

Supporting Information

Controllable Synthesis and Growth Mechanism of Interlayer Coupled Multilayer Graphene

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Characterizations: The graphene samples underwent systematic characterization using various techniques. Scanning Electron Microscopy (SEM) was performed using a Hitachi S-4800 instrument operating at 5 kV. Optical microscopy was conducted by using an Olympus BX51 microscope. Raman spectroscopy was carried out by using a Renishaw Invia plus spectrometer with a 532 nm laser for excitation. Transmission Electron Microscopy (TEM) analysis was performed by using a JEOL JEM-2100F instrument operating at 200 kV.

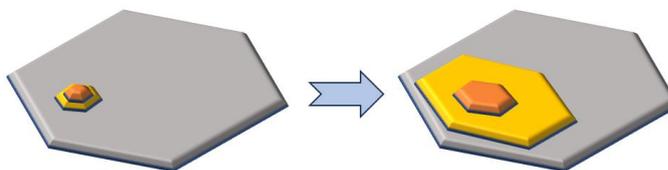


Figure S1. Growth diagram of interlayer coupling multi-layer graphene.

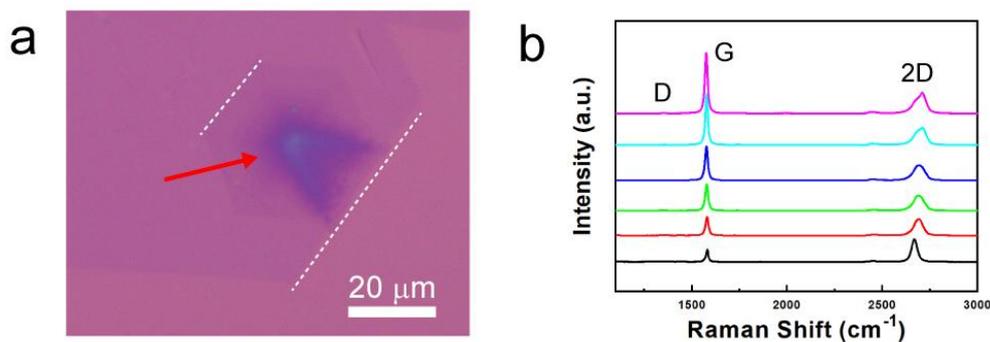


Figure S2. (a) Optical photograph of interlayer-coupled multilayer graphene transferred onto the SiO₂/Si surface, and (b) its corresponding Raman spectrum.

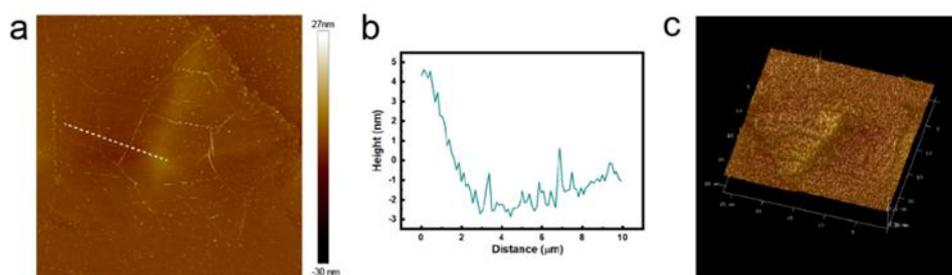


Figure S3. AFM characterization of interlayer-coupled multilayer graphene. (a) AFM image of interlayer-coupled multilayer graphene. (b) Height profile along the white dashed line in figure (a). (c) Three-dimensional AFM image of interlayer-coupled multilayer graphene.

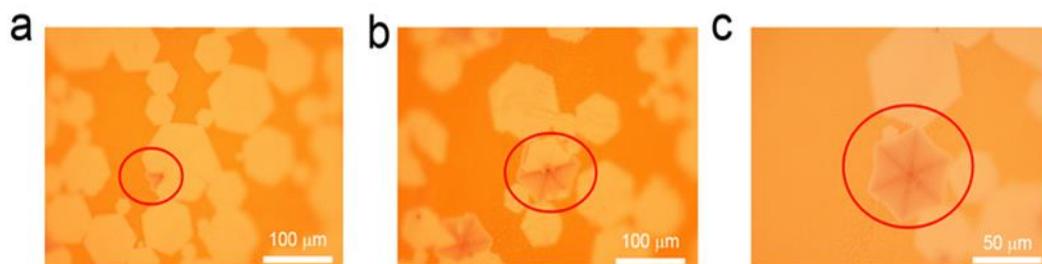


Figure S4. (a–c) Optical photographs of interlayer-coupled multilayer graphene with different morphologies. 500 Ar sccm, 100 sccm H₂ and 0.8 sccm CH₄.

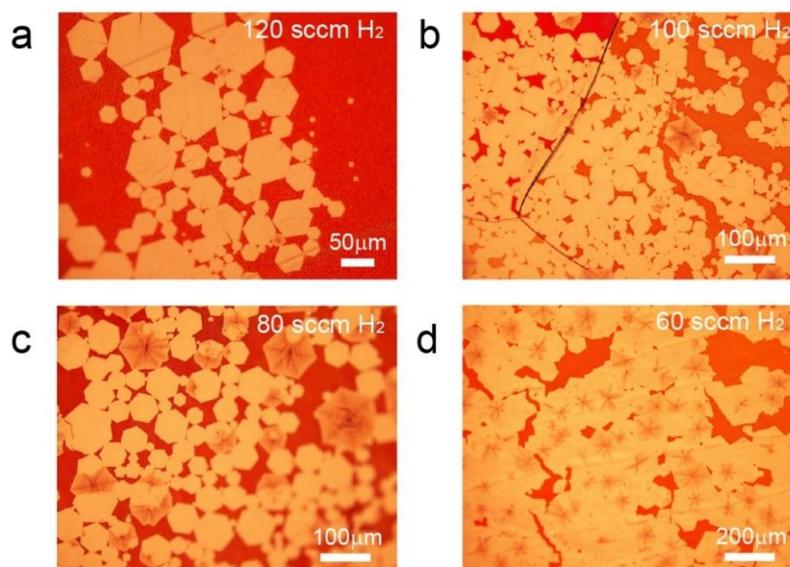


Figure S5. Optical photographs of interlayer-coupled multilayer graphene grown on liquid copper under different hydrogen gas flow rates. 500 Ar sccm, 0.8 sccm CH₄ and (a) 120 sccm H₂, (b) 100 sccm H₂, (c) 80 sccm H₂, (d) 60 sccm H₂, respectively.

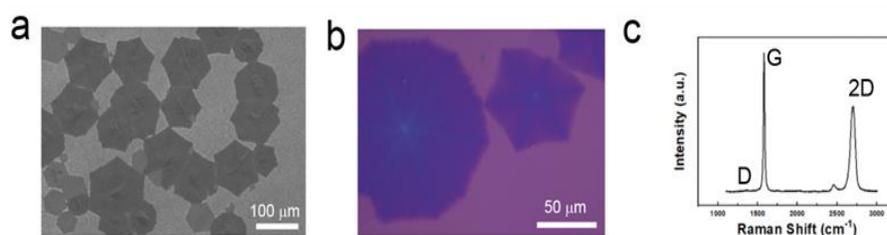


Figure S6. Morphology and structural characterization of umbrella-like multilayer graphene, (a) SEM image. (b) Optical photograph of multilayer graphene transferred onto SiO₂/Si. (c) corresponding Raman spectrum of the multilayer graphene in figure (b).

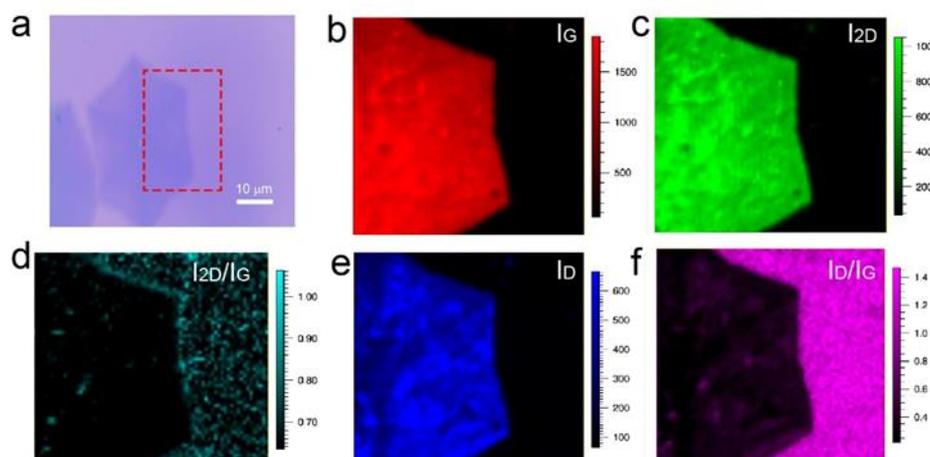


Figure S7. (a) Optical image of umbrella-like multilayer graphene, (b–f) Raman mapping spectra of umbrella-like multilayer graphene.

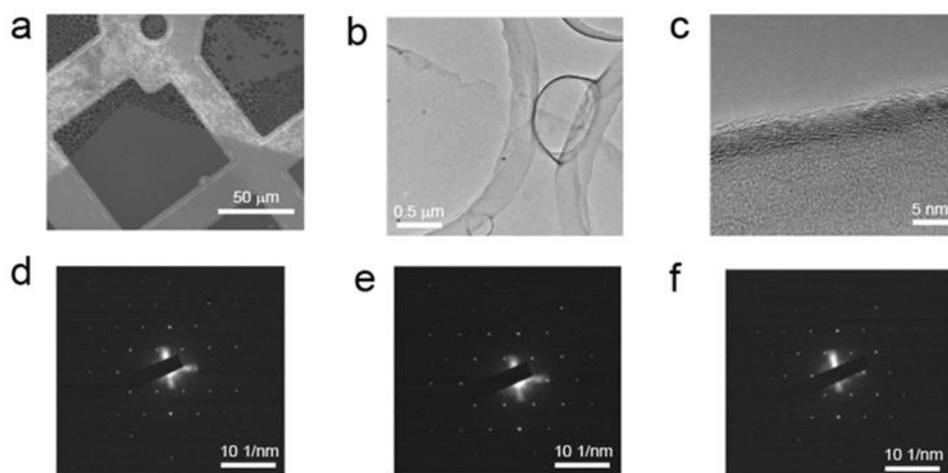


Figure S8. (a) SEM image of umbrella-like interlayer-coupled multilayer graphene transferred onto a microgrid, (b) Low-magnification TEM image of umbrella-like interlayer-coupled multilayer graphene, (c) High-magnification TEM image of umbrella-like interlayer-coupled multilayer graphene, (d–f) SEAD patterns of umbrella-like interlayer-coupled multilayer graphene at different locations.

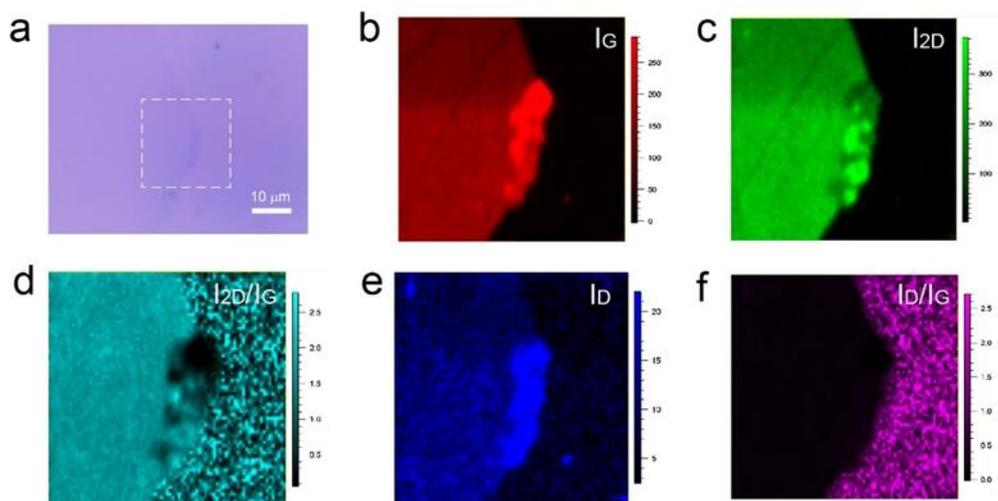


Figure S9. (a) Optical photograph of the sample where carbon species start to accumulate at the edges of graphene domains, (b–d) Raman mapping spectra at the position indicated by the white dashed box in Figure (a).

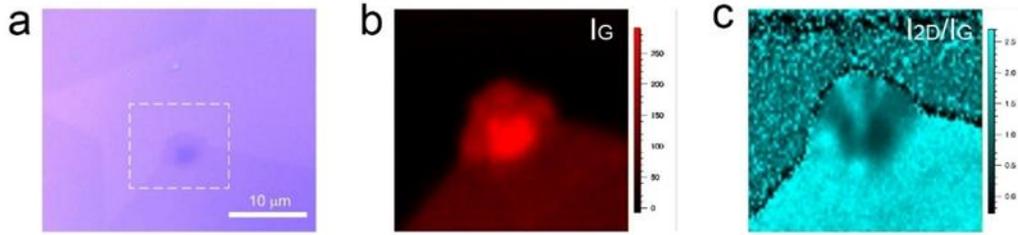


Figure S10. (a) Optical photograph of the sample where carbon species continue to accumulate at the edges of graphene domains, (b,c) Raman imaging spectra of the G peak and I_{2D}/I_G ratio at the position indicated by the white dashed box in Figure (a).

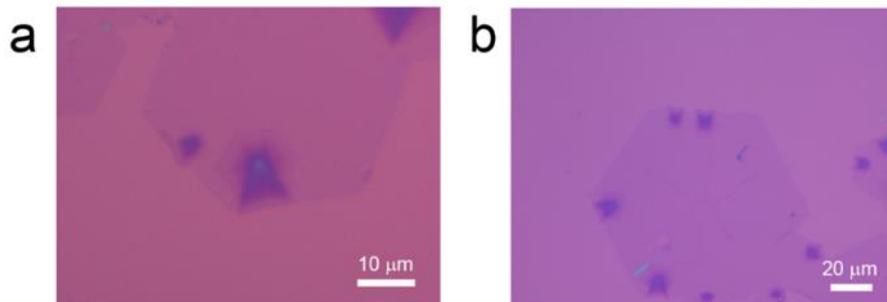


Figure S11. (a,b) Optical photographs of interlayer-coupled multilayer graphene with different morphologies transferred onto SiO_2/Si substrate.

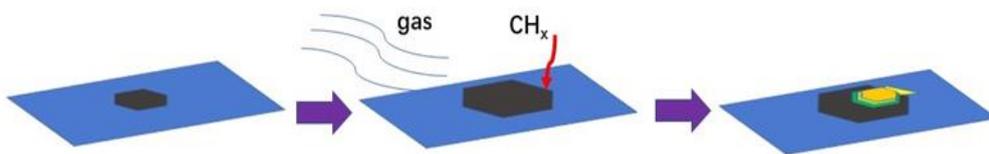


Figure S12. Schematic illustration of the growth process of winged-interlayer-coupled multilayer graphene.