Supporting Information

Revealing the Crystalline Integrity of Wafer Scale Graphene on SiO₂/Si: An Azimuthal RHEED Approach

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Figure S1. The histogram of grain boundary mis-orientation angle for the as-sputtered Cu on sapphire(0001).



Figure S2. (a) SEM image of the ~900 nm as-sputtered Cu film on a spinel(111) substrate. (b) XRD azimuthal scans of Cu {111} at chi angle χ =70° and spinel {111} at chi angle χ =19° indicating the epi relationship between Cu film and spinel to be parallel, that is, Cu(111) [100] || spinel(111) [100].



Figure S3. (a) and (b) EBSD crystallographic orientation maps of the as-sputtered Cu film on a spinel(111) substrate using (a) IPF-Y mapping component and (b) IPF-Z component, respectively. (c) Histogram of grain boundary mis-orientation angles of the as-sputtered Cu film on a spinel(111) substrate.



Figure S4. (a) Cu {111} EBSD pole figure and (b) IPF along the normal direction of the assputtered Cu film on a spinel(111) substrate.



Figure S5. The Cu film on spinel(111) after the thermal annealing and graphene growth. (a) SEM image of the graphene on Cu surface; (b) and (c) EBSD crystallographic orientation maps using (b) IPF-Y and (c) IPF-Z mapping components, respectively; (d) EBSD Cu {111} pole figure showing a clean three-fold symmetry; (e) IPF-Z showing the crystallographic directions are clustered in the <111> direction along the normal direction of sample; (f) Histogram of grain boundary misorientation angles showing only small angle boundaries (<5°).



Figure S6. (a) Optical image of the bilayer graphene sheet transferred from a twinned Cu(111) film to a SiO₂/Si substrate. (b) Optical image of the multilayer graphene sheet transferred from a twinned Cu(111) film to a SiO₂/Si substrate. The dashed outlines are speculated to reflect the twin boundaries in Cu films.



Figure S7. (a) SEM image and (b) Raman spectra of the multilayer graphene used in Figure 6.



Figure S8. WinWulff simulation of $\{10\overline{1}1\}$ poles of single-crystalline graphite projected at 72° chi angle on the (0001) crystal plane. The indices in green are for 3-digit indexing.