Supplementary Information for

Influence of Graphene Thickness and Grain Boundaries on MoS_2 Wrinkle Nanostructures

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Supplementary Figures

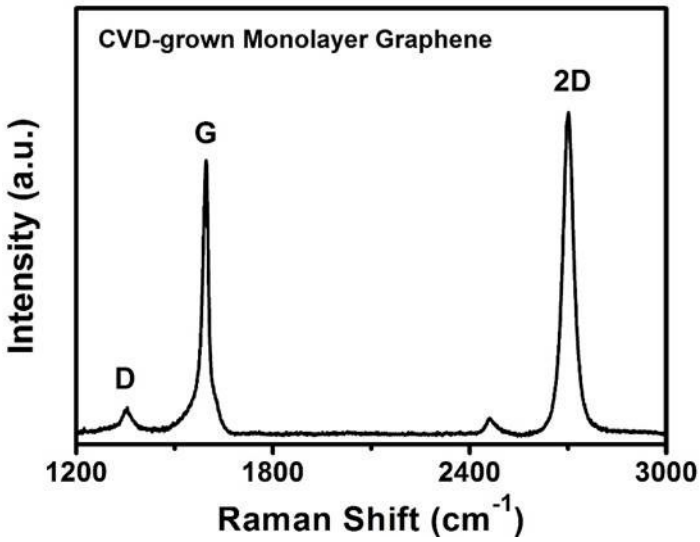

Figure S1. Raman spectrum of monolayer graphene grown by CVD

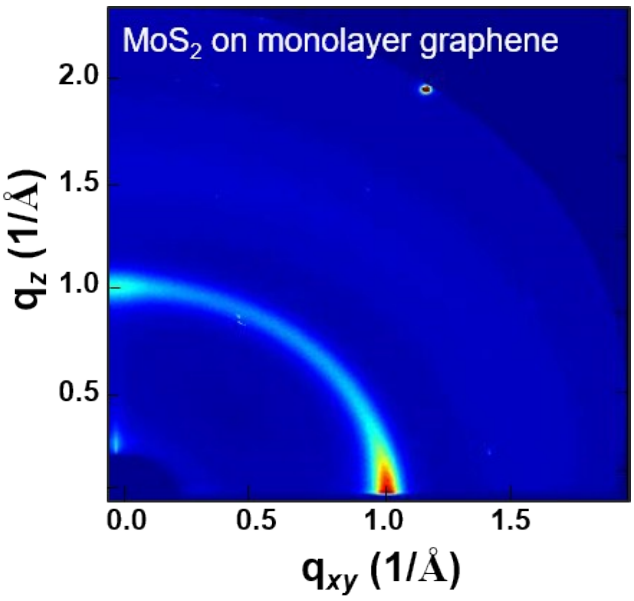

Figure S2. Orientation of MoS_2 layers on monolayer graphene. Grazing incidence X-ray diffraction (GIXRD) patterns collected from MoS_2 wrinkle films prepared on monolayer graphene.

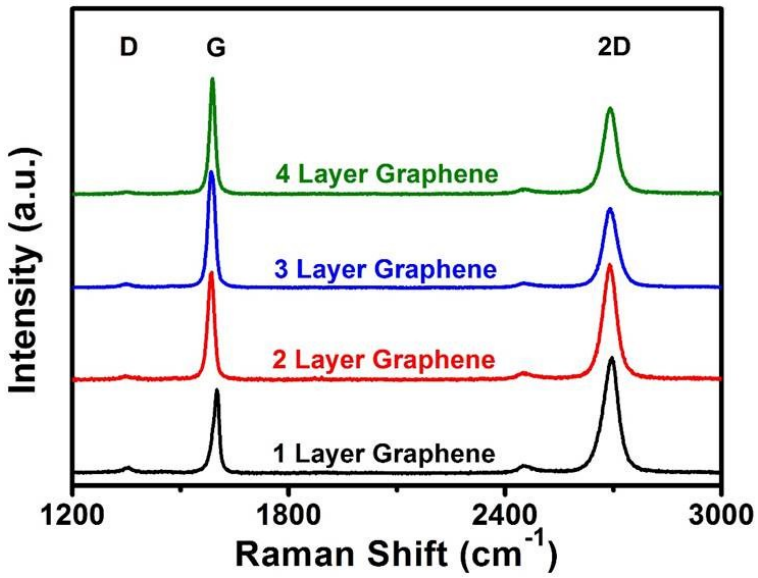

Figure S3. Raman spectra of monolayer, bilayer, trilayer, tetralayer graphene synthesized by CVD methods.

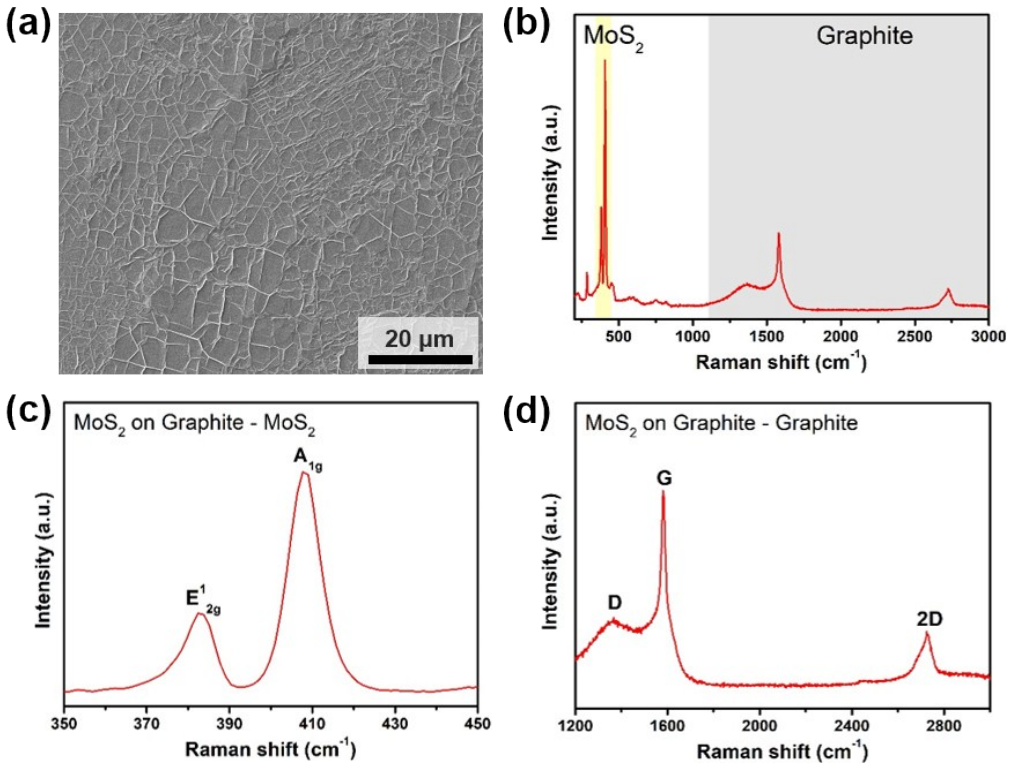

Figure S4. MoS_2 films synthesized on CVD-grown graphite. (a) SEM image of MoS_2 films synthesized on graphite. Wrinkles in the image originate from the CVD growth of graphite on Ni, not from MoS_2 growth. (b-d) Raman spectra of MoS_2 films synthesized on graphite with magnified spectra of the (c) MoS_2 region and (d) graphite region.

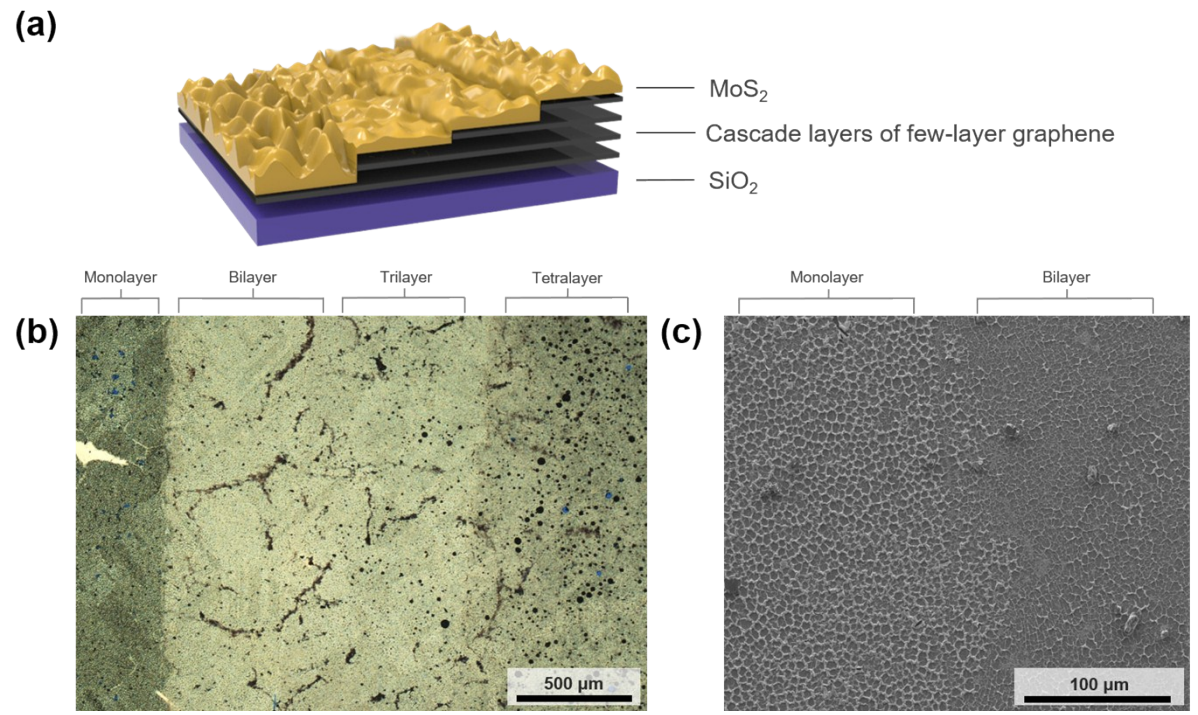

Figure S5. MoS_2 wrinkles synthesized on cascade layers of few-layer graphene on a single substrate. (a) Schematic illustration for MoS_2 synthesized on monolayer to tetralayer graphene. (b) SEM image of MoS_2 wrinkles on mono-, bi-, tri-, and tetralayer graphene on a single substrate, and (c) a SEM image of the boundary between MoS_2 wrinkles on mono- and bilayer graphene.

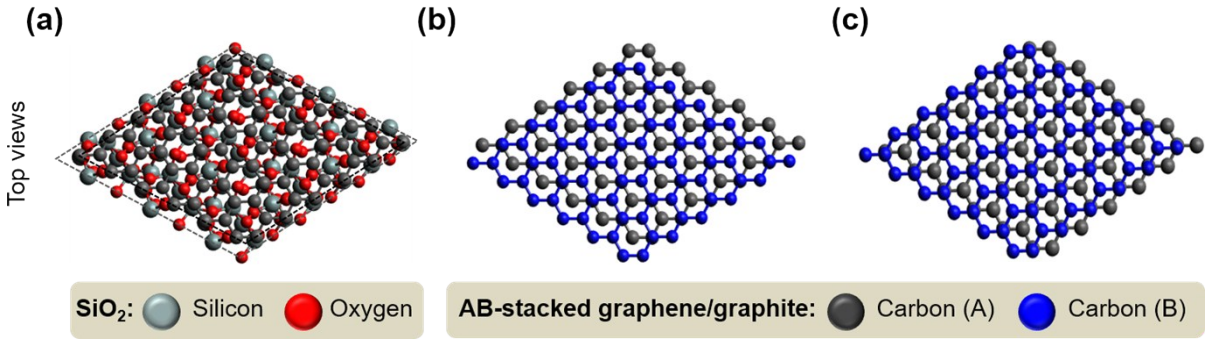

Figure S6. Molecular structures for calculating adhesion energies of various graphene layers. (a-c) Top-view configurations of (a) monolayer graphene on SiO_2, (b) bilayer graphene, and (c) bulk graphite.

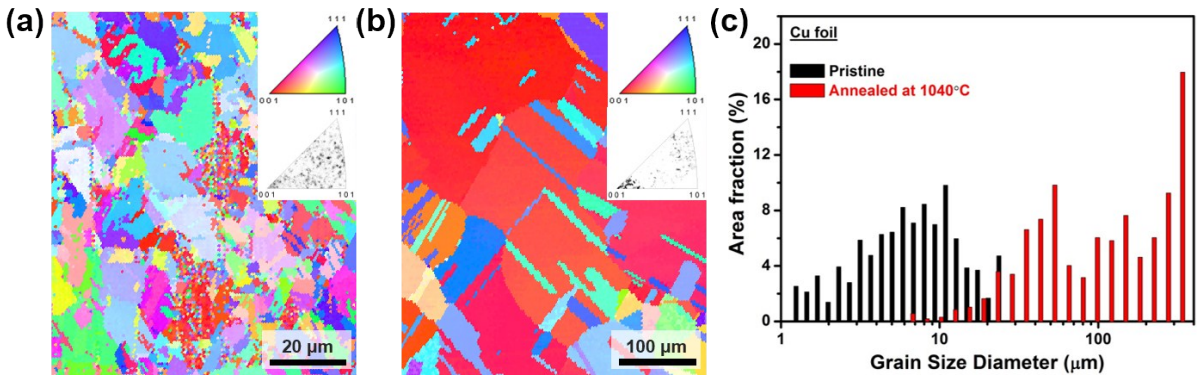

Figure S7. Grain structure of Cu foil. (a, b) Electron back-scattering diffraction (EBSD) pattern of (a) pristine Cu foil, and (b) Cu foil annealed at 1040°C for 1h. The top inset displays the grain color map, and the bottom inset displays the inverse pole figure of each pattern. (c) Area fraction of the grain size diameter for a pristine Cu foil (black) and Cu foil annealed at 1040°C (red).

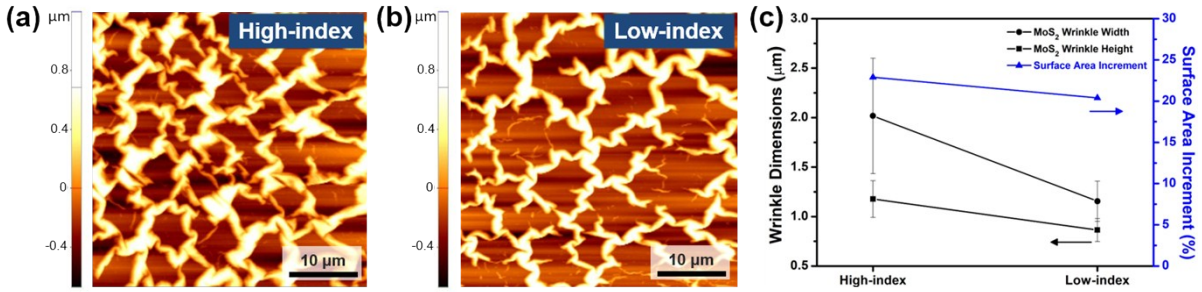

Figure S8. (a, b) AFM images of MoS_2 wrinkles synthesized on graphene originating from (a) high-index Cu crystal facets and (b) low-index Cu crystal facets. Graphs under each AFM image display representative height profiles. (c) Correlation of the Cu crystal facets with the MoS_2 wrinkle dimensions (left, black) and surface area increment relative to the geometric area (right, blue).

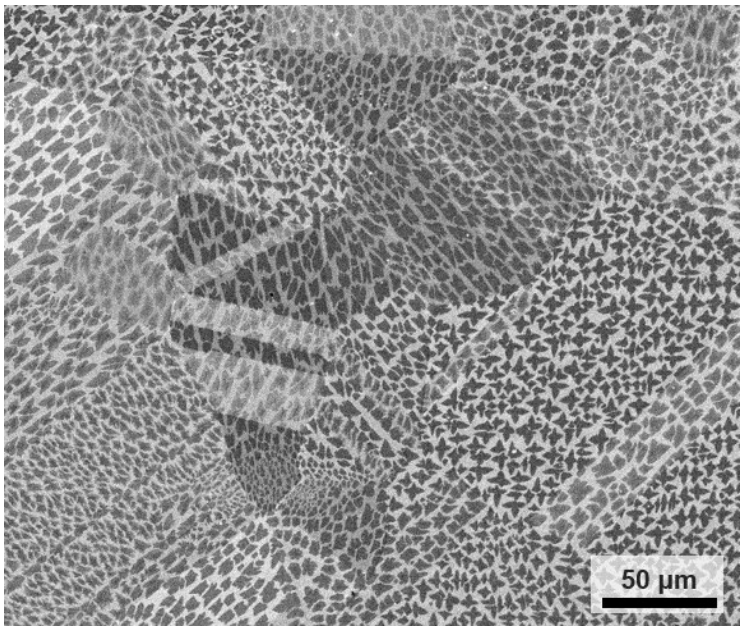

Figure S9. Nucleation behavior of graphene on a polycrystalline Cu surface. Cu was mildly oxidized to increase the visibility of individual graphene domains.

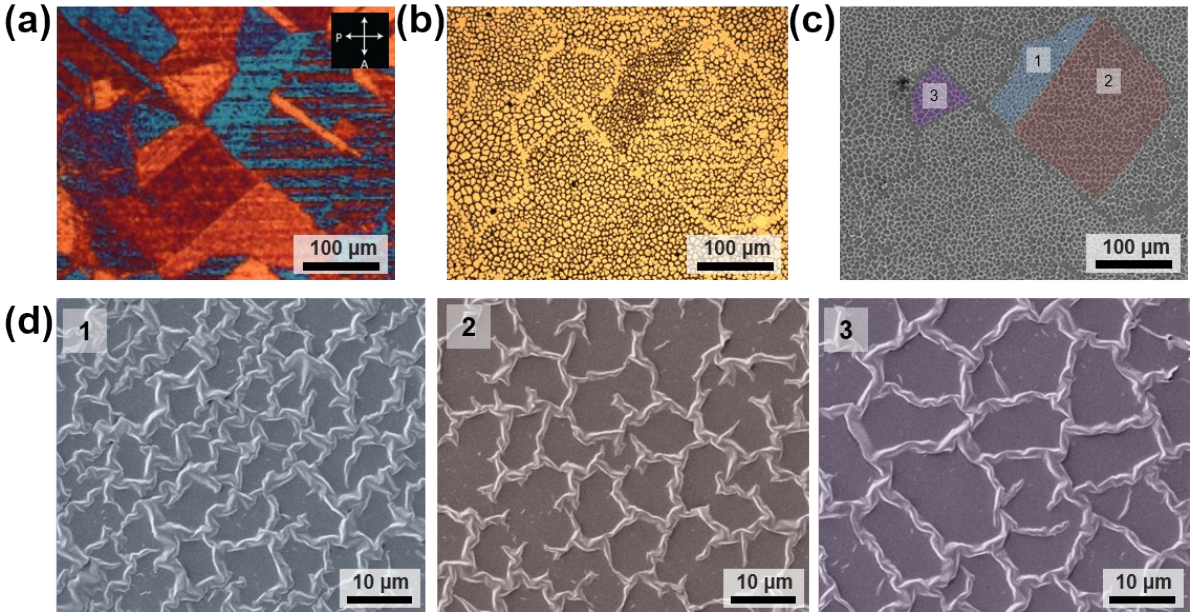

Figure S10. Synthesis and comparison of MoS_2 wrinkles on arbitrary graphene regions. (a) POM image of an arbitrary region of polycrystalline graphene coated with 5CB. (b) Optical image of MoS_2 wrinkle nanostructures at the same region as (a). (c) SEM image of MoS_2 wrinkle nanostructures at the region in (b) where areas originating from different Cu grains in (a) are highlighted in specific colors. (d) Magnified SEM images of the colored regions in (c), showing the distinct wrinkle structure for each region.

Supplementary Tables

Table S1. Density functional theory (DFT) calculations of the binding energy between SiO_2 and graphene layers.

	Stacking	Interlayer distance (Å)	Binding energy (meV/Å²)	Simulation tool
Monolayer Graphene*	AB	2.95	-7.15	LDA
Bilayer Graphene	AB	3.30	-9.57	
Bulk Graphite	AB	3.31	-18.61	
Monolayer Graphene*	AB	3.15	-14.05	vdW-DF2
Bilayer Graphene	AB	3.55	-18.24	
Bulk Graphite	AB	3.52	-39.80	

* Graphene sheets were placed on a SiO_2 substrate.

Table S2. Density functional theory (DFT) calculations of the binding energy of bilayer graphene with different stacking orientation.

	Stacking	Interlayer distance (Å)	Binding energy (meV/Å²)	Simulation tool
Bilayer Graphene	AA	3.70	-16.00	vdW-DF2
	AB	3.55	-18.24	