Supporting Information for

In-situ formed nanoparticles assistant growth of large-size single crystalline h-BN on copper

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Figure S1. Optical images of h-BN at different heating temperature in the oxideassisted CVD growth: a) 50°C, b) 55°C, c) 60°C, respectively.



Figure S2. a) AFM image of h-BN triangle transferred onto SiO₂/Si substrates. b) Lowmagnification image of an h-BN domain on a carbon-covered TEM grid and hexagonal electron diffraction pattern (inset). c) Raman spectrum of h-BN on SiO₂/Si substrate. d) XPS spectra of B 1s (left) and N 1s (right) of h-BN film with binding energy peaks at 190.1 eV and 397.8 eV, respectively. e) UV–vis spectrum of a monolayer h-BN film on transparent quartz substrate. f) OBG calculation plot from e).



Figure S3. Optical images of h-BN at different annealing time: a) 20 min, b) 60 min, c) 180 min, respectively. All Copper foils are preheated in Ar and H₂ mixture as in conventional method. d) The relation between areal nucleation density of h-BN and annealing time.



Figure S4. More examples for typical nanoparticles locating in the center of h-BN.



Figure S5. Histogram of nanoparticles height at various annealing times. These data are extracted from the corresponding AFM images in Figure 5.



Figure S6. The morphology of copper foil after preheating in Ar and H_2 mixture (namely without pre-oxidation) but annealing at hydrogen concentration of 18.6% for a) 5min, b) 20 min, c) 40 min and d) 180 min. e) The evolutions of nanoparticles size with annealing time.



Figure S7. a) The morphology of pre-oxidized copper foil with annealing at hydrogen concentration of 18.6% for 180 min. b) Corresponding optical image after growth, the nucleation density of BN rises to 1.1×10^4 nuclei/mm². c) The evolution of density of h-BN and nanoparticles (> 15 nm) density with annealing time.



Figure S8. Orientation dependence of triangular h-BN domains grown on different Cu facets. a) EBSD mapping image of polycrystalline Cu foil. b) Corresponding low-magnification SEM image and c) Optical image of h-BN domains on different Cu facets. d-f) Magnified SEM images of h-BN on Cu (111), Cu (001), Cu (101) facet respectively, showing the orientation between h-BN and Cu foil. g-h) Statistical distributions of relative angle of individual h-BN domains on each Cu facets.