Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.
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Supporting Information

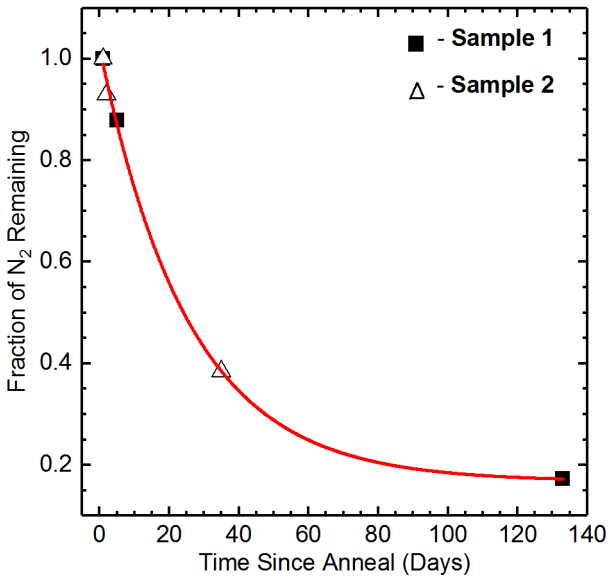

Figure S1. A graphical representation of the life-span of N_2 filled GNBs from two separate samples (1 and 2) after prolonged exposure to atmospheric conditions. Day 0 represents the first measurement after NH_3 treatment.

Graphene nanobubble life-span

XPS results of two separate NH_3 treated samples (labelled Sample 1 and Sample 2 in Figure S1) over several months lead us to believe that the GNBs persist in this time however N_2 is released gradually, following an exponential fit. Evidence of the N_2 peak was still visible over four months after the initial treatment, reduced from ~4.9 at. % on Day 0 to 0.8 at. % on Day 133. Fraction of N_2 was calculated using N_A atomic percentage relative to the C 1s sp^2 on Day 0 as 1.0 (~4.9 at. %), and consequent measurements were measured in relation to that percentage.

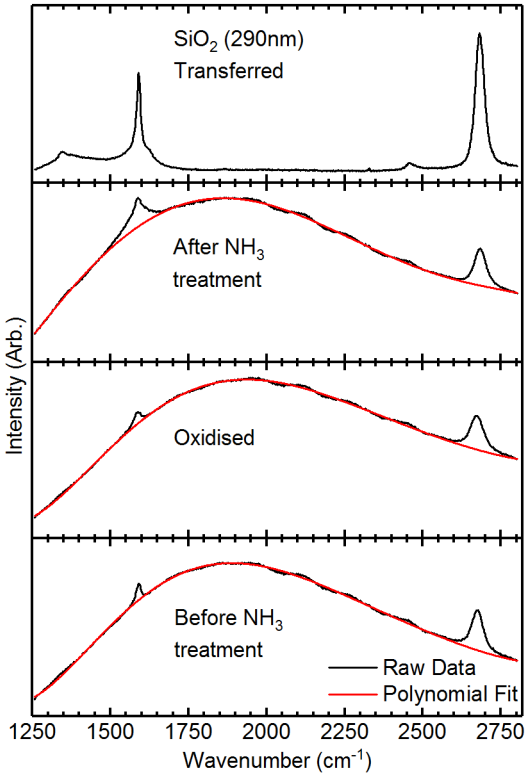

Figure S2. Raw Raman data collected from CVD graphene samples, prior to subtraction. A 6th order polynomial has been fitted to the photoluminescent background of the Cu substrate. The appearance of the *G* (~1580 cm^{-1}) and *2D* (~2700 cm^{-1}) peaks in each scan are characteristic to graphene.

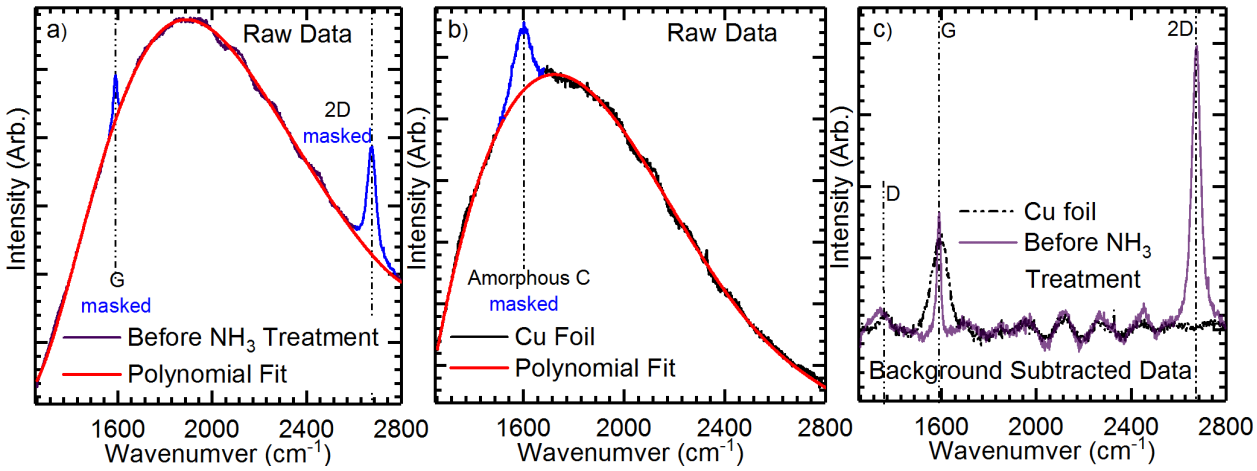

Figure S3. Raman spectra of (a) before NH_3 treatment of graphene on a Cu substrate, (b) Cu foil before background subtraction and (c) after background subtraction. The photoluminescent background in (a) and (b) is fitted with a 6th order polynomial. Blue indicates masking of peaks.

Raman analysis

Subtraction of the photoluminescent background caused by the Cu substrate was applied to all graphene/Cu sample datasets. This was done by masking the *D*, *G* and *2D* peaks (Figure S3, blue) and fitting the remaining data with a 6th order polynomial (red) (Figure S2 and S3a). Using this fit, the background was removed through a Python program. The research made use of the following Python packages: NumPy [1], Matplotlib [2], AstroPy [3,4] The oscillating component of the background is due to the photoluminescent background, which can also be seen in the spectrum of Cu foil (Figure S3b-c) and is present in all graphene on Cu spectra. The broad peak at ~1600cm^{-1} is attributed to amorphous carbon [5] on the surface of the Cu foil (Figure S3b-c). Comparison of background subtracted data of both the before NH_3 treatment graphene/Cu (purple) and Cu foil (black, dashed) demonstrate that the broad peak seen at ~1350 cm^{-1} is likely due to the Cu photoluminescent background, as it can be seen in both spectra (Figure S3c).

References

(1) Oliphant, T. {NumPy}: A Guide to {NumPy}. Trelgol Publishing 2006.

(2) Hunter, J. D. Matplotlib: A 2D Graphics Environment. *Comput. Sci. Eng.* **2007**, *9* (3), 90–95.

(3) Astropy Collaboration; Price-Whelan, A. ~M.; Sip\H ocz, B. ~M.; Günther, H. ~M.; Lim, P. ~L.; Crawford, S. ~M.; Conseil, S.; Shupe, D. ~L.; Craig, M. ~W.; Dencheva, N.; et al. The Astropy Project: Building an Open-Science Project and Status of the v2.0 Core Package. *AJ* **2018**, *156*, 123.

(4) Astropy Collaboration; Robitaille, T. ~P.; Tollerud, E. ~J.; Greenfield, P.; Droettboom, M.; Bray, E.; Aldcroft, T.; Davis, M.; Ginsburg, A.; Price-Whelan, A. ~M.; et al. Astropy: A Community Python Package for Astronomy. *AAP* **2013**, *558*, A33.

(5) Dychalska, A.; Popielarski, P.; Franków, W.; Fabisiak, K.; Paprocki, K.; Szybowicz, M.

Study of CVD Diamond Layers with Amorphous Carbon Admixture by Raman Scattering Spectroscopy. *Mater. Sci. Pol.* **2015**, *33* (4), 799–805.