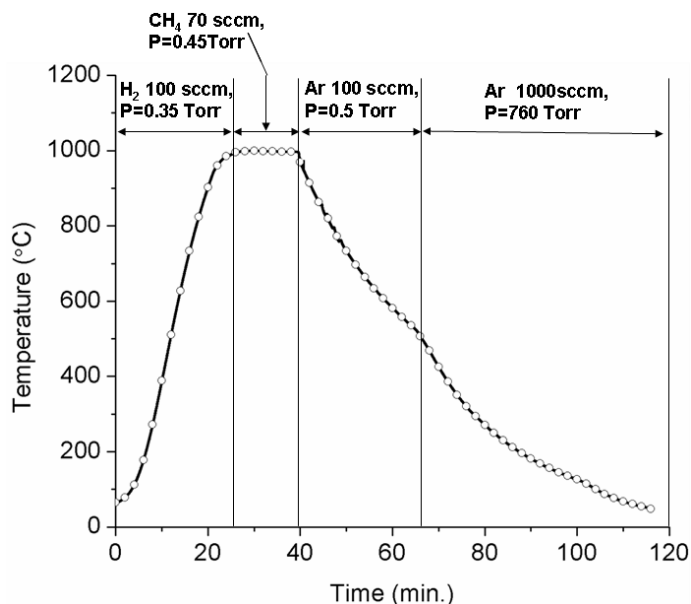


Figure S1. Temperature vs. time plot of bilayer graphene growth condition. Pressure value is denoted as "P".

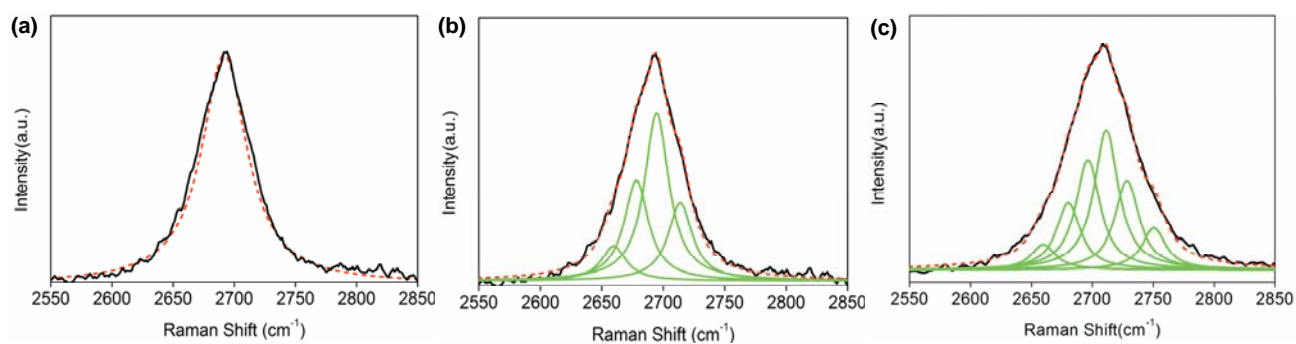


Figure S2. (a) The measured 2D Raman band of a bilayer with the FWHM of 45cm^{-1} . Single Lorentzian fit (red dash line) clearly shows deviation from the measured 2D band. (b) The peak can be well-fitted with the sum of four single Lorentzian (green solid line) of 24cm^{-1} FWHM. (c), The measured 2D Raman band of a trilayer with the FWHM of 62cm^{-1} . 2D peak of trilayer are fitted with six single Lorentzian (green solid line)

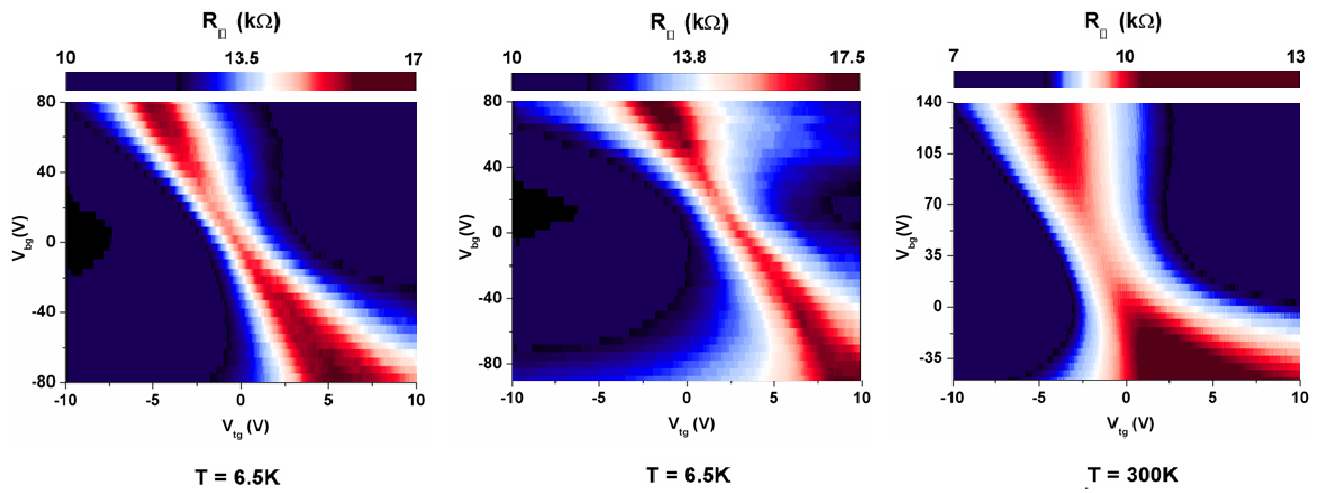


Figure S3. Three dual-gate graphene devices showing bilayer transport behaviour.

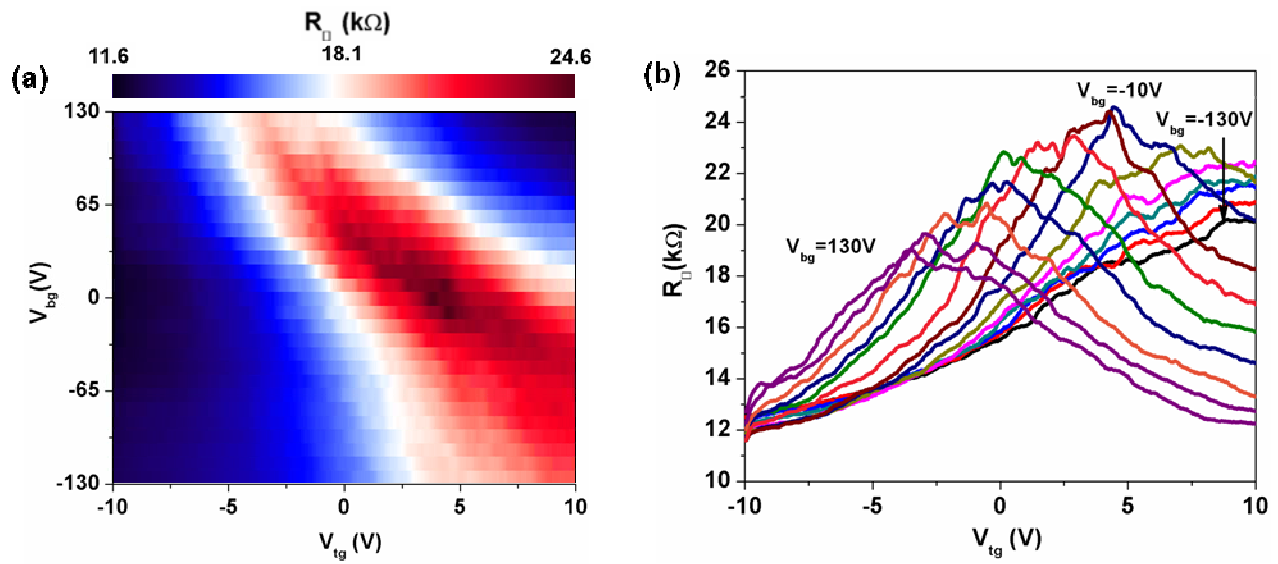


Figure S4. (a) A device showing trilayer transport behaviour. The observed peak square resistance decreases as increasing field. This is distinctively different from bilayer response. (b) Horizontal section views with R_{\square} plotted against V_{tg} at fixed V_{bg} from -130 to 130 V with 20V increment.

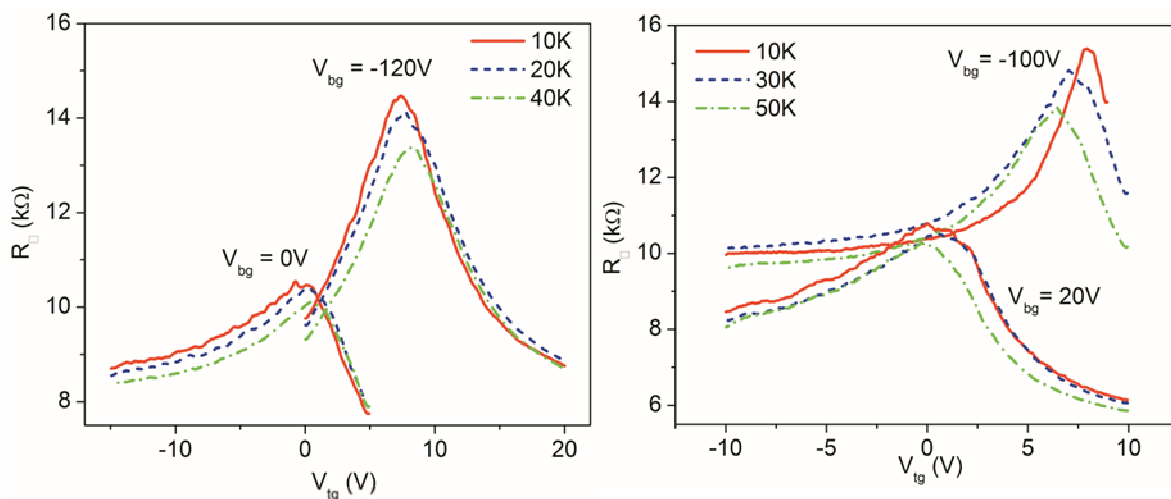


Figure S5. Two dual-gate graphene devices showing temperature dependent resistance versus top gate voltage sweep at two different back gate voltages.

Sample No.	Growth Pressure (Torr)	Growth Temperature (°C)	Growth Time (min)	Ar Flow rate (sccm)	CH ₄ Flow rate (sccm)	H ₂ Flow rate (sccm)	2D Band FWHM (cm ⁻¹)	I _{2D} /I _G	I _D /I _G	Cooling rate (°C/min)
1	0.5	1000	15	0	70	0	46.6	2.628	0.258	18
2	0.5	1000	15	0	140	0	47	2.12	0.57	18
3	Ambient	1000	15	1000	50	0	59.12	1.402	0.36	18
4	1.5	1000	15	0	40	600	60	0.64	1.11	18

Table S1. Comparison of graphene samples synthesized under different conditions.

References

1. Caldwell, J. D.; Anderson, T. J.; Culbertson, J. C.; Jernigan, G. G.; Hobart, K. D.; Kub, F. J.; Tadjer, M. J.; Tedesco, J. L.; Hite, J. K.; Mastro, M. A.; Myers-Ward, R. L.; Eddy, C. R.; Campbell, P. M.; Gaskill, D. K. *ACS Nano* **2010**, *4*, 1108.
2. Li, X.; Cai, W.; An, J.; Kim, S.; Nah, J.; Yang, D.; Piner, R.; Velamakanni, A.; Jung, I.; Tutuc, E.; Banerjee, S. K.; Colombo, L.; Ruoff, R. S. *Science* **2009**, *324*, 1312.