Supplementary Information

Quantifying the Effect of Nanosheet Thickness on the Piezoresistive Response in Printed Graphene Nanosheet Networks

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Network Thickness Measurement using White Light Interferometry

Film thickness measurements were carried out using the non-contact optical white light interferometry (WLI) technique. This technique illuminates the sample with a low coherence light source and using a Mirau lens images the interference fringes arising from different heights on the sample. During the measurement the sample is moved in the z-axis relative to the lens, which moves the interference fringes. The fringes in WLI have maximum contrast at the point of perfect focus, and so the point of perfect focus for each pixel in the image, based on the z-scan position at which it occurs. This can then be reconstructed into a 3D surface and analysed using the ProfilmOnline software. On order to measure step heights accurately, scratches in the films were imaged to see both the top surface of the film and the substrate below. Taking a scan of this area and applying a levelling on the substrate enables the use of the Histogram function for step height, where the height of each pixel is plotted on a histogram and the distribution of surface heights can be observed. For a thin film, this histogram typically shows tow peaks, one associated with the substrate and a second associated with the top surface of the film. Taking the difference between the position of both peaks gives the film thickness.

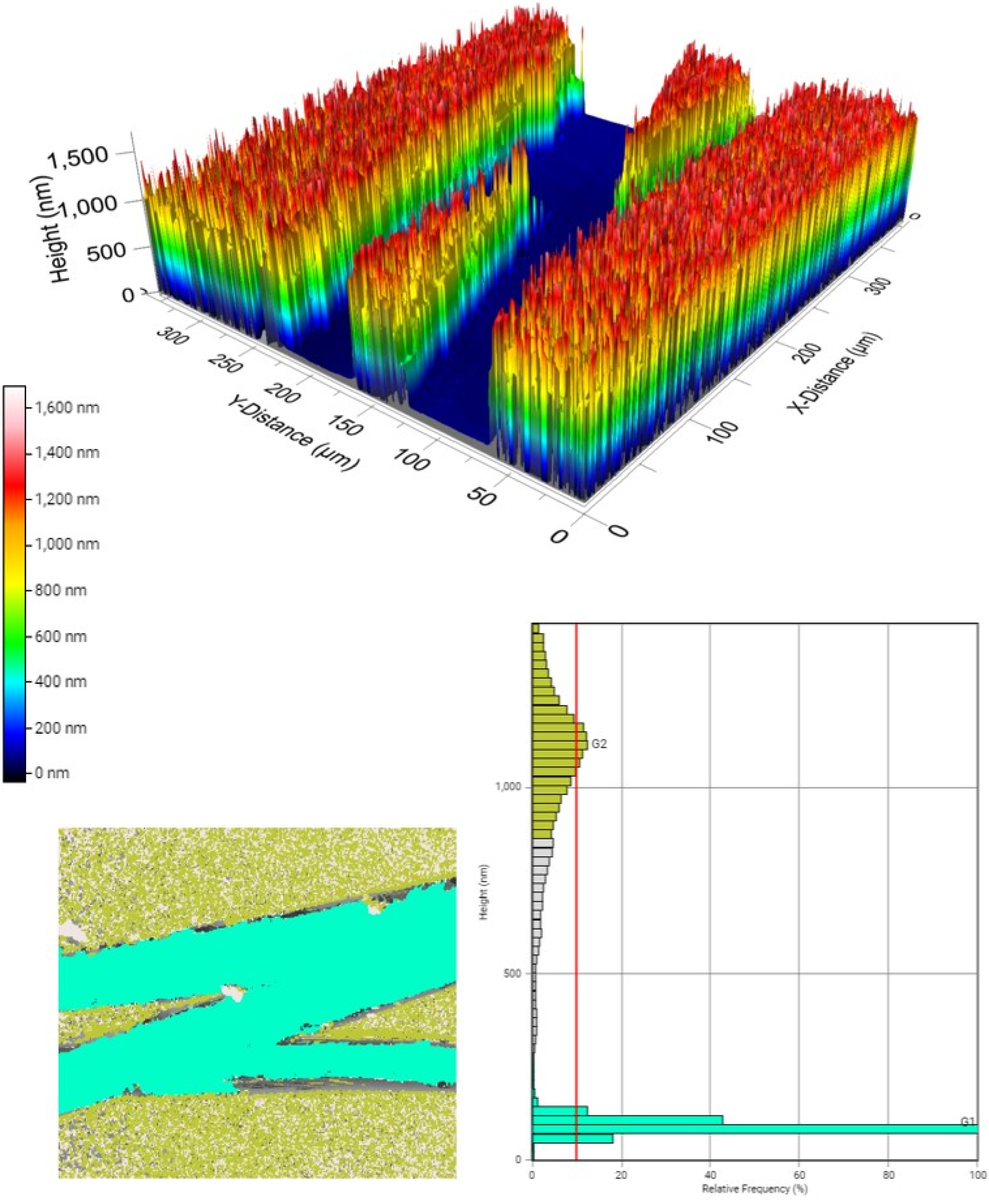

Figure S1: 3D representation of the surface of a nanosheet network, scratched to the substrate reconstructed from white light interferometry data. Below the histogram of pixel height is shown, in which two distinct distributions of heights are observed, one represents the substrate and the other the top surface of the network.

Cyclic Strain Testing

Cyclic strain testing of the piezoresistors was conducted to investigate the distribution of gauge factors for each of the devices, as will be outlined in later sections. But in order to extract this data, each device was cycled using a triangular sawtooth strain profile, from 0% to 0.5% strain. At a strain rate of 0.2 %/s for 200 cycles. The electronic response of each device is shown below, for clarity, each profile is normalised to the initial resistance of the device.

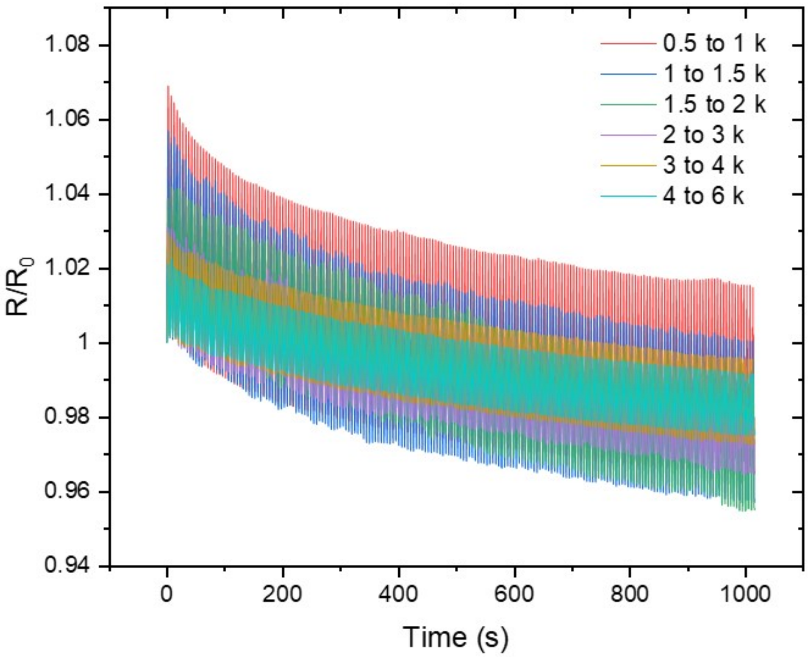

Figure S2: Piezoresistive response to cycling for 200 cycles at 0.5% strain, 0.2%/s using a triangular sawtooth profile for a device from each size fraction. The resistances are normalised to the zero-strain resistance measured before the first straining cycle.

Gauge Factor Distribution from Cyclic Data

In order to extract gauge factor values from cycling data in the linear regime, the resistance vs. time graph is plotted in OriginPro 2022b. The region of interest is chosen using the 'data selector' function. In this study that was the region after any initial decay in resistance associated with conditioning. Once the region was selected, it was analysed to identify peaks using the 'Peak Analyzer', with the goal defined to be 'find peaks'. To find the maximum and minimum point of each cycle, the baseline was taken to be the mean of the data set. Peaks were identified in both directions using a local maximum method with at least 5 local points. This reduced the likelihood of finding false peaks as a result of noise in the electrical signal. Additionally, peaks were filtered by 'height %' with a threshold height of 20%. Once completed, this operation outputs a list of resistance values and the timestamp associated with each. As a sanity check, the difference between time stamps should be equal to half the cycling period. The gauge factor can then be calculated by using the following equation.

$$G_{n,\varepsilon} = \left(\frac{R_{n,\varepsilon} - R_{n,0}}{R_{n,0}} \right) \Big/ \varepsilon$$

Where the subscript n describes which cycle the value is associated with, ε is the maximum strain applied, and 0 signifies that the measurement is at the unstrained position. From this analysis, a distribution of gauge factor values can be extracted for each device. This enables a method of testing the consistency of individual samples, along with minimising the potential error in measured gauge factor due to signal variations in the electrical response from a single measurement, which can be significant especially in the case of low gauge factor samples.

Nanosheet Thickness

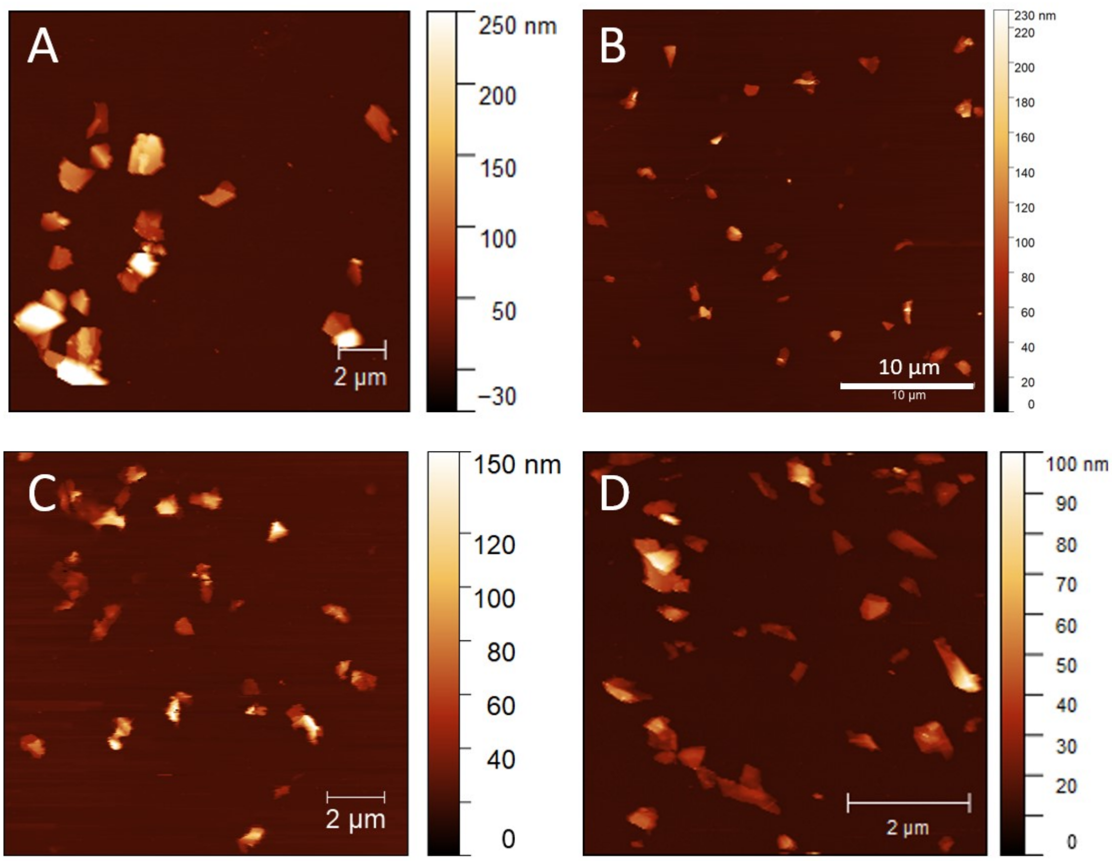

Figure S3: Representative AFM images of flakes from across the range of size selected fractions. A) 0.5 to 1 kRPM, B) 1 to 1.5 kRPM, C) 2 to 3 kRPM, D) 3 to 4 kRPM.

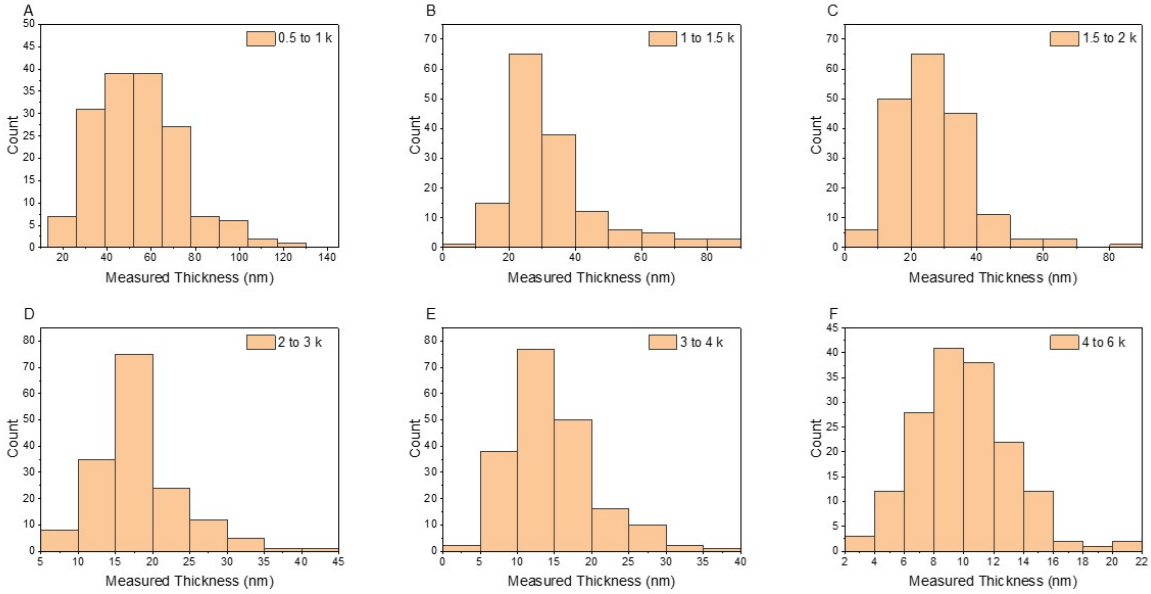

Figure S4: Measured apparent thickness distribution histograms for nanosheets measured using AFM from all six size fractions. A) 0.5 to 1 kRPM, B) 1 to 1.5 kRPM, C) 1.5 to 2 kRPM, D) 2 to 3 kRPM, E) 3 to 4 kRPM, F) 4 to 6 kRPM.

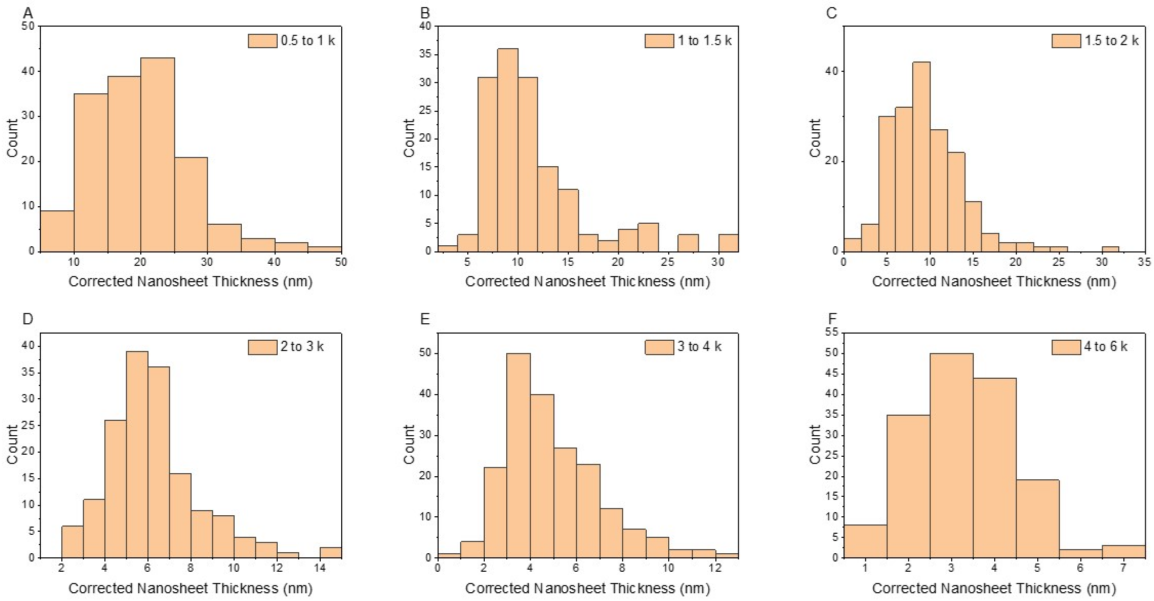

Figure S5: Converted nanosheet thickness distribution histograms, converted from the measured thicknesses (Figure S4) for all six size fractions. A) 0.5 to 1 kRPM, B) 1 to 1.5 kRPM, C) 1.5 to 2 kRPM, D) 2 to 3 kRPM, E) 3 to 4 kRPM, F) 4 to 6 kRPM.

Table S1: Nanosheet sizes determined from AFM for each size fraction.

Centrifugation Trapping Speeds	L_{NS} (µm)	Thickness before correction (nm)	T_{NS} (nm)
0.5 to 1 kRPM	1.50	54.6	19.7
1 to 1.5 kRPM	1.30	32.4	11.6
1.5 to 2 kRPM	0.83	27.0	9.6
2 to 3 kRPM	0.76	17.9	6.2
3 to 4 kRPM	0.50	14.4	4.9
4 to 6 kRPM	0.45	9.8	3.2

Model Derivation

For a nanosheet network ($t>t_x$)

$$\rho_{Net} \approx \frac{\left[\rho_{NS} + 2t_{NS}R_J\right]}{(1-P_{Net})}\left[1+\frac{2}{n_{NS}t_{NS}l_{NS}^2}\right]$$

For graphene, the carrier density, n_{NS} is large so the above expression can be simplified to

$$\rho_{Net} \approx \frac{\left[\rho_{NS} + 2t_{NS}R_J\right]}{(1-P_{Net})}$$

The gauge factor of a naetwork can be written as

$$G_{Net} = 2+\frac{1}{\rho_{Net}}\frac{d\rho_{Net}}{d\varepsilon}$$

We see that the strain derivitive of network strain is a key parameter, and so we take this derivitive of the above expression for network resistivity.

$$\rho_{Net} \approx [\rho_{NS} + 2t_{NS}R_J](1-P_{Net})^{-1}$$

$$\frac{d\rho_{Net}}{d\varepsilon} \approx [\rho_{NS} + 2t_{NS}R_J]\frac{d}{d\varepsilon}(1-P_{Net})^{-1} + (1-P_{Net})^{-1}\frac{d}{d\varepsilon}[\rho_{NS} + 2t_{NS}R_J]$$

$$= [\rho_{NS} + 2t_{NS}R_J](1-P_{Net})^{-2}\frac{dP_{Net}}{d\varepsilon} + (1-P_{Net})^{-1}\left[\frac{d\rho_{NS}}{d\varepsilon} + 2t_{NS}\frac{dR_J}{d\varepsilon}\right]$$

$$= \frac{[\rho_{NS} + 2t_{NS}R_J]}{(1-P_{Net})^2}\frac{dP_{Net}}{d\varepsilon} + \frac{[d\rho_{NS}/d\varepsilon + 2t_{NS}dR_J/d\varepsilon]}{(1-P_{Net})}$$

Substituting this derivative into the expression for network gauge factor, yields.

$$G = 2 + \frac{\frac{[\rho_{NS} + 2t_{NS}R_J]}{(1-P_{Net})^2}\frac{dP_{Net}}{d\varepsilon} + \frac{[d\rho_{NS}/d\varepsilon + 2t_{NS}dR_J/d\varepsilon]}{(1-P_{Net})}}{\frac{[\rho_{NS} + 2t_{NS}R_J]}{(1-P_{Net})}}$$

$$G = 2 + \frac{[\rho_{NS} + 2t_{NS}R_J]\frac{dP_{Net}}{d\varepsilon} + (1-P_{Net})[d\rho_{NS}/d\varepsilon + 2t_{NS}dR_J/d\varepsilon]}{(1-P_{Net})[\rho_{NS} + 2t_{NS}R_J]}$$

Further simplifyng we are left with a three term expression for network gauge factor.

$$G = 2 + \frac{\frac{dP_{Net}}{d\varepsilon}}{(1-P_{Net})} + \frac{[d\rho_{NS}/d\varepsilon + 2t_{NS}dR_J/d\varepsilon]}{[\rho_{NS} + 2t_{NS}R_J]}$$

Dividing the third term through by ρ_{NS}, and modifying the second term using the relation

$$\frac{\frac{dP_{Net}}{d\varepsilon}}{(1-P_{Net})} \approx -\frac{d\ln(1-P_{Net})}{d\varepsilon}$$

$$G_{Net} = 2 - \frac{d\ln(1-P_{Net})}{d\varepsilon} + \frac{\left[\frac{1}{\rho_{NS}}\frac{d\rho_{NS}}{d\varepsilon} + \left(\frac{1}{R_J}\frac{dR_J}{d\varepsilon}\right)\left(\frac{2R_J}{\rho_{NS}}\right)t_{NS}\right]}{\left[1 + \left(\frac{2R_J}{\rho_{NS}}\right)t_{NS}\right]}$$

For fitting, the $\left(\frac{2R_J}{\rho_{NS}}\right)$ term can be extracted from the thickness dependent resistivity fit.