Supplementary Information

Large scale epitaxial graphite grown on twin free nickel(111)/spinel substrate

Zonghuan Lu, Xin Sun, Yu Xiang, Gwo-Ching Wang, Morris A. Washington, and Toh-Ming Lu Department of Physics, Applied Physics, and Astronomy, and Center for Materials, Devices, and Integrated Systems (cMDIS), Rensselaer Polytechnic Institute, Troy, New York 12180, the United States



Figure S1. (Top) The low pressure CVD system setup and (bottom) the process diagram for the growth of graphite film on epitaxial Ni(111) film on spinel(111) substrate.



Figure S2. AFM image of epitaxial Ni(111) surface. The film was DC sputtered at 475 °C on spinel(111) substrate



Figure S3 Raman G, 2D, and G/2D maps for graphite grown on Ni (111) at 925 °C before transferring to SiO_2/Si substrate. The scanning area is 20 x 20 μ m.



Figure S4. AFM images and roughness characterizations of graphite films on Ni(111) before transferring to SiO₂/Si substrate. The graphite films were grown at (a) 925 °C, (b) 1000 °C, and (c) 1075 °C, respectively. Note the scale bar is 10 μ m.



Figure S5. (a) AFM image and (b) Raman spectrum of a graphite on Ni(111) sample grown at 925 °C, after the surface has been treated using a UV ozone cleaning process for 10 min to completely remove graphite from Ni (111) substrate.



Figure S6. Representative AFM images of graphite edges after transferred to SiO₂/Si substrate. The films were grown at different temperatures: (a) 850 °C, (b) 925 °C, (c) 1000 °C, and (d) 1075 °C, respectively. The inset for each image is the AFM line scan across the edge showing thickness of the graphite.



Figure S7. EBSD pole figure and XRD $2\theta \cdot \omega$ scan of a commercial HOPG sample. (a) The EBSD ($10\overline{11}$) pole figure with color contour to show the strength of a fiber texture and (b) the XRD $2\theta \cdot \omega$ scan for the HOPG sample with 2θ ranging from 10° to 90° .



Figure S8. (a) EBSD IPF-Z map shows the out-of-plane crystal orientation distribution for the graphite film grown at 925 °C. The graphite film is transferred to SiO₂/Si substrates before characterization. (b) EBSD IPF-X map shows in-plane crystal orientation distribution for the graphite film. The insets are the EBSD color legends for a hexagonal crystal system.



Figure S9. XRD 360° azimuthal Φ scans of graphite (10¹1) peaks for graphite films grown at (a) 1000 °C and (b) 1075 °C. The χ angle was set at 72.4° for the Φ scans. The XRD data were collected from graphite films after transferring to SiO₂/Si substrates.