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Electronic Supplementary Material

Direct evaluation of CVD multilayer graphene elastic properties

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We briefly expose here the structural model used for the calculation of tube radius in our InGaAs/Cr/Graphene system. The model, which can be used for curvature calculations in a multilayer system is thoroughly explained in ref. [1]. In our case, we consider an elastic multilayer structure under conditions of plane strain (see Fig. S1):

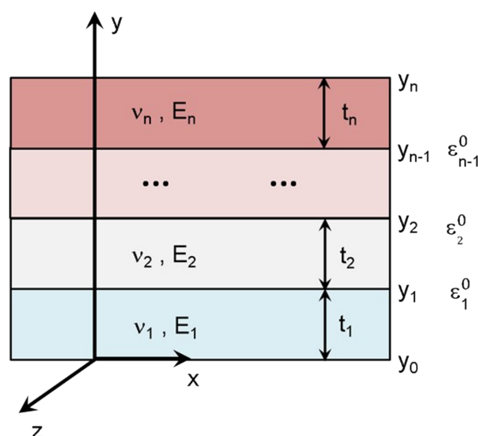

Fig. S1 – Representation of a multilayer structure defined within the frame of the analytical model used in our work [1]. The variables shown in the figure refer to elastic properties of each layer (E_i, ν_i), layer thickness (t_i), interface position (y_i) and interfacial strain (ε_i^0) for each film of the stack.

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The stacked film structure consists of n layers with thickness t_i, $i = 1, 2, ..., n$ and interfacial layer position y_i, i = 1, 2,..., n . Distinct materials can be considered, with layers characterized by specific Young's modulos E_i and Poisson's ratio ν_i. The layers are under the influence of initial strains ε_i^0, which may be due to lattice mismatch or due to thermal loading. Under the considered plane strain conditions full strain in the transverse direction z is equals to zero. Once we know a set of values of the parameters listed above (e.g., for InGaAs and Cr layers) we can fit the tube radius (Fig. 5, main text) as a function of Graphene parameters.

During the winding process of the strained thin films, separated from the substrate due to the etching of the sacrificial layer, the stress along the layer stack is accommodated by changing the interatomic distances within each film in order to minimize the elastic energy of the entire system. Although the forces in successive layers can have opposite directions, the formation of the tube indicates that their sum produces a torque that acts to roll the bilayer.

The starting point to be considered here is the total displacement of the layers that roll. In a multilayered tube the films remain connected to one end (flat region) of the sample. If we consider the circumference with tangential strain component ε_t, the outer surface of the tube has tensile strain. Inside the tube wall the strain sign changes according to the radial position, ranging from $\varepsilon_t < 0$ (for in-plane compression at the inner wall), to $\varepsilon_t > 0$ for in-plane expansion. Within this range there is a position where the resulting strain reaches zero, represented by y_b. It can be considered that the resulting strain distribution ε_t inside the tube wall is a linear function of the coordinate y and can be decomposed into a uniform component and a component which varies along the curvature:

$$\varepsilon_t = c + \frac{y - y_b}{R_i} \quad . \tag{1}$$

The parameter c is the uniform deformation component, corresponding to the scenario after the multilayer is released from the substrate, while R_i is the tube inner radius of curvature. A multilayer structure under plane strain conditions is considered here (the longitudinal strain ε_l is constant, denoting no relaxation along the tube direction), which is adequate whenever the system is wide enough in the longitudinal direction (tube axis) and presents limited (well-defined) bending in one direction.

Taking into account that the surface tangential stress is $\sigma_t = 0$ and using Hooke's law the :

$$\sigma_l = \nu\sigma_r - E\varepsilon^0, \tag{2}$$

$$\sigma_r = E\left(\varepsilon_t - \varepsilon^0\right) + \nu\sigma_l, \tag{3}$$

where the *r* subscript refers to the tube radial direction and ε^0 is the initial strain (for the flat layer condition, considered here due to the lattice mismatch between the InGaAs layer and the GaAs substrate). Here the resultant force in the longitudinal direction is not zero. However, this force does not affect the curvature of the rolled multilayer. Substituting equations 1 and 2 into equation 3 and making the variables changes $(1 + \nu) \rightarrow \eta$ and $\frac{E}{(1 - \nu^2)} \rightarrow E'$ one obtains:

$$\sigma_r = E'\left(c + \frac{y - y_b}{R_i} - \eta\varepsilon^0\right). \tag{4}$$

In order to determine the unknown parameters *c*, y_b , and *R* it is possible to write down three equilibrium conditions, which sum over the n-layers (each with sub-index *i*):

I – Force due to uniform strain

$$\sum_{i=1}^{n} E'_i t_i\left(c - \eta_i\varepsilon_i^0\right) = 0. \tag{5}$$

II – Force due to bending strain:

$$\sum_{i=1}^{n}\int_{y_{i-1}}^{y_i}\frac{E^{'}(y-y_b)}{R}dy=0 \quad (6)$$

III – Bending moment created by the normal stress in respect to bending axis:

$$\sum_{i=1}^{n}\int_{y_{i-1}}^{y_i}E^{'}_{i}\left(c+\frac{(y-y_b)}{R}-\eta_i\varepsilon_i^0\right)(y-y_b)dy=0. \quad (7)$$

The coordinates y_i used as integration limits above are as: $y_0 = 0$; $y_i = y_{i-1} + t_i$, with i =1, ... n. Solving these three equations one obtains the following solution for parameters c and y_b and radius of curvature R:

$$y_b=\frac{\sum_{i=1}^{n}E_i/(1-\nu_i^2)t_i(y_i+y_{i-1})}{\sum_{i=1}^{n}[E_i/(1-\nu_i^2)]t_i}, \quad (8)$$

$$c=\frac{\sum_{i=1}^{n}E_i/(1-\nu_i^2)t_i(1+\nu_i)\varepsilon_i^0}{\sum_{i=1}^{n}[E_i/(1-\nu_i^2)]t_i}, \quad (9)$$

$$R_i=\frac{2\sum_{i=1}^{n}[E_i/(1-\nu_i^2)]t_i[y_i^2+y_iy_{i-1}+y_{i-1}^2-3y_b(y_i+y_{i-1}-y_b)]}{3\sum_{i=1}^{n}[E_i/(1-\nu_i^2)]t_i(y_i+y_{i-1}-2y_b)[c-(1+\nu_i)\varepsilon_i^0}. \quad (10)$$

These three equations were implemented in a computer script, which is used to fit the data of Fig. 5.

Finally, the sensitivity of the fit parameters used in figures 5 and 4 of the manuscript are depicted in figures S2 and S3. Using the model described above one can fit the tube radius measured by SEM for different Cr layer thickness, using a fixed thermal strain of 2.03% between the InGaAs and Cr layers, with no strain between the Cr layer and the GLs (values of elastic constants and thickness for InGaAs and Cr layers are declared in the main text). Each graphene layer added to the layer stack is accounted as an additional layer of 0.4 nm thickness with elastic constants to be determined. We show below the result of varying individual parameters for a tube with 1170 nm radius (18.8 nm-thick Cr layer) and 1 graphene layer. The Young Modulus, Poisson's ratio and graphene layer thickness are varied slightly above the error bars, in order to expose the sensitivity of our fits to the experimental results (solid dots).

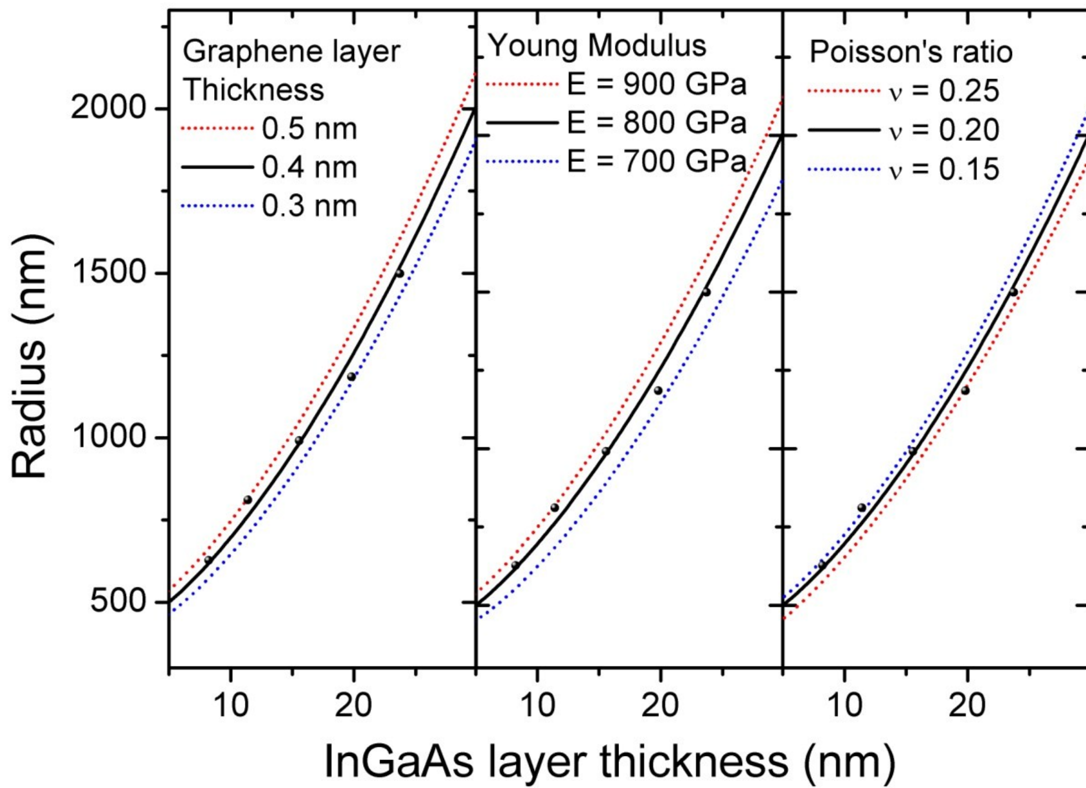

Fig. S2 – Result of varying fit parameters of the model described above for a series of tube radius (different Cr thickness). The best fit is provided by the black solid line, while dashed lines represent variations of individual parameters slightly above the error bars of the fitting procedure.

Finally, the sensitivity of the fitting of the X-ray data is depicted in Fig. S3. The model, thoroughly described in ref. [2] will not be reproduced here due to its strong specificity (requires single-crystalline layers and synchrotron radiation measurements). It is, however, crucial to mention that the radius of a given tube configuration strongly depends of the layer strain (the same 2.03 % initial strain between InGaAs and Cr layers is used), which implies on unambiguous tube radius, layer thickness and final tangential and radial strain distributions. In this sense, such parameters obtained by the elastic model of Figs. 5 (main text) and S2 are supported by XRD data, which makes the choice of parameters for fitting fig. 4 (main text) more robust. Figure S3 shows directly the effect that changes on the tube inner radius (1 graphene layer, 18.8 nm Cr thickness) impact on the fitting of experimental data and imply on distinct lattice parameter distributions. The fits shown already take into account the contributions of the three turns, which are seen separately in Fig. 4 of the main text.

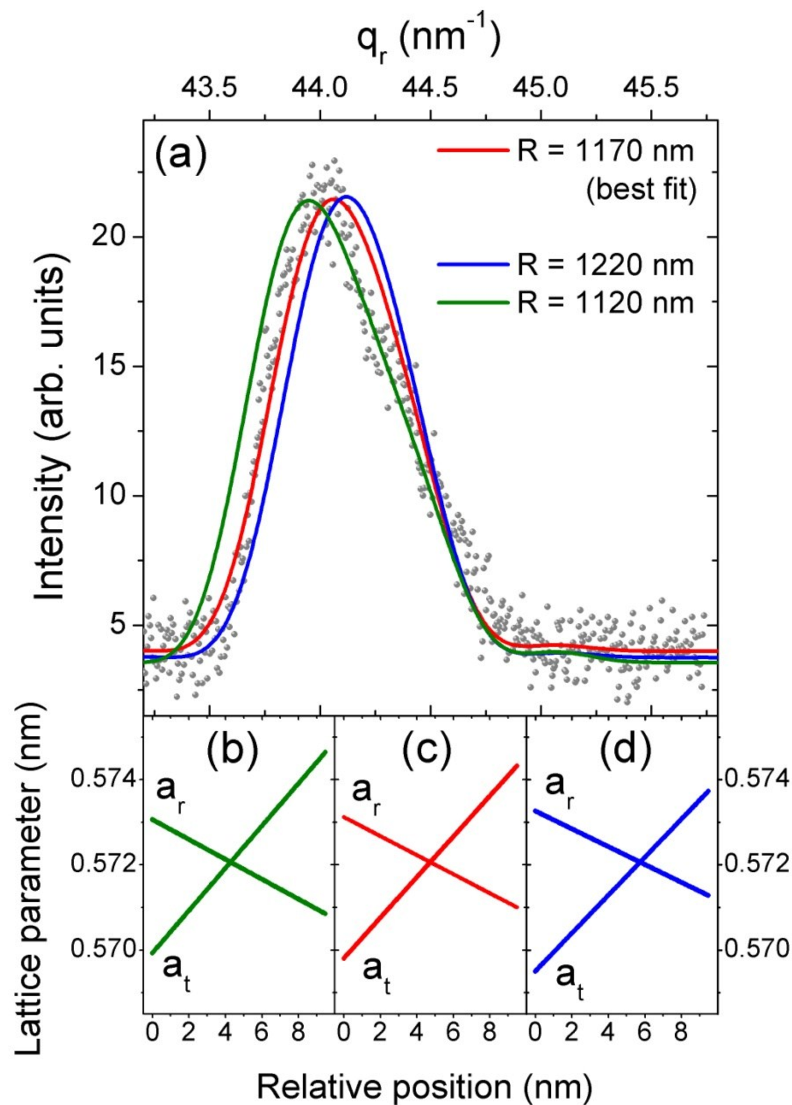

Fig. S3 – (a) fits (solid lines) of the experimental XRD data from a tube with 1 graphene layer (18.8 nm Cr film thickness). The best fit is shown in red for a tube radius of 1170 nm. (b) Lattice parameter profile for R = 1120 nm. (c) Lattice parameter profile for R = 1170 nm (best fit). (d) Lattice parameter profile for R = 1220 nm. All fits represent the sum of scattering contributions of three tube turns (see main text).

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