

Insights into Relative Lower Frequencies and Buckling Loads of Monolayer Graphene Sheets via Nonlocal Elasticity Theory: Size-Dependent Young's Modulus Approach

T. Murmu^{1,*}, S. Adhikari², M. A. McCarthy¹, and C. Y. Wang²

¹*Department of Mechanical, Aeronautical and Biomedical Engineering, Irish Centre for Composites Research, Materials and Surface Science Institute, University of Limerick, IRL, Ireland*

²*College of Engineering, Swansea University, Singleton Park, Swansea SA2 8PP, Wales, UK*

The nonlocal natural frequencies and buckling loads of a graphene sheet decreases as the scale (nonlocal) parameter increases. The reasons for these behaviours of graphene sheet have not been exhaustively addressed earlier. We discuss these important predictions of graphene when modelled via nonlocal elasticity. This letter answers theoretically the cause behind these values of nonlocal frequencies and buckling loads being different (lower) from classical theories. The concept of size-dependent Young's modulus and the pseudo-in plane loads are introduced, which bridges the low effective structural stiffness (or frequency/buckling load) to its physical origin, i.e., nonlocal interaction among atoms.

Keywords: Graphene, Nonlocal Elasticity, Classical Elasticity.

Graphene¹ promises as one of the exciting material of the future. It is a two-dimensional structure consisting of an one-atom-thick planar sheet of sp^2 -bonded carbon atoms organised in a honeycomb crystal lattice.² The graphene sheets can be mono-layered or multi-layered. Knowledge of the behavior of graphene under mechanical loading is essential for its optimal design and applications. The mechanical behaviour includes its bending, vibration and buckling characteristics etc. The graphene sheets can be potentially used as a resonators³ and mass sensors.^{4,5} As experiments are not always practical at the nanoscale, molecular dynamics,^{6,7} (MD) is often used for understanding the graphene sheets. Molecular dynamics simulations can yield accurate and highly detailed information concerning mechanical response of nanostructures. However, from an engineer's perspective, expertise in molecular dynamics is not common among engineers. Further MD is very time-consuming and computationally expensive if the number of atoms is large. So finding techniques for exploring the behaviour of graphene sheets for bending, vibration, buckling and other studies are important issues. When the mechanical behavior of graphene

sheet is understood, it can be used as resonators³ and mass sensors.^{4,5} Continuum methods are becoming popular for design of nano-scale devices. Continuum mechanics theories play an indispensable role in characterising overall mechanical response of nanoscale structures. The mechanics is the building blocks of engineering nanostructures. However theories at a nanoscale level should consider the distinct features that distinguish nanomaterials/structures from their macroscopic counterparts. One distinct feature is the size-effects at nanoscale⁸⁻¹⁴ related to atoms and molecules.

One popular size-dependent continuum theory, frequently used to model bending, vibrational and instability behaviour of graphene sheets is Eringen's nonlocal elasticity theory.¹⁵ Local elasticity is based on the concept of localness (point) irrespective of the surrounding, while nonlocal elasticity takes into account the effect of the surroundings. This effect is more prominent and intuitive at atomic scale (nanoscale) where effect on an atom is affected by other surrounding atoms. Nonlocal elasticity theory (NET) is based on atomic theory of lattice dynamics. The theory states that the stress components at a point depend not only on the strain components at the same position but also on all other points of the body. The theory is

*Author to whom correspondence should be addressed.

found to be in accordance with lattice dynamics and experimental observation on phonon dispersion. NET for structural analysis of graphene is developed from a nonlocal constitutive differential form due to simplicity. Using the nonlocal constitutive differential form, researchers have developed nonlocal plate theories based on Kirchhoff's plate theory, Mindlin plate theory, and higher order plate theories.¹⁶ It is important to note that monolayer graphene is assumed to be an idealised nonlocal plate with constant effective thickness (actually the thickness is not well defined).¹⁷ From molecular dynamics it is also shown¹⁸ for wave propagation, that a graphene monolayer if considered as continuum thin plate would have a thickness of $h = 0.104$ nm. Idealised plate means that the surface is flat without any presence of ripples.¹⁹

The NETs are based on a single important parameter known as nonlocal parameter or scale coefficient.^{15,20} When this parameter is considered zero, the model is the same as the classical elasticity, and this implies no size-effect. The size-effects term is very broad and is related to atomic, molecular and lattice effects. In fact it is the extrinsic length of the given structure which is important. Extrinsic length may be sample size, wavelength or crack length. We can analysis a macroscopic plate by nonlocal elastic plate theory as the macroscopic plate also contains atoms and lattices. However the intrinsic length (e.g., lattice parameter) will be so small compared to extrinsic length (e.g., sample size), that it is very reasonable to neglect it.

A number of studies have been conducted using the nonlocal plate theory for a graphene (idealised plate) structure.¹⁶ It should be noticed that the local elasticity is not able to impart accurate prediction of graphene behaviour when compared to molecular dynamics. The local theory results are not accurate as we ignore the important size effects at the nanoscale. From nonlocal plate theories, studies on bending, vibration and buckling analysis of graphene (also in carbon nanotubes²¹) have predicted larger deflection and smaller frequencies and buckling loads in comparison to classical plate theories. There are two important broad areas of nonlocal elasticity plate theories which require attention i.e., (i) the closeness of the nonlocal structural predictions compared to realistic behaviour and (ii) the correct (universal) values of nonlocal parameter for graphene if there is one. Researchers have attempted to solve the second question but the first question has not been addressed in depth as it is assumed that graphene would have larger deflection, smaller frequencies and buckling loads. Experimental evidence is thus necessary in this investigation. The motivation of the present work is to understand why the nonlocal plate theory predicts this way and its application to graphene sheets. This letter attempts to quantitatively explore the rationale behind the decreased frequencies and buckling loads of a graphene sheet via NET.

Assume an idealised rectangular flat single-layer graphene sheet (SLGS) having dimensions ($a \times b$) in x and y direction, respectively (Fig. 1). It undergoes deflection w with time t . The effective thickness of the SLGS, h is assumed constant. Using NET, the governing equation for free vibration of a rectangular monolayer graphene sheet at room temperature is given as²²

$$D\nabla^2 \cdot \nabla^2 w + \rho h [1 - (e_0 l_c)^2 \nabla^2] \frac{\partial^2 w}{\partial t^2} = 0 \quad (1)$$

where e_0 is the nonlocal parameter. Term l_c is the internal characteristic length (e.g., distance between C–C atoms, lattice parameter etc.). Here $D = (Eh^3)/(12(1 - \nu^2))$, and ρ is the density. The term ∇^2 is the Laplacian operator $(\partial)/(\partial x^2) + (\partial)/(\partial y^2)$. E and ν denote the Young's modulus and the Poisson's ratio of the SLGS. It should be noted that the SLGS here is considered isotropic in nature. For nonlocal simply-supported graphene plate, we assume the solution of Eq. (1) as

$$w = \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} W_{mn} \sin(m\pi/a)x \sin(n\pi/b)y e^{i(\omega_{mn}t)} \quad (2)$$

With aspect ratio, $R = (a/b)$, the natural frequencies are expressed as

$$\omega = \frac{1}{a^2} \sqrt{\frac{D}{\rho h}} \sqrt{\frac{[(m\pi)^2 + R^2(n\pi)^2]^2}{1 + \mu^2[(m\pi)^2 + R^2(n\pi)^2]}} \quad (3)$$

The term μ in the frequency expression is the nonlocal parameter (dimensionless) and is expressed as $\mu = e_0(l_c/a) = e_0 l_R$, where e_0 is constant appropriate to each material. The prediction of the natural frequencies of graphene has been validated by researchers using molecular dynamics (MD) simulation.²³ For a macroscopic (say a thin graphite plate) plate, the expression for the frequencies is, $\omega = \frac{1}{a^2} \sqrt{\frac{D}{\rho h}} [(m\pi)^2 + R^2(n\pi)^2]$. The expression is the cause of the effect that $\mu = 0$. In fact μ is not exactly zero but $\mu \rightarrow 0$, ($l_R \rightarrow 0$) and is very small and is neglected in classical elasticity theory. Details on μ can be found in Ref. [15]. Similarly, the governing equation of buckling of rectangular graphene sheet is given as

$$D\nabla^4 w - P[1 - (e_0 l_c)^2 \nabla^2] \frac{\partial^2 w}{\partial x^2} = 0 \quad (4)$$

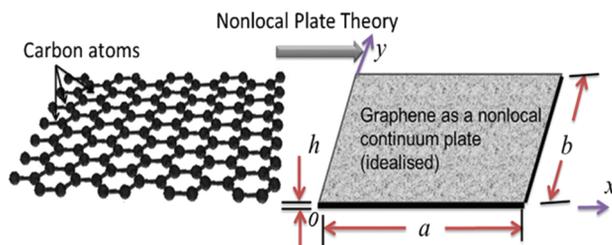


Fig. 1. Single layer graphene sheet considered as a continuum nonlocal thin plate.

The buckling loads for simply-supported monolayer graphene plate can be obtained as

$$P = \frac{D}{a^2} \frac{[(m\pi)^2 + R^2(n\pi)^2]^2}{(m\pi)^2[1 + \mu^2[(m\pi)^2 + R^2(n\pi)^2]]} \quad (5)$$

The nonlocal expressions for frequencies and buckling loads imply that the nonlocal frequencies and buckling loads are smaller than the local counterpart ($\mu = 0$) i.e., there is always a reduction in the values of parameters when nonlocal effects are considered $\mu > 0$. It is important to note, that values of elastic properties of graphene sheets are quite variable in literature.^{24,25} Details of various Young's modulus values for graphene sheets are discussed in Reddy et al.²⁶ Say, we consider the properties of graphene sheets as Young's modulus, $E = 3.4$ TPa,²⁵ density $\rho = 2250$ kg/m³ and Poisson's ratio as $\nu = 0.16$.²⁷ Thickness is taken as 0.1 nm.²⁵ We would notice that the reductions in frequencies and buckling loads increase as the nonlocal parameter increases. The values of critical buckling load are more affected than first natural frequencies. The effects are even more prominent in higher modes. As the length of the SLGS increases the nonlocal effect diminishes.

These observations have been also reported in earlier papers.¹⁶ However the defence of the results obtained from nonlocal elasticity analysis is not clearly and comprehensively reported in literature. The underlying physics and the insights have been generally missing. If the physics is understood, and related experiments can be performed, then the gap between mathematical theory and real applications can be bridged. In this letter experiments are beyond the scope of the present work, so attempt has been made to understand the rationale behind the nonlocal elastic analysis for graphene sheets.

We first look at the nonlocal elasticity constitutive relation as proposed by Eringen.¹⁵ The differential form of the nonlocal constitutive relation is¹⁵

$$[1 - (e_0a)^2 \nabla^2] \sigma_{NL} = E \varepsilon \quad (6)$$

From NET the stress at a point in a body not only depends on the strain at that point but also on all strains in the body. The expression in Eq. (6) imparts Young's modulus as

$$E_{NL} = \frac{[1 - (e_0a)^2 \nabla^2] \sigma_{NL}}{\varepsilon}$$

For a nanostructure, if the stress at point (atom) behaves as local elastic stress then we have $\sigma_{NL} = \sigma_L = E \varepsilon$, and the size-independent Young's modulus is obtained from $E_L = (\sigma_L)/(\varepsilon)$. Considering the term containing nonlocal effects is positive, we get E_L (size-independent) $>$ E_{NL} (size-dependent), it happens that the bending rigidities (stiffness) as D_L (size-independent) $>$ D_{NL} (size-dependent). This implies that we use a constant size-independent (local) Young's modulus in governing equations considering the cross section is defined. The reason for decreased

frequencies and buckling load is thus the consideration of size-independent over-predicted constant Young's modulus. In Figure 2 we have emphasized on nonlocal vibration of SLGS which shows the change of absolute frequencies with nonlocal parameter μ . The frequency reduces as μ increases. Applying classical elasticity, for the same drop of values of frequencies the bending stiffness (or Young's modulus) has to decrease as illustrated in Figure 2(b). These are true for buckling loads too. These trends are justified by the observations from experiments and molecular dynamics as we discussed below.

Literature shows that experiments and molecular dynamic simulations have been carried out to measure and determine the elastic moduli of graphene sheets. One of the methods by which the Young's modulus of graphene can be determined is by examining thermal vibrations via molecular dynamics simulations. Jiang et al.²⁸ first used this method to study the Young's modulus of graphene. The reliability of their results are illustrated by comparing their work with that of Lee et al.²⁹ According to Jiang et al.,²⁸ at small scales, Young's modulus is not constant and size-independent; and is in fact nonlinearly proportional to length of the graphene sheet. Thickness was considered as 3.35Å. The Young's modulus decreases as the length of square graphene decrease. This trend is however beyond a threshold length. Above the threshold length the Young's modulus is constant. Considering certain conditions, Jiang et al.²⁸ reported the threshold Young's modulus is 1.1 TPa.

Using the molecular dynamics method with the orthogonal tight binding method, Zhao et al.⁸ determined the magnitude of Young's modulus. Similar thickness was considered as 3.35Å. Their results match well with the experimental one. It was shown that at small scales Young's modulus is dependent on the size and chirality (arm chair and zigzag) of graphene. After extensive study it is reported that the Young's modulus is nonlinearly proportional to the size of graphene. The Young's modulus increases with increase of the diagonal length of the nanoribbon and slowly converges to the Young's modulus of bulk graphene. In other words it is concluded that stiffness decreases when the size of graphene nanoribbon decreases. The size-dependent effect is negligible when the diagonal length of the graphene nanoribbon is over 10.0 nm. Considering the present parameters, this critical length of graphene can be important in nonlocal plate theory. In vibration analysis of graphene, in some studies it is shown that the nonlocal effect is negligible after a certain length of graphene.

It has also been reported that the Young's modulus decreases with decreasing number of carbon atoms in a graphene sheet.²⁶ In this study, the Young's modulus of graphene was found by combining continuum mechanics with Brenner's potential and applying the Cauchy-Born rule.²⁶ A similar size dependence of Young's modulus in

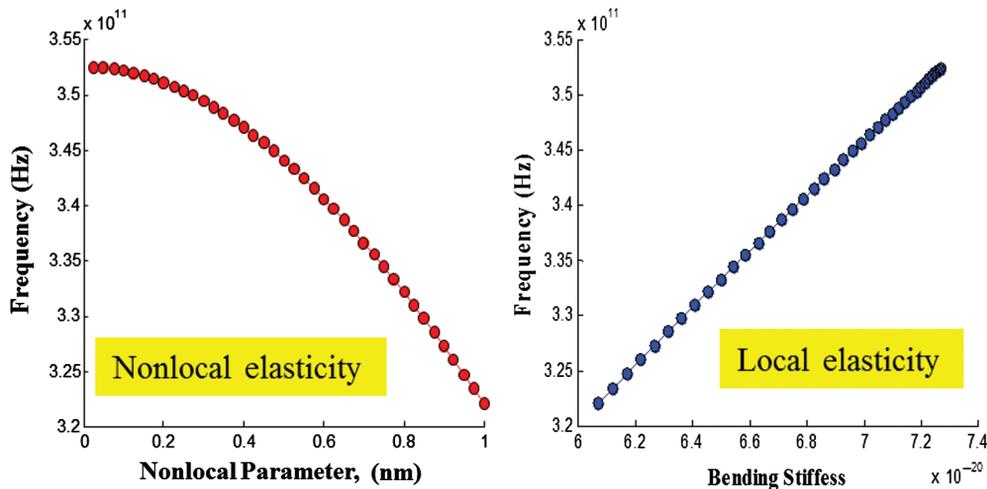


Fig. 2. (a) Variation of frequency with nonlocal parameter ($e_0 a$) for a single layer graphene sheets using nonlocal elasticity, (b) variation of frequency with bending stiffness for the same single layer graphene sheets using classical elasticity.

the form of bending rigidity of graphene is reported in Liu and Zhang.³⁰ In this study the bending rigidity K is a function of the bending rigidity at zero Kelvin, average thermal energy kT , temperature T and a parameter η as $K = K_0 - (3\eta kT)/(4\pi) + \dots$. The parameter η is dimensionless value and depends on the system size and a characteristic length (for example, the C–C bond length). It is worth noting that the single nonlocal parameter $e_0 a$ used in describing the nonlocal effects depends on the characteristic length and on the system size also. Though bending rigidity depends on temperature and loading conditions, it also depends on the Young's modulus. So when Young's modulus decreases, bending rigidity decreases. Taking the first two terms, if the dimensionless parameter η increases, the bending rigidity decreases. However it is not clear what the behaviour of η is with the increase or decrease of system size (e.g., graphene edge length). In summary, from the evidence discussed earlier, the cause for decreased frequencies and buckling loads is thus the consideration of size-independent over-predicted constant Young's modulus in Eqs. (1) and (4).

We now discuss about analogy of nonlocal effect in plate theory at small-scale. The nonlocal effect can be modelled equivalent as classical elasticity with a pseudo in-plane load. In a uniaxial buckling analysis of a graphene sheet where the nonlocal effects are active, the graphene sheet can be thought of a classical elastic plate with some virtual in-plane load. The virtual load or pseudo-in-plane load is a consequence of the nonlocal effects at this scale. The resulting nonlocal bending rigidity is less than the conventional one. Because of the presence of the pseudo-in-plane load, the buckling load is smaller than the classical buckling load. We say that the pseudo-in-plane load is a function of nonlocal parameter e_0 . For a biaxial buckling analysis, there would be pseudo-in-plane load both in the length and the width directions of rectangular graphene plate.

Similar in a uniaxial buckling analysis of a graphene sheet where the nonlocal effects are active, the vibrating graphene sheet can be thought of a classical elastic plate with some virtual in-plane load (related to curvature and compressive stress³¹). As a result the nonlocal bending rigidity is less than that of conventional one. Because of the presence of pseudo-in-plane load, the natural frequencies are smaller than the classical ones. Along the same lines, instead of pseudo-in-plane load we can think of a pseudo-mass adhered on the classical plate. Because of the presence of additional pseudo-mass the frequency is smaller than the classical one. The pseudo-mass would be a function of nonlocal parameter e_0 . For the particular experiment on graphene sheets with specific geometrical properties, we may get a definite exclusive value of nonlocal parameter. However it is expected that for other material properties of graphene sheets, the experiment result will predict different nonlocal parameter (as Young's modulus will be dependent on material properties). The nonlocal parameter will thus be function of material properties. Therefore there may different pseudo-mass for different nonlocal parameters. The nonlocal theory will be correct with proper choice of nonlocal parameter.

Another popular size-dependent plate theory used for small scale structures is the modified couple stress theory (MCST).³² How well the theory can be employed for graphene sheets (nanoplate) is a question, as the theory was originally used for micro-scale plates.³³ However recently, it is being applied to nanotubes and graphene sheets.³⁴ The theory predicts that the frequencies and buckling load increases with increase of scale parameter (equivalent to nonlocal parameter). This is contradictory to nonlocal elastic predictions. For size-dependent functionally graded beams, Reddy³⁵ has reported that the two theories (nonlocal and MCST³²) bring two different mechanisms into the upgraded theories. For MCST, the observation can be attributed to the fact that there would be

a hardening of bending stiffness (contradictory to nonlocal elasticity). We account for both the theories. The difference arises in both theories because of the consideration of different extrinsic length, i.e., for the MCST it is thickness of the plate whereas in NET it is the length of the specimen. Nonlocal plate theory does not take into account the effect of thickness (say SLGS of 100 m² and one-atomic thick). The hardening effect in MCST arises due to the twisting effect in the cross section of the specimen. Both of the predictions may be true as the Young's modulus (material properties) is not ambiguous and dependent on many parameters.^{8,24,28,30} For accurate prediction of structural behaviour of nanostructures, a united model may be necessary which considers both thickness and length as the related extrinsic scale parameters and provide an accurate model. The motive for combining NET and MCST would be to consider nonlocal or other small-scale effects in both transverse and in-plane directions of graphene sheets.

In summary, in this letter we analysed why the nonlocal natural frequencies and buckling loads of a graphene sheet decreases as the scale (nonlocal) parameter increases. The paper attempts to answer issues relating to frequencies and buckling loads of graphene sheets different from classical predictions. The concept of size-dependent Young's modulus and the pseudo-inplane load in nonlocal elasticity are discussed, which bridges the low effective structural stiffness (or frequency/buckling load) to its physical origin, i.e., nonlocal interaction among atoms. The employment of size-independent over-predicted constant Young's modulus in nonlocal elastic modelling is considered to be the reason for decreased frequencies and buckling load in monolayer graphene sheets. The analogy of pseudo-inplane load can be used for accounting the nonlocal effects. This letter also investigates the conflicting physics with other existing size-dependent theories such as modified couple stress theory for graphene sheets; and suggests a possible way forward. Further, points outlined in the letter could be accounted for the nonlocal elastic models applicable to other one-dimensional nanoscale structures such as nanotubes³⁶⁻⁴⁹ and nanorods⁵⁰⁻⁵³ also.

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