Abstract

Starting from an atomistic approach we have derived a hierarchy of successively more simplified continuum elasticity descriptions for modeling the mechanical properties of suspended graphene sheets. The descriptions are validated by applying them to square graphene-based resonators with clamped edges and studying numerically their mechanical responses. Both static and dynamic responses are treated. We find that already for deflections of the order of 0.5 Å a theory that correctly accounts for nonlinearities is necessary and that for many purposes a set of coupled Duffing-type equations may be used to accurately describe the dynamics of graphene membranes.

Introduction. Recent progresses in fabricating graphene, a monolayer of graphite, have considerably boosted the attention for this material [1]. Among its unique features are remarkable electronic properties [2, 3], which make graphene of considerable interest for both fundamental science and technological applications. Moreover, its exceptionally large mechanical strength [4] and ability to sustain large electrical currents can be of great value in the broad field of nanodevices. In particular, in the field of nanoelectromechanical systems (NEMS), graphene-based mechanical resonators were recently demonstrated [4, 5] and theoretical work indicates a strong coupling between deformation and intrinsic electronic properties of graphene [6, 7].

Most research on graphene has hitherto focused on the electronic properties of graphene, and less attention has been directed to mechanical properties. For modeling NEMS a reliable and efficient description of the mechanical response of nanocarbons to external forces is essential [8, 9]. While continuum elasticity theory has been applied successfully to the study of mechanical properties of nanotubes for a long time, [10] [11] [12] it has only recently been applied to graphene membranes [13] [14] [15] [16].

In this work we first formulate a general nonlinear elasticity theory for graphene sheets starting from an atomistic description in terms of a valence force field model. Through successive approximations, we then derive simplified models which are easier to solve and study analytically. We then apply these continuum descriptions to a drum-like resonator and investigate the mechanical response in both the static and dynamic cases. The results obtained agrees well with the experimentally observed responses [17, 18].

Continuum elasticity model for graphene.

The interaction potential between carbon atoms in graphene can be modeled by a valence force field model [19] [20] where the potential energy \( U_{sp^2} \) between \( sp^2 \) bonds is given by

\[
U_{sp^2} = \frac{1}{2} \sum_{i=1}^{N_{at}} \sum_{j=1}^{3} \frac{\alpha}{4a_0^2} (\bar{r}_{ij}^2 - a_0^2)^2 + \sum_{i=1}^{N_{at}} \sum_{j<k} \frac{\beta}{a_0^2} (\bar{r}_{ij} \cdot \bar{r}_{ik} + \frac{1}{2}a_0^2)^2 + \sum_{i=1}^{N_{at}} \gamma \bar{D}_i \cdot \bar{D}_i.
\]

Here the index \( i \) labels the carbon atoms while indices \( j \) and \( k \) are bond labels for the nearest neighbor atoms of \( i \). Thus, \( \bar{r}_{ij}, j = 1, 2, 3 \), are the three...
bond vectors that connect the atom $i$ to its nearest neighbors. The parameters $\alpha$, $\beta$ and $\gamma$ are constants, $N_{at}$ the number of atoms, $a_0 = 1.421$ Å is the equilibrium bond length in graphite, and $D_i = \sum_{j=1}^{3} \vec{r}_{ij}$ is the dangling bond vector. The first two terms of Eq. (11) represent the energy cost necessary to change the length and angle between covalent C-C bonds. In the continuum description, we refer to it as the stretching energy. The last term of Eq. (11) gives the energy cost necessary to change the angle between $p_z$-orbitals, which are approximately normal to the graphene surface. We refer to energy contributions from this term as bending energy.

Provided that the length scale of the deformation is large compared to the lattice spacing (long wavelength limit), continuum theory can be used for graphene [15] and we can write

$$U_{sp}^2 = \int dx dy W_0[\vec{u}(x,y)]$$

by parametrizing the deformed surface as $\vec{u}(x,y) = [u(x,y), v(x,y), w(x,y)]$. The elastic energy density $W_0$ can be divided into stretching and bending contributions, $W_0 = W_0^S + W_0^B$. The stretching energy density can be written as [21]

$$W_0^S = \frac{Eh}{2(1-\nu^2)}[E_{xx}^2 + E_{yy}^2 + 2\nu E_{xx}E_{yy} + 2(1-\nu)E_{xy}^2],$$

(2)

where the components of the Green strain tensor $E_{ij}$ are

$$E_{xx} = u_x + (u_x^2 + v_x^2 + w_x^2)/2,$$
$$E_{xy} = (u_y + v_x + u_x u_y + v_x v_y + w_x w_y)/2,$$
$$E_{yy} = v_y + (u_y^2 + v_y^2 + w_y^2)/2.$$  

Here the subscripts on $u,v,w$ denote differentiation, i.e. $u_x = \partial u/\partial x$ etc., and the coefficient $Eh$ represents the Young modulus multiplied by the thickness of thin plate theory [22] [23]. It is important to note that in our theory $Eh$ is a single parameter and we do not consider any thickness in our formulation. The Poisson ratio is denoted by $\nu$. Both $Eh$ and $\nu$ are related to the Lamé parameters $\lambda$ and $\mu$ as $Eh = 4\mu(\lambda + \mu)/(\lambda + 2\mu)$ and $\nu = \lambda/(\lambda + 2\mu)$. The Lamé parameters are determined from $\alpha$ and $\beta$ as $6\mu = \sqrt{3}(\alpha + 8\beta)$ and $6\lambda = \sqrt{3}(\alpha - 4\beta)$ (for details, see [21]). Using values from Ref. [19] ($\alpha = 155.9$ J/m$^2$ and $\beta = 25.5$ J/m$^2$), gives $\mu = 103.89$ J/m$^2$ and $\lambda = 15.55$ J/m$^2$. The continuum theory is in an excellent agreement with molecular dynamics simulations of graphene [15].

The bending energy density can be approximated as

$$W_0^B = \frac{\kappa}{2}(2a_0H)^2,$$

where $\kappa = \sqrt{3}a_0^2\gamma/2 \approx 0.8$ eV is the bending rigidity and $H = (w_{xx} + w_{yy})/2$ is the local mean curvature.

In clamped graphene-based NEMS applications (e.g.: mechanical resonators), we find that the stretching energy is much greater than the bending energy. In thin plate theory, the bending regime (linear theory) is valid only for out-of-plane deflections less than the plate thickness [23]. Following this argument, for graphene the linear regime is almost nonexistent and the nonlinear stretching regime is dominant. We have determined this to be the case for deflections in excess of 0.5 Å. Thus, in the following we neglect the bending contribution.

For a given applied external body force $\vec{F}_0$, the equilibrium shape can be obtained by direct minimization of the free energy functional

$$F[\vec{u}(x,y)] = \int dx dy W_0 - \int dx dy \rho_0 m_c^{-1} \vec{F}_0 \cdot \vec{u},$$

where $\rho_0$ is the mass density and $m_c$ is the carbon mass. Alternatively, the equilibrium shape may be found by solving for the dynamics of the system including dissipation. We will here work with the latter approach. The dynamic equation in the stretching regime, where Eq. (2) is used as the elastic energy density, can be obtained from standard variational principles. The corresponding equation of motion, with an added phenomenological damping term $c\dot{u}(x,y)$, is

$$\ddot{u}(x,y) + c\dot{u}(x,y) = \rho_0^{-1} \dot{\vec{P}}[\vec{u}(x,y)] + m_c^{-1} \vec{F}_0(x,y,t),$$

(3)

where $\dot{\vec{P}}$ is the Piola stress tensor. In cartesian coor-
estimates \((xy)\), the Piola stress tensor is
\[
P_{xx} = (1 + u_x)[(\lambda + 2\mu)E_{xx} + \lambda E_{yy}] + 2\mu u_y E_{xy},
P_{xy} = 2\mu(1 + v_y)E_{xy} + v_x[(\lambda + 2\mu)E_{xx} + \lambda E_{yy}],
P_{xz} = \lambda w_x(E_{xx} + E_{yy}) + 2\mu w_x E_{xx} + 2\mu w_y E_{xy},
P_{yz} = 2\mu(1 + u_x)E_{xy} + u_y[(\lambda + 2\mu)E_{yy} + \lambda E_{xx}],
P_{yy} = (1 + v_y)[(\lambda + 2\mu)E_{yy} + \lambda E_{xx}] + 2\mu v_x E_{xy},
P_{yz} = \lambda w_y(E_{xx} + E_{yy}) + 2\mu w_y E_{yy} + 2\mu w_x E_{xy}(d)
\]
and the linear differential operator \(D\) acts on \(\hat{P}\) as
\[
D\hat{P} = \sum_{\chi=x,y,z} (\partial_{xx}P_{\chi} + \partial_{y}P_{\chi})\hat{x}.
\]
Dirichlet boundary conditions can be imposed through \(\bar{u} = \bar{u}_0(t)\) and Neuman boundary conditions through \(\bar{n}_0 \cdot \hat{P} = \bar{P}_0(t)\), where \(\bar{u}_0(t)\) and \(\bar{P}_0(t)\) are specified on the boundary.

We refer to Eqs. (3) and (4) as the full nonlinear system or general elasticity equations. Since the general expressions for the Piola stress tensor are quite cumbersome and make the equations difficult to solve. The full equations can be approximated by a simpler set of equations where we neglect the second order contributions of the in-plane displacements \(u\) and \(v\). This yields the following expressions for the components of the strain tensor
\[
E_{xx} \approx u_x + w_x^2/2,
E_{xy} \approx (u_y + v_x + w_x w_y)/2,
E_{yy} \approx v_y + w_y^2/2,
\]
and the Piola tensor
\[
P_{xx} \approx (\lambda + 2\mu)E_{xx} + \lambda E_{yy},
P_{xy} \approx 2\mu E_{xy},
P_{xz} \approx \lambda w_x(E_{xx} + E_{yy}) + 2\mu(w_x E_{xx} + w_y E_{xy}),
P_{yz} \approx 2\mu(1 + u_x)E_{xy} + u_y[(\lambda + 2\mu)E_{yy} + \lambda E_{xx}],
P_{yy} \approx (1 + v_y)[(\lambda + 2\mu)E_{yy} + \lambda E_{xx}] + 2\mu v_x E_{xy},
P_{yz} \approx \lambda w_y(E_{xx} + E_{yy}) + 2\mu w_y E_{yy} + 2\mu w_x E_{xy}(d)
\]
we have arrived at these equations without treating the graphene as a thin plate with some thickness.

We may further simplify the above expressions by completely removing the in-plane displacements. The resulting nonlinear equation for \(w(x, y, t)\) is given by
\[
\ddot{w}(x, y, t) + c\dot{w}(x, y, t) - \rho_0^{-1} \sum_{\chi=x,y} \partial_{\chi}(w_{\chi}T_{\chi}) = \frac{F_0}{m_c},
\]
where
\[
T_x = (\lambda + 2\mu)\delta x + \lambda \delta y + (\lambda/2 + \mu)(w_x^2 + w_y^2),
T_y = (\lambda + 2\mu)\delta y + \lambda \delta x + (\lambda/2 + \mu)(w_x^2 + w_y^2).
\]
Here we have introduced the constants \(\delta x\) and \(\delta y\) representing initial strains in the \(x\) and \(y\) directions, respectively. Such strains may appear during the manufacturing process of graphene resonators. The functions \(T_x\) and \(T_y\) are tensions in the \(x\) and \(y\) directions induced by stretching of the graphene. We refer to Eq. (7) as the out-of-plane approximation.

If one mode is expected to dominate the out-of-plane deformations, the out-of-plane approximation may be projected onto this mode to obtain an ordinary rather than a partial differential equation. If the dominant mode is the fundamental, we can write for a square sheet \((-a \leq x, y \leq a)\)
\[
\frac{w(x, y, t)}{\rho_0} = \frac{w(t)\cos(\pi x/2a)\cos(\pi y/2a)}{\rho_0}.
\]
By using this Ansatz, we obtain a Duffing equation for the amplitude of the fundamental mode,
\[
\ddot{w}(t) + \omega_0^2 w(t) + \frac{5\pi^4(\lambda + 2\mu)}{128a^4\rho_0} w^3(t) = \frac{F(t)}{m_c},
\]
where
\[
F(t) = \frac{1}{a^2} \int \int F(x, y, t)\cos(\pi x/2a)\cos(\pi y/2a)dxdy
\]
is the overlap of the driving force with the shape of the fundamental mode, and the resonant frequency \(\omega_0\) is given by
\[
\omega_0 = \frac{\pi}{a} \sqrt{\frac{(\lambda + \mu)(\delta + \delta^2/2)}{\rho_0}}.
\]
and we have assumed $\delta_x = \delta_y = \delta$. Notice if several modes are included in the Ansatz, we obtain a set of coupled Duffing-type equations for the mode amplitudes.

**Numerical results: Statics.** We now proceed to investigate the accuracies of the different approximations — full nonlinear system (44), von Kármán approximation (50), out-of-plane approximation (7), and the Duffing equation (8). For illustration, we consider in the simulations a clamped square graphene sheet of side $2a = 1 \mu m$, (Dirichlet boundary condition with $u_0(t) = 0$ on all edges) subject to uniform pressure $F_{dc} \overline{z}/A$ where $A = 3\sqrt{3}a^2/4$ is the area per carbon atom in graphene. In all computations we consider a pre-tensile graphene with initial strains of $\delta_x = \delta_y = 0.5\%$. We solve the Eq. 6 numerically by using the spectral method [24, 25] with 81 global basis functions.

The tension in the graphene sheet varies depending on how much stretching is imposed by the external forces. For pre-tensile graphene, small deflections do not change the built-in tension and the graphene-based resonator behaves as a linear tensile elastic membrane [26]. However, when the deflections are large enough, the stress changes significantly from the initial value. To illustrate this, we present Figs. 1 and 2. The first figure shows the stress distributions for two values of driving forces $F_{dc}$ per carbon atom, $F_{dc} = 1 \text{nN}$ and $F_{dc} = 1 \text{pN}$. For the smaller force, the deflection at equilibrium is small and the tension $P_{xx}$ is almost uniform and equal to the initial value $1.2 \text{N/m}$. This agrees with the theoretical value of $\sigma = Eh/(1-\nu)\delta \approx 1.2 \text{N/m}$, where $\delta = 0.5\%$ is the initial strain of the pre-tensile graphene. On the other hand, for the larger force $F_{dc} = 1 \text{pN}$, the equilibrium built-in tension $P_{xx}$, as well as the other components of the Piola stress tensor, is not uniform and it varies from $2 \text{N/m}$ to $14 \text{N/m}$. The inset in Fig. 2 shows the dependence between the average value of the tension $P_{xx}$ and the driving force $F_{dc}$. Note that for small $F_{dc}$ the tension is almost constant and equal to $1.2 \text{N/m}$, but for large $F_{dc}$ the tension varies as $F_{dc}^{2/3}$.

Next, we present the results for the vertical deflection at the center of the graphene sheet as function of the external force $F_{dc}$. We find that for a pre-tensile graphene sheet the response is linear for small values of $F_{dc}$. This is due to an almost uniform tension, independent of the amount of deflection, see Fig. 2. On the other hand, when the out-of-plane deflections are large, we see a nonlinear response $w \sim F_{dc}^{1/3}$, where $w$ is the deflection at the center of the graphene.

Finally, evaluations of the bending energy for the equilibrium shapes, for a range of $F_{dc}$ between $0.1 \text{fN}$ and $1 \text{pN}$, show that the stretching energy is at least four orders of magnitude larger than the bending energy. This validates the assumption that bending energy in clamped graphene-based nanoresonators may be neglected. In addition, we find that all the simplified mechanical descriptions show good agreement with the general theory in the static case.

**Numerical results: Dynamics.** In this section, we study the dynamic response of pre-tensile clamped graphene subject to small and large deformations. For small deformations, when the tension is set by the initial strain, the clamped graphene sheet behaves as a linear tensile elastic membrane [26]. On the other hand, when the out-of-plane deformations are large and tension depends on the amount of deflection, a
Figure 2: Elastostatics study for clamped pre-tensile graphene. The figure shows the vertical deflection at the center of the graphene sheet in response to a uniform force ranging from 0.1 fN to 1 pN per atom. Good agreement is obtained between the simplified models and the full nonlinear system. The inset shows the dependence between the tension on the driving force.

nonlinear response governs the dynamic behaviour. Here we show that the dynamics exhibits Duffing-type behavior, e.g. we observe the characteristic instability in the amplitude when we sweep the driving frequency.

We divide the analysis into three parts. First, we consider a driving force with the same spatial distribution as the fundamental mode. Here we have only one mode present which makes this case easy to analyse. In the second part we consider a uniform force where several modes couple to the external force. Here we observe the presence of up to three modes with similar Duffing-type response as the fundamental mode. Finally, we study small oscillations around an equilibrium determined by the dc-component of the exciting forces. We evaluate the resonance frequencies as function of the $F_{dc}$ and the amplitude of oscillation as a function of the ac-component of the driving force $F_{ac}$.

For a driving force with the same spatial dependence as the fundamental mode,

$$F(x, y, t) = F(t) \cos(\frac{\pi x}{2a}) \cos(\frac{\pi y}{2a}),$$

the out-of-plane and Duffing approximations are equivalent. We perform numerical computations for small and large harmonic driving forces ($F(t) = F_{ac} \cos(\omega t)$). The results are shown in Fig. 3. The figure shows that for small driving force $F_{ac} = 0.5$ fN, the response is that of a linear harmonic resonator. However, for the larger driving forces of $F_{ac} = 6$ fN and 20 fN, we see a nonlinear response which manifests as an amplitude instability at a critical, amplitude dependent frequency; e.g., at $f = 1100$ MHz for $F_{ac} = 6$ fN. The Duffing equation predicts this instability at a slightly higher frequency. This is because the tension is slightly reduced when the in-plane displacements are included (general elasticity theory or von Karman approximation), see Fig. 2. There is an excellent agreement between the general elasticity theory and the von Karman approximation.

Next, we study the case of a uniform driving force. Here several modes couple to the driving forces. The fundamental mode has the greatest overlap followed by the modes

$$F_{dc} = 0.5 \text{ fN, Duffing equation}$$

$$F_{ac} = 5 \text{ fN, General elasticity theory}$$

$$F_{ac} = 6 \text{ fN, von Karman theory}$$

$$F_{ac} = 6 \text{ fN, Duffing equation}$$

$$F_{dc} = 20 \text{ fN, General elasticity theory}$$

$$F_{dc} = 20 \text{ fN, von Karman theory}$$

$$F_{dc} = 20 \text{ fN, Duffing equation}$$

Figure 3: Amplitude of the fundamental mode for different driving forces strength. Observe the amplitude instability for large values of $F_{ac}$. All points were obtained starting from a configuration with the sheet at rest. If the dynamic state of the system is kept between frequency steps a hysteretic response obtains.
These modes are found by plotting the stretching energy against the driving frequency. This is shown in Fig. 4. The figure shows three peaks corresponding to the different modes: the peak at 1200 MHz belongs to the fundamental mode, and the peaks at 2360 MHz and 2840 MHz correspond to the other modes mentioned above. Notice that the first peak at 350 MHz does not correspond to any mode but to a 1/3 sub-harmonic of the fundamental. The presence of this sub-harmonic is due to the cubic nonlinear term in Eq. (8).

Finally, we study small oscillations around an equilibrium shape determined by a dc-part of the driving force $F_{dc} + F_{ac} \cos(\omega t)$, typical for a NEMS resonator. We assume a spatially uniform driving force. We find that the resonance frequency for small oscillations increases as we increase the stretching or deflection at the equilibrium position. This may be used to tune the frequency of the oscillator by changing the dc-bias as $f_{res} \propto F_{dc}^{1/3}$.

Next, we study the dependence between the amplitude of the oscillations as function of the driving ac-force $F_{ac}$. We find that there is a linear response for small $F_{ac}$ and then it tends to follow the law $F_{ac}^{1/3}$. This is shown in Fig. 5. Here the driving frequency is the resonance frequency for $F_{dc} = 250$ fN, which is 3360 MHz.

**Conclusions.** We have derived and analyzed a nonlinear finite elasticity theory for graphene resonators, both for elastostatics and elastodynamics problems. Moreover, we have studied how this general elasticity theory can be simplified to more easily solvable equations. In particular, the out-of-plane approximation, Eq. (7), gives good agreement with the general elasticity theory while maintaining the advantage of being computationally efficient. We have also shown that the dynamic response of clamped graphene resembles that of coupled Duffing-type resonators.

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