Non-linear dynamics for contactless characterization of graphene

Farbod Alijani[1], Dejan Davidovikj[2], Marco Amabili[3] and Peter G. Steeneken [1,2]

[1]*Department of Precision and Microsystems Engineering, TU Delft, The Netherlands*

[2] *Kavli Institute of Nanoscience, TU Delft, The Netherlands*

[3]*Department of Mechanical Engineering, McGill University, Montreal, Quebec, Canada*

Summary. The goal of this work is to develop a fast and accurate method for determining the Young's modulus of 2D materials exploiting their non-linear dynamics response. To demonstrate the method, we perform a series of measurements on strongly driven circular graphene membranes, showing the transition from linear to non-linear behavior. The non-linear response is approximated by a single Duffing oscillator, and harmonic balance method together with a least squares technique is used to identify non-linear parameters following an iterative approach. From the identified non-linear stiffness parameter and a geometrically non-linear model of the membrane, the Young's modulus is determined. Close agreement is observed between the Young's modulus obtained exploiting non-linear dynamics response and those obtained by AFM nanoindentation.

Introduction

Since graphene was first isolated, a plethora of two-dimensional (2D) materials have been discovered. The remarkable mechanical properties of pristine 2D nanomaterials have sparked interest for potential uses such as pressure, gas and mass sensors, representing the ultimate limits of 2D Nano-Electro-Mechanical Systems (NEMS) [1, 2]. Although promising, the development of 2D NEMS devices is still far from being considered well-established, predominantly due to the large variability that can be seen in mechanical properties obtained by available techniques [3].The conventional method for determining mechanical properties of suspended 2D materials is Atomic Force Microscopy (AFM) [4]. AFM operation requires mechanical contact between a sharp tip and the membrane that potentially leads to large stress and adhesion effects near the tip causing possible membrane fracture. Moreover, to get accurate measurements, large deflections are used, which can cause slippage, flattening of imperfections, and thus modifying the tension and Young's modulus of the 2D material. For studying the intrinsic mechanical properties of these materials, it is essential to characterize them at the smallest possible forces and strains, while still maintaining sufficient accuracy in determining the mechanical properties [3]. Therefore, this study focuses on developing a reliable non-contact method for characterizing the mechanical properties of 2D nanomaterials by utilizing their non-linear dynamics response.

Experimental procedure

Circular cavities are fabricated by etching holes in a gold-palladium (100 nm) and SiO_2 (285 nm) layer on a silicon wafer, resulting in cavities with a depth of 385 nm and a diameter of 5μm. In order to create circular nanodrums, a few-layer graphene flake is then transferred on top of the cavities using an all-dry transfer technique [5]. The sample is then mounted in a vacuum chamber to minimize damping by the surrounding gas. The flexural motion of the nanodrum is detected using an optical interferometer, which has been used previously in frequency and time-domain studies of nanomechanical properties of 2D materials [6]. All measurements are conducted at the center of the drum.
The drum's motion is probed by a Helium−Neon laser, and the intensity variations caused by the interfering reflections from the moving membrane and the fixed silicon substrate underneath are detected by a photodiode. The detection is done in a homodyne scheme, using a Vector Network Analyzer (VNA). Fig. 1(a) shows a simplified schematic setup. The drum is actuated by subjecting it to both dc and ac voltages while the excitation frequency is varied around the fundamental frequency in small steps. To relate the measured amplitude (in V/V) to the actual motion of the membrane, a calibration measurement of the drum's Brownian motion is performed [6].

Non-linear identification

In order to perform the identification procedure, first the geometrically non-linear response of the fixed graphene membrane is modelled by using a Lagrangian approach. The numerical analysis is performed by assuming axisymmetric vibrations and expanding the radial displacement of the membrane in terms of admissible functions satisfying fixed boundary conditions. Then, by neglecting radial inertia, Lagrange equations are reduced to a single dimensionless Duffing oscillator with viscous damping as follows:

$$r^2\ddot{x}+\zeta\dot{x}+x+\eta_3 x^3=\lambda\cos(t)\,,\quad \lambda=\xi\mathrm{F}\big/\left(m\omega_1^2 h\right), \tag{1}$$

where x is the generalized coordinate associated with the fundamental mode of the membrane. Moreover, x and t are made dimensionless with respect to the drum's thickness (h) and the excitation frequency, respectively. In addition, r is the frequency ratio (the ratio between the excitation frequency and the fundamental frequency (ω_1)), ξ is the modal participation factor, F is the driving force, and m is the mass of the membrane. The unknown parameters are the damping ratio ζ, the dimensionless force λ, and η_3. The non-linear stiffness η_3 is function of the Young's modulus and its convergence and accuracy is determined by using different number of terms in the expansion for the radial displacement of the membrane. For a graphene membrane with Poisson ratio 0.16, η_3 converges to:

$$\eta_3=0.58Eh^3\big/\left(R^2N_0\right)\;,\quad N_0=R^2\rho h\omega_1^2\big/(5.78)\;, \tag{2}$$

where R is the radius of the drum, E is the Young's modulus, ρ is the mass density, and N_0 is the pretension that is determined from the experimentally measured fundamental frequency. Next, harmonic balance method is applied and the solution of equation (1) is approximated by a truncated Fourier series:

$$x \approx x_N = x_0 + \sum_{k=1}^{N}\left[x_{2k-1}\sin kt + x_{2k}\cos kt\right], \tag{3}$$

where N is the chosen order of truncation and x_N is the truncated Fourier series representation of x. By substituting equation (3) into (1) and equating the coefficients of each of the N harmonics, a system of algebraic equations is obtained that relates the frequency ratio r to the amplitudes x_N. Next, the identification is conducted by assuming that the vibration amplitude x_N, and the frequency ratio r are already known for every frequency step from experiments. Therefore, in order to obtain the unknown parameters, the following system is solved for every j-th frequency step, $r^{(j)}$:

$$\left[2r^{(j)}\mathrm{D}\mathrm{S}_{x^{(j)}} \quad \mathrm{Q}_{x^{(j)}} \quad -\mathrm{S}_{f^{(j)}}\right]\cdot\begin{Bmatrix}\zeta\\ \eta_3\\ \lambda\end{Bmatrix} = \left[-\mathrm{S}_{x^{(j)}} - r^{(j)}\mathrm{D}^2\mathrm{S}_{x^{(j)}}\right], \quad \mathrm{D}=\begin{pmatrix}0 & 0 & \cdots & 0\\ 0 & \mathrm{D}_1 & \cdots & 0\\ \vdots & \vdots & \ddots & \vdots\\ 0 & 0 & \cdots & \mathrm{D}_N\end{pmatrix}, \ \mathrm{D}_k = \begin{pmatrix}0 & -k\\ k & 0\end{pmatrix}, k = 1,\dots,N \ , \ j = [\![1:m]\!]. \tag{4}$$

where m is the number of excitation frequency steps for which the experimental data was obtained at a specific excitation level. S_x, and Q_x are vectors comprising truncated Fourier coefficients of x, and x^3, respectively, and S_f is the dimensionless force vector. System (4) is over-constrained, since it contains $(2N+1)\times m$ equations. Therefore, in order to obtain unknown parameters the least squares technique has been applied [7].

Results

The nanodrum of interest has a thickness of 5nm as confirmed by Raman spectroscopy and atomic force microscopy. The membrane is driven electrostatically using the silicon wafer as a backgate. Fig. 1(b) shows a set of calibrated frequency response traces of the fundamental mode while varying the driving ac voltage. The dc voltage has been kept fixed (V_{dc} = 3V) throughout the entire measurement. The frequency of the fundamental mode is 14.7MHz. The RMS force is the electrostatic driving force corrected by the modal participation factor of the drum's fundamental mode. For driving forces above F_{RMS} = 15pN, the resonance peak exhibits a non-linear hardening behaviour, which holds information about the Young's modulus. These set of curves are then fitted by equation (1) and the cubic non-linear term is obtained to extract the Young's modulus. Figs. 1(c) and (d) compare identified curves and experimental ones for different F_{RMS}. The fitted curves are in full agreement with the experimental results, giving a Young's modulus of 0.6 TPa.

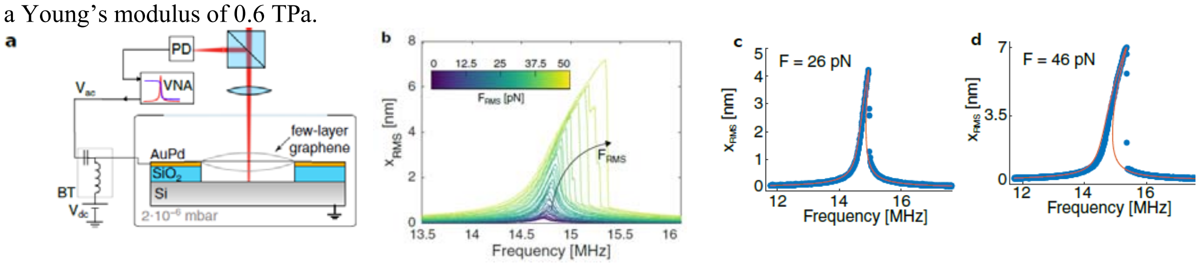

Fig. 1. (a) The schematic experimental set up; (b) experimentally measured non-linear amplitude-frequency response curves; (c) comparison between the experimental (dots) and identified curve (red line) for F=26pN; (d) comparison between the experimental (dots) and identified curve (red line) for F=46pN.

Conclusions

A non-contact measurement procedure has been developed to obtain non-linear frequency response curves of strongly driven graphene nanodrums. In order to estimate Young's modulus, a non-linear identification technique based on harmonic balance method was presented and the experimentally obtained frequency-amplitude curves were fitted by a forced Duffing oscillator. The identified value of Young's modulus is within the range of elastic modulus previously reported in the literature via AFM nanoindentation. The new method allows high frequency determination of the Young's modulus at small amplitudes without mechanical contact.

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