Electronic Supplementary Material (ESI) for RSC Advances.

Supplemental information for

Toward Clean Suspended CVD Graphene

Alexander Yulaev[1,2,3]*, Guangjun Cheng*[4]*, Angela R. Hight Walker*[4]*, Ivan V. Vlassiouk*[5]*, Alline Myers*[1]*, Marina S. Leite*[2,6]*, and Andrei Kolmakov*[1]*

[1]Center for Nanoscale Science and Technology, NIST, Gaithersburg, MD 20899, USA;

[2]Department of Materials Science and Engineering, University of Maryland, College Park, MD 20742, USA;

[3]Maryland NanoCenter, University of Maryland, College Park, MD 20742, USA;

[4]Physical Measurement Laboratory, NIST, Gaithersburg, MD 20899, USA;

[5]Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA;

[6]Institute for Research in Electronics and Applied Physics, University of Maryland, College Park 20742, USA

Keywords: graphene transfer, anthracene, purity, scanning electron microscopy

*To whom correspondence should be sent: andrei.kolmakov@nist.gov

Anthracene thermal evaporation

The anthracene film was deposited onto a Gr/copper stack by thermal evaporation in an evacuated glass test tube. The copper foil with CVD-grown graphene was tightly attached to the copper supporting tube. The copper tube was used for pumping of the test tube before anthracene deposition, as well as a cold finger for the substrate. In order to grow a uniform anthracene film, the copper tube was cooled with liquid nitrogen during the anthracene deposition. The bottom part of the glass tube was filled with 1 mm size anthracene particles (purity ≥ 99 %), pumped to ≈ 10^{-3} Pa, and then immersed into a boiling water bath to evaporate anthracene. After 30 min of anthracene evaporation, 14 µm ± 7 µm thick layer of anthracene film has been deposited onto the Gr-copper specimen. To avoid water condensation onto the anthracene film upon venting, the glass tube with the sample was constantly evacuated until the temperature of the copper tube and mounted sample reached room temperature.

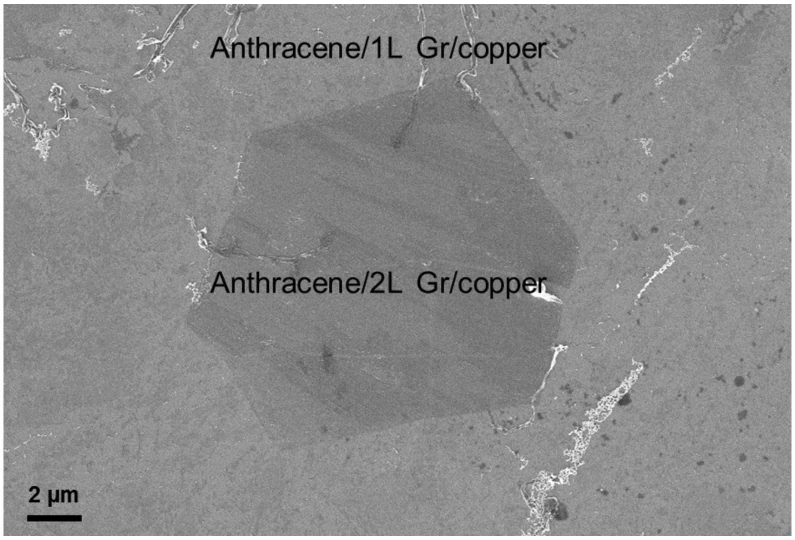

Figure S1. SEM image of an anthracene film thermally evaporated on Gr/copper at ≈ -20 °C. The hexagon in the middle of the image is the island of two-layer Gr. Both single and two-layer graphene are buried under the anthracene film. The film deposited onto a substrate at room temperature tends to form an incomplete film of weakly bound crystallites as large as few tens of μm (Figure S2). Capillary forces can easily destroy such a sacrificial layer during wet copper etching, which would make the transfer problematic.

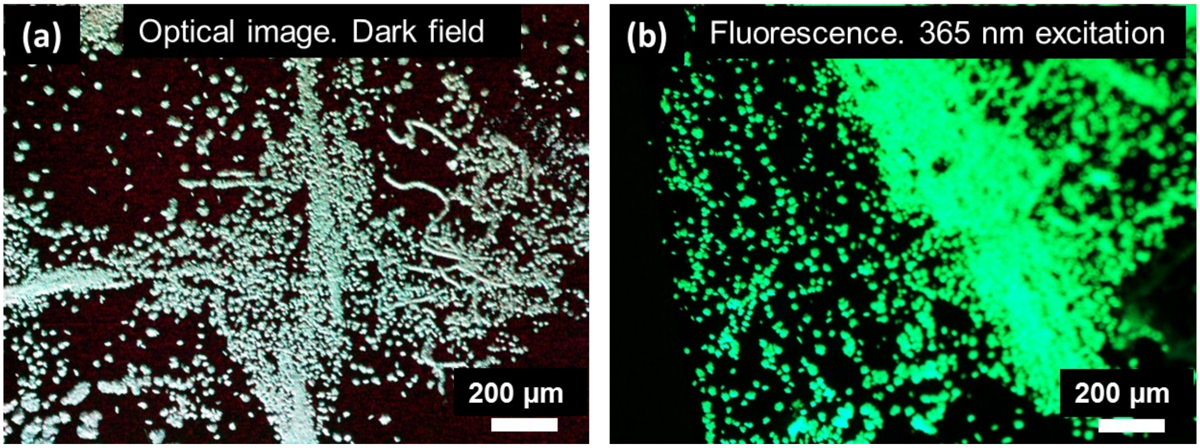

Figure S2. The morphology of the anthracene film deposited onto Gr/copper stack held at the room temperature. (a) Dark field optical image. The dark areas correspond to pristine graphene on copper. (b) Fluorescent microscopy image of the same region as in (a) excited by 365 nm UV light.

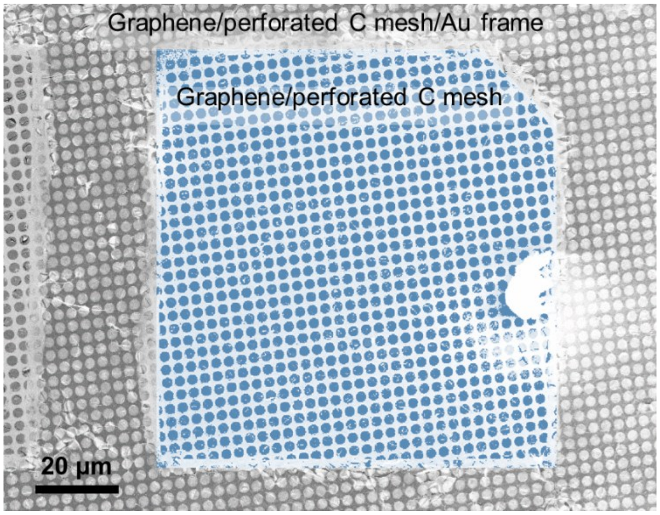

Figure S3. SEM image showing perforated carbon mesh covered with as transferred suspended graphene (false blue color) Surface coverage **(≈ 95 % before anthracene cleaning procedures)** was calculated as the ratio of orifices covered with graphene to all orifices in carbon mesh.

Raman Characterization

Raman spectra were acquired under ambient conditions with a micro-Raman spectrometer equipped with a 514.5 nm (2.41 eV) wavelength excitation laser and a grating with a 1800 mm^{-1} pitch, while operating in 180° backscattering geometry. A 50× objective was used to focus the excitation laser to an approximately 1 µm spot onto the sample with an incident power of less than 2 mW to avoid local heating effects.

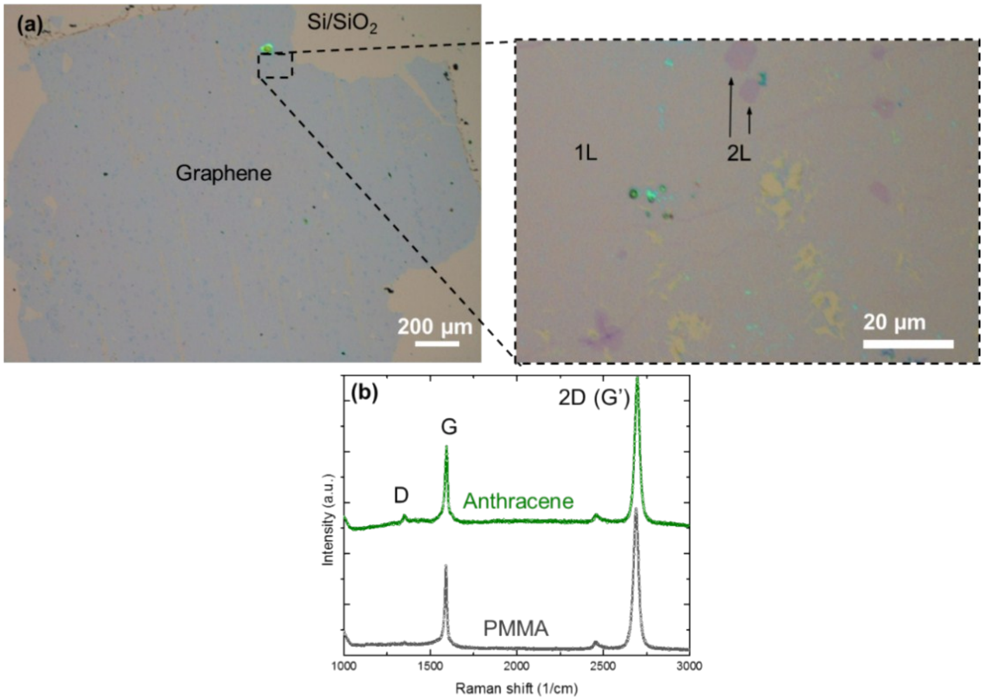

Figure S4. Optical image of graphene transferred on SiO$_2$ and Raman analysis. (a) Optical image of CVD Graphene transferred by anthracene onto SiO2 (285 nm)/ Si(100) substrate (see Figure 1a, (vi)), and (b) Raman spectra of graphene transferred by PMMA and anthracene onto SiO2 (285 nm)/Si(100) substrate. The D-peak (1360 cm^{-1}) indicates the defect density of graphene layers, the G-peak (1580 cm^{-1}) is due to Stokes phonon energy shift of the in-plane vibrational mode, and the 2D (G')-peak (2690 cm^{-1}) is the second-order overtone of another in-plane vibration. The wavelength of the excitation laser was 514.5 nm.

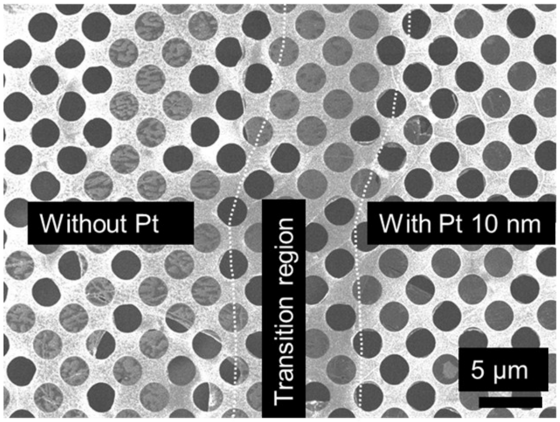

Figure S5. Spillover of Pt catalysis. SEM image of suspended graphene transferred by PMMA on a carbon mesh with a pre-deposited 10 nm Pt layer (right side) and without Pt (left side). Both the region with and without Pt are separated by a transition section (in the center) which is not covered by Pt, but represents graphene membranes cleaned by hydrogen diffused here from the Pt area.

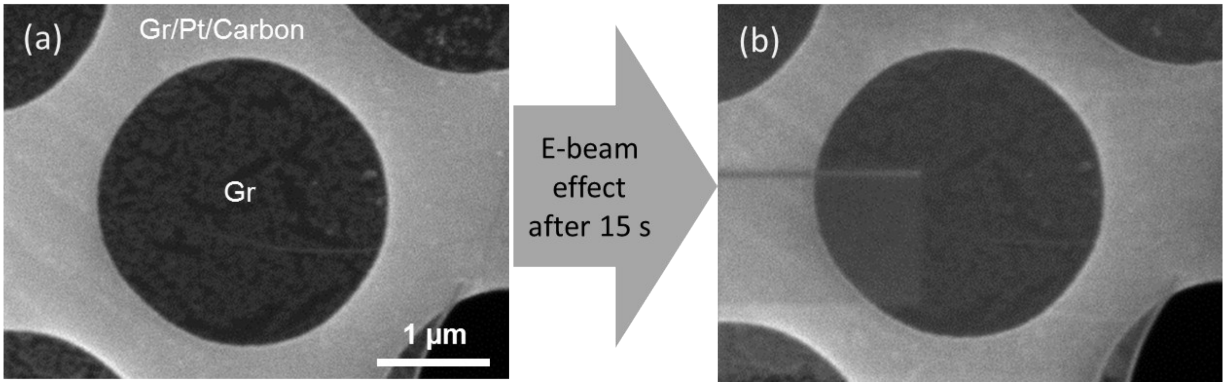

Figure S6. E-beam induced carbon contamination of a graphene membrane transferred by PMMA onto a carbon mesh with Pt and annealed on a hot plate.

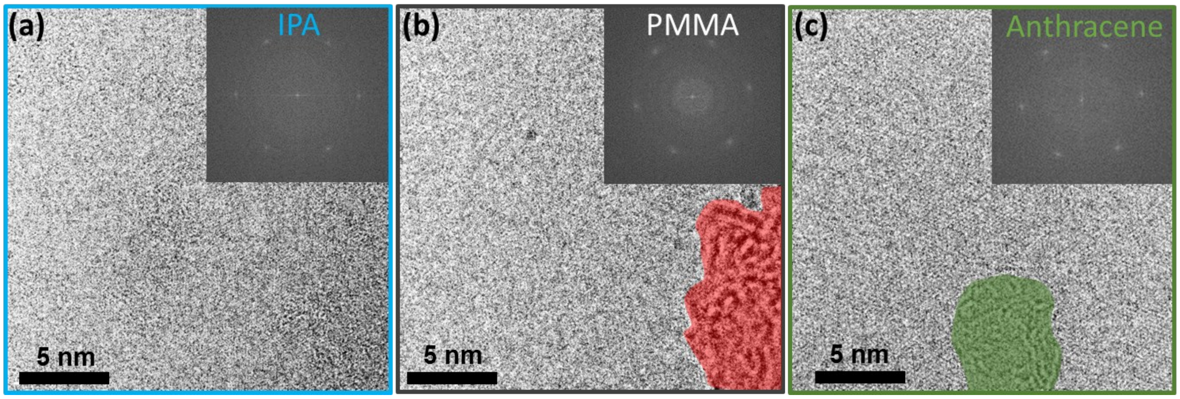

Figure S7. HRTEM images and fast Fourier transforms (FFTs) (insets) of graphene transferred by IPA (a), PMMA (b), and anthracene (c) onto a carbon mesh with Pt catalyst. Color coded regions have been excluded from FFT analysis.