Supporting Information for

Suspended Monolayer Graphene under True Uniaxial Deformation

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S1. Sample preparation

![Microscopy image of graphene flake between PMMA layers. The sample is deposited onto a Si/300nm SiO$_2$ substrate (3.8x3.8 mm$^2$), pre-patterned with a 40x40 Au marking grid (each square is 80x80 µm$^2$), (b) Microscopy image of suspended graphene inside the e-beam lithography window (3µm). The sample is transferred on a glass substrate.]

(a) (b)

Figure 1

S2. Strain calculation from Raman Spectra

The applied strain is calculated from the G-band peak shift according to the following well established secular equation$^1$

$$\Delta \omega_G^x = \Delta \omega_G^y \pm \frac{1}{2} \Delta \omega_G^z = -\omega_G^0 \gamma_G (\epsilon_{ii} + \epsilon_{nn}) \pm \frac{1}{2} \omega_G^0 \beta_G (\epsilon_{ii} - \epsilon_{nn})$$

in which $\epsilon_{ii}$ and $\epsilon_{nn}$ are the parallel and perpendicular strain components, $\gamma_G$ is the Grüneisen parameter and $\beta_G$ is the shear deformation potential$^1$. The phonon wavenumber at rest is $\omega_G^0$.

From the above equation we have:

$$\begin{cases}
\Delta \omega_G^+ = -\omega_G^0 \gamma_G (\epsilon_{ii} + \epsilon_{nn}) + \frac{1}{2} \omega_G^0 \beta_G (\epsilon_{ii} - \epsilon_{nn}) \\
\Delta \omega_G^- = -\omega_G^0 \gamma_G (\epsilon_{ii} + \epsilon_{nn}) - \frac{1}{2} \omega_G^0 \beta_G (\epsilon_{ii} - \epsilon_{nn})
\end{cases} \Rightarrow$$
The numeric values of $\varepsilon_\parallel$ and $\varepsilon_\perp$ are calculated from the above general equations assuming $\gamma_G = 1.99$, $\beta_G = 0.99$ and $\omega_0^{0} = 1581 \text{cm}^{-1}$.

**Figure 2** $G^+$ (empty rectangles) and $G^-$ (full rectangles) peak positions for graphene embedded in polymer beam in tension. Labeled lines correspond to the predicted values for suspended graphene. This graph is taken from Frank et al. 2011, Nat Commun 2: 255.
**S3. Converting spectroscopic data into stress-strain.**

The procedure for converting the spectroscopic data into stress-strain is presented in the following. For carbon materials, the shift of the Raman frequency scales with the applied stress by a factor $k$:

$$|\Delta \omega| = k \sigma$$  \hspace{1cm} (1)

Where $\Delta \omega$ is the shift of the 2D Raman band, $k$ is a scale factor and $\sigma$ is the mechanical stress. In tension the shift frequency of the 2D Raman band, can be captured by a first order polynomial curve (eq. 2), thus:

$$\Delta \omega = f_0 + f_1 \varepsilon$$  \hspace{1cm} (2)

By combining equations (1) and (2), the spectroscopically derived Raman shifts can be converted into mechanical stresses, with the factor $k=5.75 \text{ cm}^1 \text{GPa}^{-1}$ for the 514 nm wavelength. Finally the mechanical stress can be estimated from the next equation:

$$\sigma = \frac{f_0}{k} + \frac{f_1}{k} \varepsilon$$  \hspace{1cm} (3)

where $f_i$ are the polynomial constants estimated from the fitting of the graphs of $\Delta \omega(\varepsilon)$, $\sigma$ and $\varepsilon$ are the mechanical stress and strain respectively and $k$ as defined previously.

**S4. Critical buckling strain**

With regards to compression, atomically thin membranes are not expected to have any compressive strength in air. The critical buckling strain for a flake in the classical Euler regime in air, can be determined through the following equation:

$$\varepsilon_c = \frac{\pi^2 k D}{C ab}$$  \hspace{1cm} (4)
where $a$ and $b$ are the length and width of the flake for axial compression, $k$ is a geometric term, and $D$ and $C$ are the bending and tension rigidities, respectively. The above equation, is mainly valid for suspended thin films and yields extremely small ($\sim 10^{-9}$) $\varepsilon_c$ values for graphene monolayers of thicknesses of the order of atomic radii. Such extremely small critical buckling strains are also predicted by molecular dynamics calculations. This certainly explains the tendency of graphene to wrinkle easily because of its evidently low resistance to buckling instability as expressed by (4).

References
