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# Large amplitude vibration of a bilayer graphene embedded in a nonlinear polymer matrix

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## HIGHLIGHTS

- ▶ Nonlinear vibration of a double layered graphene sheet embedded in polymer medium is studied.
- ▶ A new model is presented for the van der Waals interactions between graphene and matrix.
- ▶ Nonlinear natural frequencies are presented for both zigzag and armchair graphene sheets.

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## ABSTRACT

In the present article, nonlinear free and forced vibration of a bilayer graphene embedded in a polymer medium is studied based on the nonlocal elasticity theory. As a nonlinear function of deflection of graphene sheets, a refined pressure expression is established to describe the van der Waals (vdW) interactions between graphene layers and polymer medium. Assuming the large displacements and anisotropic model for graphene layers, the nonlinear couple partial differential equations of a double layered graphene sheet (DLGS) are obtained. The in-phase and out of phase nonlinear to linear natural frequencies are shown for both zigzag and armchair geometries. The effects of small scale parameter, nonlinear coefficients of vdW between two layers, nonlinear factor of polymer matrix and geometric properties on the nonlinear vibrational behavior of a DLGS are discussed in detail. It is found that the nonlinear vdW coefficient has a significant effect on the out of phase frequencies while the nonlinear polymer coefficient has considerable influence on in-phase frequencies.

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## 1. Introduction

Recently, researchers worldwide have engaged in fundamental studies of graphene sheets and have investigated the potential of their technological applications including graphene-based composite structures. Remarkable and unique properties of graphene sheets have attracted particular interest to serve them as excellent reinforcements for high performance polymer composites (see e.g. [1–2]). High stiffness and strength, superior fracture toughness and enhanced electrical conductivity of the graphene sheets produce composites with tailored physical and mechanical properties. Due to exceptional properties of graphene sheets, these two dimensional nano-structures become a suitable candidate as a reinforcement for polymers. It was shown that graphene

can significantly improve the physical properties of host polymers such as stiffness, strength, thermal and electrical conductivities and dimensional stability [3].

Due to difficulties in experimental analyses of nanostructures, there are several articles in literature that carried out the mechanical analysis of nanostructures such as nanotubes and graphene sheets based on the continuum mechanics concepts. The vibration analysis of multilayered graphene sheets using a continuum model was reported by He et al. [4]. They derived an explicit formula to predict the linear van der Waals (vdW) interaction between any two sheets. Behfar and Naghdabadi [5] investigated the nanoscale vibrational analysis of a multi-layered graphene sheet embedded in an elastic medium and determined the corresponding natural frequencies and the associated modes. Jiang et al. [6] established the cohesive law for interfaces between a carbon and polymer atoms which were not well bonded and were characterized by the van der Waals force. Xu et al. [7] studied the vibration of a double walled carbon nanotube in which the interlayer vdW forces was described as a nonlinear function of displacement. Based on the

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continuum mechanics concept and a multiple-elastic beam model, the nonlinear free vibration of embedded multiwall carbon nanotubes was carried out by Fu et al. [8] using the incremental harmonic balanced method. Liew et al. [9] proposed a continuum based plate model to derive the natural frequencies and associated vibration modes of multi-layered graphene sheets embedded in an elastic matrix. He et al. [10] used an elastic, multiple shell model for the vibration analysis of multi-walled carbon nanotubes. They modeled the vdW interaction between any two layers as the radius-dependent function. Duan and Wang [11] developed a molecular mechanics simulation to investigate the nonlinear deformation of a single layer, circular, graphene sheet under a central point load. Lu et al. [12] projected the vdW forces along the normal direction of the carbon nanotubes and showed that the vdW pressures are not the same on the walls but inversely proportional to wall radius. Mahdavi et al. [13] studied the nonlinear vibration of a single walled carbon nanotube embedded in a polymer matrix. The interfacial vdW forces were described by a nonlinear function in terms of deflection of carbon nanotubes. Murmu and Pradhan [14] used the nonlocal elasticity theory to study the vibration response of single layered graphene sheets embedded in an elastic medium. Ke et al. [15] considered the nonlinear free vibration of embedded double walled carbon nanotubes based on the Timoshenko beam model and von Karman geometric nonlinearity. They employed the differential quadrature method to solve the nonlinear governing equations.

Yang et al. [16] studied the nonlinear free vibration of single walled carbon nanotubes based on von Karman geometric nonlinearity and Eringen's nonlocal elasticity theory for different boundary conditions. The elastic modulus was obtained through molecular mechanics simulation. He et al. [17] derived a more refined pressure distribution expression to describe the van der Waals interaction between any two tubes of a multi-walled carbon nanotube to study the buckling and post-buckling of such materials. Shen et al. [18] presented nonlinear vibration behavior of a simply supported, rectangular, single layer graphene sheet in thermal environments and obtained the value of nonlocal parameter. The nonlocal 3-D Navier equations of motion were reformulated and decoupled by Jomehzadeh and Saidi [19] to study the vibration behavior of nano-plates. They [20] investigated the large amplitude vibration of multi-layered graphene sheets by considering linear vdW interactions between any two layers. Shen et al. [21] studied the nonlinear bending behavior of a single layer rectangular graphene sheet subjected to a transverse uniform load in thermal environments. Nonlinear bending, vibration and postbuckling analyses were investigated for a simply supported single thin film resting on a two-parameter elastic foundation by Shen [22]. Farajpour et al. [23] considered the small scale effect on buckling analysis of circular graphene sheets. Pradhan and Kumar [24] studied the small scale effect on the vibration analysis of orthotropic single layered graphene sheets. Ansari and Ramezannezhad [25] presented nonlinear vibrations of embedded multiwalled carbon nanotubes in thermal environments. Effects of nonlinear van der Waals interaction forces from both surrounding medium and adjacent tubes on nonlinear vibration of an embedded double-walled carbon nanotube were studied by Mahdavi et al. [26]. Recently, Wang et al. [27] developed a nonlinear continuum model for nonlinear vibration analysis of multi-layered isotropic graphene sheets without considering the small scale effect.

In most conditions of nano environments, the large displacements incessantly occur for nano devices including graphene sheets. The large amplitude vibration of graphene sheets is a key role in the design of resonator structural components. Also, in order to fully achieve the more accurate results, the van der Waals (vdW) interaction and polymer pressure should be refined

and the small length scale parameter should be considered. Moreover, the mechanical properties of graphene sheets are directionally dependent and the orthotropic material properties should be used to describe the mechanical properties of graphene layers. In this paper, the nonlinear free and forced vibrations of double layered orthotropic graphene sheets embedded in a polymer medium are presented by considering the nonlinear terms for both layers and polymer interactions. Based on the von Karman and nonlocal elasticity theory, the governing equations of motion are obtained for a double layered graphene sheet (DLGS) with arbitrary direction of chiral angle. The Galerkin's approximate method is used to reduce the governing nonlinear partial differential equations to ordinary differential equations and the harmonic balance method is employed to determine nonlinear frequencies of DLGS's. The effects of small length scale, nonlinear vdW interaction, nonlinear coefficient of polymer matrix, geometrical and anisotropic material properties on the nonlinear vibration behavior of DLGS's are studied in detail.

## 2. Modeling of nonlinear expression for vdW pressure

For a DLGS in a polymer medium, the interaction between layers is governed by the van der Waals force, which is characterized by the Lennard-Jones's pair potential. The vdW pressure between each layer of graphene and the surrounding medium can also be expressed by the Lennard-Jones potential. The Lennard-Jones 6–12 model is given as [28]

$$U_{ij} = 4\varepsilon \left[ \left( \frac{\sigma}{d} \right)^{12} - \left( \frac{\sigma}{d} \right)^6 \right] \quad (1)$$

where  $\varepsilon$  is the bond energy at the equilibrium distance, and it takes the values  $\varepsilon_{C-C} = 0.002390$  eV for carbon atoms of the graphene sheets and  $\varepsilon_{C-CH_2} = 0.004656$  eV for carbon atoms and the  $-CH_2-$  units of polyethylene [29].  $\sigma$  is a parameter which is determined by the equilibrium distance and its value is 0.3825 nm for carbon-polyethylene. Also,  $d$  is the distance between interacting atoms. Depending on the positions of atoms of the top layer relative to those on the bottom layer, there are two different stacking modes for the bilayer graphene (BLG), AA- and AB-stacking graphite, as their geometrical structures are shown in Fig. 1. Two graphene layers stacked directly on top of each other in AA-stacking while carbon atoms of one layer are placed in the center of Brillouin zone of other layer in the AB-stacking. Although the C–C bond length is the same for both of them, the equilibrium distance between two layers is different in both of them, which is nearly 3.59 Å for AA-stacking BG and 3.31 Å for the AB-stacking one [30].

The vdW force can be obtained by taking the derivative of the Lennard-Jones pair potential with respect to distance  $d$  as

$$F(d) = -\frac{\partial U_{ij}}{\partial d} = \frac{24\varepsilon}{\sigma} \left[ 2 \left( \frac{\sigma}{d} \right)^{13} - \left( \frac{\sigma}{d} \right)^7 \right] \quad (2)$$

It can be seen that the van der Waals force is a highly nonlinear function of distance. Therefore, it is not reasonable to model it by a linear function. Expressing the Taylor expansion of the vdW force around the equilibrium position  $\bar{d}$ , and taking into account that the vdW force is an odd function of the interlayer spacing, one can easily obtain:

$$F(d) = -\frac{24\varepsilon}{\sigma^2} \left[ 26 \left( \frac{\sigma}{\bar{d}} \right)^{14} - 7 \left( \frac{\sigma}{\bar{d}} \right)^8 \right] (d - \bar{d}) - \frac{336\varepsilon}{\sigma^4} \left[ 65 \left( \frac{\sigma}{\bar{d}} \right)^{16} - 6 \left( \frac{\sigma}{\bar{d}} \right)^{10} \right] (d - \bar{d})^3 \quad (3)$$

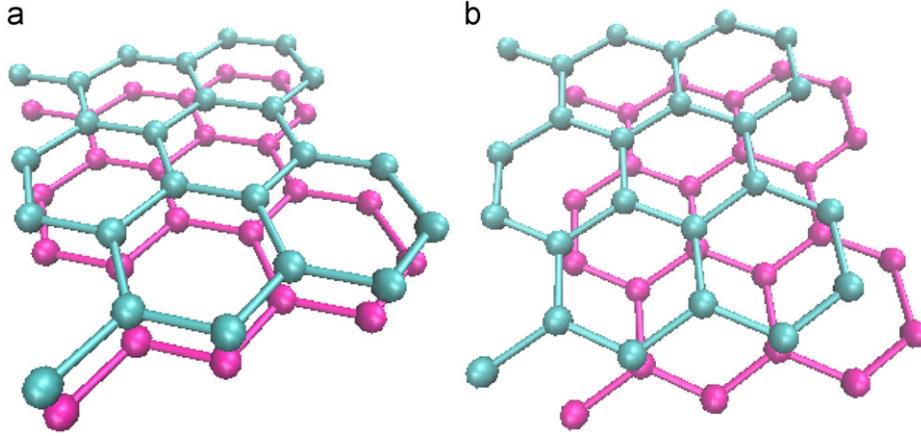


Fig. 1. Two different geometrical configurations of a bilayer graphene. (a) AA-stacking (b) AB-stacking.

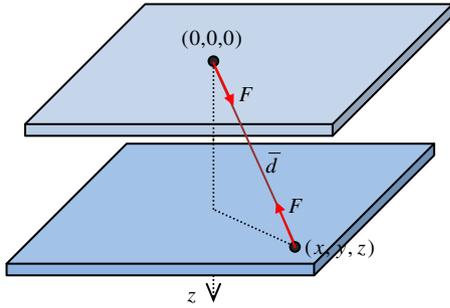


Fig. 2. Schematic diagram for direction of forces of graphene layers.

The Taylor expansion of the vdW force between atoms is truncated to third term in order to obtain the more precise result. As it can be seen from Fig. 2, this vdW force is along the atoms direction. In order to find the interaction pressure between two layers of the graphene sheets in  $z$  direction, the vdW force should be projected along the  $z$  direction and then integrated over the entire sheet as

$$q_{gr-gr} = \rho_C^2 \left\{ -\frac{24\epsilon_{C-C}}{\sigma_{C-C}^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \left[ 26 \left( \frac{\sigma_{C-C}}{\sqrt{x^2+y^2+z^2}} \right)^{14} - 7 \left( \frac{\sigma_{C-C}}{\sqrt{x^2+y^2+z^2}} \right)^8 \right] (d-\bar{d}) \frac{z}{d} dx dy - \frac{336\epsilon_{C-C}}{\sigma_{C-C}^4} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \left[ 65 \frac{\sigma_{C-C}^{16}}{(\sqrt{x^2+y^2+z^2})^{14}} - 6 \frac{\sigma_{C-C}^{10}}{(\sqrt{x^2+y^2+z^2})^8} \right] (d-\bar{d})^3 \frac{z}{d^3} dx dy \right\} \quad (4)$$

where  $\rho_C = 4/3\sqrt{3}l_c^2$  is the area density of the carbon atoms and  $l_c$  is the carbon–carbon bond length. By considering relation between the distance difference of two atoms and transverse displacements of the graphene layers ( $d-\bar{d} = (w_1-w_2)d/z$ ), the vdW pressure between two layers of graphene in Eq. (4) can be expressed as

$$q_{gr-gr} = -\rho_C^2 \left( \frac{24\epsilon_{C-C}}{\sigma_{C-C}^2} \right) \left[ \frac{13\pi\sigma_{C-C}^{14}}{3h^{12}} - \frac{7\pi\sigma_{C-C}^8}{3h^6} \right] (w_1-w_2) - \rho_C^2 \left( \frac{336\epsilon_{C-C}}{\sigma_{C-C}^4} \right) \left[ \frac{65\pi\sigma_{C-C}^{16}}{6h^{14}} - \frac{2\pi\sigma_{C-C}^{10}}{h^8} \right] (w_1-w_2)^3 = c_1(w_1-w_2) + c_3(w_1-w_2)^3 \quad (5)$$

where  $h$  is the distance between two layers,  $w_1$  and  $w_2$  are transverse displacements of bottom and top layers, respectively. It can be seen that the pressure at a point between two layers is a nonlinear function of their deflections at that point and the vdW force is also projected in transverse direction to obtain more accurate results.

The interaction force between the carbon atoms of a graphene layer and polymer medium is also van der Waals force. In order to find the interaction pressure between a layer of graphene and the polymer matrix in  $z$  direction, the vdW force in Eq. (3) should be projected in transverse direction and integrated over the entire volume of the polymer medium. By considering the distance difference of two atoms in terms of transverse displacements of the bottom or top layers  $w_i$  ( $i=1,2$ ), the vdW interaction between the graphene layer and polymer matrix can be expressed as

$$q_{gr-pol} = \rho_C \rho_{CH_2} \left\{ -\frac{24\epsilon_{C-CH_2}}{\sigma_{C-CH_2}^2} \int_{-h_i}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \left[ 26 \left( \frac{\sigma_{C-CH_2}}{\sqrt{x^2+y^2+z^2}} \right)^{14} - 7 \left( \frac{\sigma_{C-CH_2}}{\sqrt{x^2+y^2+z^2}} \right)^8 \right] w_i dx dy dz - \frac{336\epsilon_{C-CH_2}}{\sigma_{C-CH_2}^4} \int_{-h_i}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \left[ 65 \frac{\sigma_{C-CH_2}^{16}}{(\sqrt{x^2+y^2+z^2})^{14}} - 6 \frac{\sigma_{C-CH_2}^{10}}{(\sqrt{x^2+y^2+z^2})^8} \right] \frac{w_i^3}{z^2} dx dy dz \right\} \quad (6)$$

where  $\rho_{C-CH_2}$  is the volume density of polymer molecules (number of polymer molecules per unit volume) which is equal to  $3.1 \times 10^{28} \text{ 1/m}^3$  for the  $-\text{CH}_2-$  unit in polyethylene and  $h_i$  is the distance between polymer medium and each layer of graphene sheets. Also, subscript  $i$  represents layer number which is 1 for bottom layer and 2 for top layer. The interaction pressure between a layer of graphene and polymer matrix can be simplified as

$$q_{gr-pol} = -\rho_C \rho_{CH_2} \left( \frac{8\epsilon_{C-CH_2}}{\sigma_{C-CH_2}^2} \right) \left[ -\frac{13\pi\sigma_{C-CH_2}^{14}}{11h^{11}} + \frac{7\pi\sigma_{C-CH_2}^8}{5h^5} \right] w_i - \rho_C \rho_{CH_2} \left( \frac{\epsilon_{C-CH_2}}{\sigma_{C-CH_2}^4} \right) \left[ -\frac{280\pi\sigma_{C-CH_2}^{16}}{h_i^{13}} + \frac{96\pi\sigma_{C-CH_2}^{10}}{h_i^7} \right] w_i^3 = k_{1i}w_i + k_{3i}w_i^3 \quad (7)$$

In this model, unlike the previous models, the vdW pressure of the polymer medium has been expanded to the third order of Taylor's series. Therefore, this model is capable to capture the

nonlinear effect of polymer matrix on vibrational behavior of DLGS's and is more accurate than the previous models.

For a specific model of DLGS in a polyethylene medium where the equilibrium distances are  $h_1 = 0.858\sigma_{C-CH_2}$  and  $h_2 - h_1 = 0.341 \text{ nm}$  [31], the numerical values of the vdW coefficients can be obtained as

$$\begin{aligned} c_1 &= -84.1593 \text{ GPa/nm} & c_3 &= -44621.4053 \text{ GPa/nm}^3 \\ k_{11} &= 28.4941 \text{ GPa/nm} & k_{31} &= 12825.3287 \text{ GPa/nm}^3 \\ k_{12} &= -0.7720 \text{ GPa/nm} & k_{32} &= -0.1407 \text{ GPa/nm}^3 \end{aligned} \quad (8)$$

Also, the vdW coefficients can be obtained as  $c_1 = -27.3546 \text{ GPa/nm}$ ,  $c_3 = -2041.1352 \text{ GPa/nm}^3$  and  $c_1 = -146.8247 \text{ GPa/nm}$ ,  $c_3 = -7176.3890 \text{ GPa/nm}^3$  for AA- and AB-stacking bilayer graphenes, respectively.

Note that the negative and positive signs represent an attraction and repulsion between the atoms, respectively. It can be seen that the effects of polymer matrix on top layer is negligible with respect to its effect on bottom layer.

### 3. Nonlocal equations of motion for a DLGS

Consider a double layered orthotropic graphene sheet resting on an elastic foundation as a polymer medium (Fig. 3). The length of each sheet is  $a$  in  $x$ -direction, the width is  $b$  in  $y$ -direction and the thickness is  $h$ . Also, the coordinate system is assumed to be at the middle of each layer. Based on the classical plate theory, the displacement components of an arbitrary point of the sheet can be expressed as

$$\begin{aligned} u_{1i} &= u_i(x,y,t) - z \frac{\partial w_i(x,y,t)}{\partial x} \\ u_{2i} &= v_i(x,y,t) - z \frac{\partial w_i(x,y,t)}{\partial y} \\ u_{3i} &= w_i(x,y,t) \end{aligned} \quad (9)$$

where  $u_i(x,y,t)$ ,  $v_i(x,y,t)$  and  $w_i(x,y,t)$  are displacement components of the midplane of each layer in the  $x$ ,  $y$  and  $z$  directions, respectively and  $t$  is time. Also, subscript  $i$  represents layer number. Since the graphene sheet is assumed to have large amplitude deformation, the nonlinear von Karman type strain–displacement relations are used as

$$\begin{aligned} \varepsilon_{xi} &= \varepsilon_{x0i} + ZK_{xi} \\ \varepsilon_{yi} &= \varepsilon_{y0i} + ZK_{yi} \\ \gamma_{xyi} &= \gamma_{xy0i} + ZK_{xyi} \end{aligned} \quad (10)$$

where in-plane strains and curvature parameters are defined as

$$\left\{ \begin{matrix} \varepsilon_{x0i} \\ \varepsilon_{y0i} \\ \gamma_{xy0i} \end{matrix} \right\} = \left\{ \begin{matrix} \frac{\partial u_i}{\partial x} + \frac{1}{2} \left( \frac{\partial w_i}{\partial x} \right)^2 \\ \frac{\partial v_i}{\partial y} + \frac{1}{2} \left( \frac{\partial w_i}{\partial y} \right)^2 \\ \frac{\partial u_i}{\partial y} + \frac{\partial v_i}{\partial x} + \frac{\partial w_i}{\partial x} \frac{\partial w_i}{\partial y} \end{matrix} \right\}, \quad \left\{ \begin{matrix} K_{xi} \\ K_{yi} \\ K_{xyi} \end{matrix} \right\} = \left\{ \begin{matrix} -\frac{\partial^2 w_i}{\partial x^2} \\ -\frac{\partial^2 w_i}{\partial y^2} \\ -2 \frac{\partial^2 w_i}{\partial x \partial y} \end{matrix} \right\} \quad (11)$$

The equations of motion for a DLGS can be obtained by using the Hamilton's principle. The Hamilton's principle states that

$$\int_0^t (\delta T - \delta U + \delta V) dt = 0 \quad (12)$$

where  $T$ ,  $U$  and  $V$  are kinetic energy, strain energy of the graphene layers and potential energy of the external loads, respectively. Expressing these parameters based on the von Karman theory, the nonlinear equations of motion for a DLGS can be obtained as follows [20]:

$$\frac{\partial N_{xi}}{\partial x} + \frac{\partial N_{xyi}}{\partial y} = \left( I_{0i} \ddot{u}_i - I_{1i} \frac{\partial \ddot{w}_i}{\partial x} \right) \quad (13a)$$

$$\frac{\partial N_{xyi}}{\partial x} + \frac{\partial N_{yi}}{\partial y} = \left( I_{0i} \ddot{v}_i - I_{1i} \frac{\partial \ddot{w}_i}{\partial y} \right) \quad (13b)$$

$$\begin{aligned} \frac{\partial^2 M_{xi}}{\partial x^2} + 2 \frac{\partial^2 M_{xyi}}{\partial x \partial y} + \frac{\partial^2 M_{yi}}{\partial y^2} + \frac{\partial}{\partial x} \left( N_{xi} \frac{\partial w_i}{\partial x} + N_{xyi} \frac{\partial w_i}{\partial y} \right) \\ + \frac{\partial}{\partial y} \left( N_{xyi} \frac{\partial w_i}{\partial x} + N_{yi} \frac{\partial w_i}{\partial y} \right) + (q_{gr-gr} + q_{gr-pol})_i + P_i \\ = I_{0i} \ddot{w}_i + I_{1i} \left( \frac{\partial \ddot{u}_i}{\partial x} + \frac{\partial \ddot{v}_i}{\partial y} \right) - I_{2i} \left( \frac{\partial^2 \ddot{w}_i}{\partial x^2} + \frac{\partial^2 \ddot{w}_i}{\partial y^2} \right) \end{aligned} \quad (13c)$$

where a dot denotes differentiation with respect to time and  $P_i$  is the external load in which involves two parts, first due to external pressure exerted by the van der Waals forces and second due to the applied force or disturbance in the forced vibration analysis.  $I_j$ 's ( $j=0,1,2$ ) are the inertia parameters which are expressed in term of the mass density of the graphene sheets ( $\rho_i$ ) as

$$(I_{0i}, I_{1i}, I_{2i}) = \int_{-h/2}^{h/2} \rho_i(1, z, z^2) dz \quad (14)$$

In order to obtain the nonlocal governing equations of motion, the resultant forces and moments in Eq. (13) should be described based on the nonlocal elasticity theory. According to nonlocal elasticity theory, the stress at a definite point in a body depends not only on the strain at that point but also on those at all other points of the body. Nonlocal continuum mechanics allows one to account for the small length scale effect that becomes significant when dealing with microstructures or nanostructures. Eringen [32] showed that the nonlocal stress tensor can be related to local stress tensor as

$$[1 - (e_0 l)^2 \nabla^2] \sigma = \sigma' \quad (15)$$

where  $e_0 l$  is a small scale parameter,  $e_0$  is a constant to adjust the model to match the reliable experimental results,  $l$  is an internal characteristic length such as C–C bond length and  $\nabla^2$  is the Laplacian operator.

Due to the hexagonal structure of the unit cells in graphene sheets, the mechanical properties of these structures are anisotropic. The directionally dependent properties of the graphene sheets depend on the direction of chiral angle. Obtaining the force and moments resultants for an anisotropic graphene sheet and

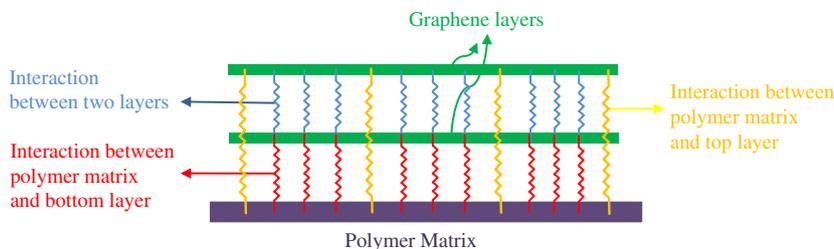


Fig. 3. Double layered graphene sheet embedded in a polymer matrix.

introducing the stress function  $\varphi$  as

$$N_{xi} = \frac{\partial^2 \varphi_i}{\partial y^2}, N_{yi} = \frac{\partial^2 \varphi_i}{\partial x^2}, N_{xyi} = -\frac{\partial^2 \varphi_i}{\partial x \partial y}, \tag{16}$$

and considering the small scale effect, the nonlocal nonlinear governing equations of motion for a double layered orthotropic graphene sheet in a polymer matrix can be obtained as

$$\begin{aligned} D_{11} \frac{\partial^4 w_1}{\partial x^4} + 2(D_{12} + 2D_{33}) \frac{\partial^4 w_1}{\partial x^2 \partial y^2} + D_{22} \frac{\partial^4 w_1}{\partial y^4} \\ + (I_0 - I_2) [1 - (e_0 l)^2 \nabla^2] (\dot{w}_1 + \nabla^2 \dot{w}_1) \\ = [1 - (e_0 l)^2 \nabla^2] [c_1 (w_1 - w_2) + c_3 (w_1 - w_2)^3 - k_{11} w_1 - k_{31} w_1^3 + p_1] \\ + [1 - (e_0 l)^2 \nabla^2] \left( \frac{\partial^2 w_1}{\partial x^2} \frac{\partial^2 \varphi_1}{\partial y^2} - 2 \frac{\partial^2 w_1}{\partial x \partial y} \frac{\partial^2 \varphi_1}{\partial x \partial y} + \frac{\partial^2 w_1}{\partial y^2} \frac{\partial^2 \varphi_1}{\partial x^2} \right) \end{aligned} \tag{17a}$$

$$\begin{aligned} A_{11} \frac{\partial^4 \varphi_1}{\partial x^4} + 2(A_{12} + 2A_{33}) \frac{\partial^4 \varphi_1}{\partial x^2 \partial y^2} + A_{22} \frac{\partial^4 \varphi_1}{\partial y^4} \\ = \left( \frac{\partial^2 w_1}{\partial x \partial y} \right)^2 - \frac{\partial^2 w_1}{\partial x^2} \frac{\partial^2 w_1}{\partial y^2} \end{aligned} \tag{17b}$$

$$\begin{aligned} D_{11} \frac{\partial^4 w_2}{\partial x^4} + 2(D_{12} + 2D_{33}) \frac{\partial^4 w_2}{\partial x^2 \partial y^2} + D_{22} \frac{\partial^4 w_2}{\partial y^4} \\ + (I_0 - I_2) [1 - (e_0 l)^2 \nabla^2] (\dot{w}_2 + \nabla^2 \dot{w}_2) \\ = [1 - (e_0 l)^2 \nabla^2] [-c_1 (w_1 - w_2) - c_3 (w_1 - w_2)^3 - k_{12} w_2 - k_{32} w_2^3 + p_2] \\ + [1 - (e_0 l)^2 \nabla^2] \left( \frac{\partial^2 w_2}{\partial x^2} \frac{\partial^2 \varphi_2}{\partial y^2} - 2 \frac{\partial^2 w_2}{\partial x \partial y} \frac{\partial^2 \varphi_2}{\partial x \partial y} + \frac{\partial^2 w_2}{\partial y^2} \frac{\partial^2 \varphi_2}{\partial x^2} \right) \end{aligned} \tag{17c}$$

$$A_{11} \frac{\partial^4 \varphi_2}{\partial x^4} + 2(A_{12} + 2A_{33}) \frac{\partial^4 \varphi_2}{\partial x^2 \partial y^2} + A_{22} \frac{\partial^4 \varphi_2}{\partial y^4} = \left( \frac{\partial^2 w_2}{\partial x \partial y} \right)^2 - \frac{\partial^2 w_2}{\partial x^2} \frac{\partial^2 w_2}{\partial y^2} \tag{17d}$$

where the parameters  $A_{jk}$  and  $D_{jk}$  ( $jk=11, 12, 22$  and  $33$ ) are the constant coefficients which are defined in terms of mechanical properties of graphene sheets in [Appendix A](#), also, the local force moments are considered as ( $N = \sum_{n=0}^{\infty} (e_0 l_i)^{2n} \nabla^{2n} N$ ).

It can be seen that the governing equations of motion of a DLGS embedded in a polymer matrix based on the von Karman assumptions are four nonlinear coupled partial differential equations with total degree of sixteen in each direction in terms of transverse displacement and stress function. It is assumed that the properties of both layers are the same. The nonlinear terms in these equations occur due to three issues as large displacement assumption for graphene sheets, nonlinear behavior of vdW interaction between two layers and nonlinear coefficients in pressure interaction of the polymer matrix.

Also, by the help of strain–displacement relations, the in-plane displacement components can be expressed in terms of the transverse deflection  $w_i$  and stress function  $\varphi_i$  as

$$u_i = \int_0^x \left( A_{11} \frac{\partial^2 \varphi_i}{\partial x^2} + A_{12} \frac{\partial^2 \varphi_i}{\partial y^2} + A_{13} \frac{\partial^2 \varphi_i}{\partial x \partial y} - \frac{1}{2} \left( \frac{\partial w_i}{\partial x} \right)^2 \right) dx \tag{18a}$$

$$v_i = \int_0^y \left( A_{12} \frac{\partial^2 \varphi_i}{\partial x^2} + A_{22} \frac{\partial^2 \varphi_i}{\partial y^2} + A_{23} \frac{\partial^2 \varphi_i}{\partial x \partial y} - \frac{1}{2} \left( \frac{\partial w_i}{\partial y} \right)^2 \right) dy \tag{18b}$$

Hence, by defining the transverse displacement and stress function, the in-plane displacements of the graphene sheets can be also obtained.

#### 4. Free vibration analysis

Let us consider the large amplitude free vibration analysis of double layered graphene sheets with all edges movable simply supported or totally clamped boundary conditions. In this case, the external loads due to applied forces on graphene layers are considered to be zero. For the movable simply supported edges, which are kept straight by a distribution of normal stresses, and clamped edges, the following boundary conditions require:

Simply supported:

$$\begin{aligned} w_i = 0, \quad M_{xi} = 0, \quad \frac{\partial^2 \varphi_i}{\partial x \partial y} = 0, \quad \int_{-b/2}^{b/2} \frac{\partial^2 \varphi_i}{\partial y^2} dy = 0 \quad \text{at } x = \pm \frac{a}{2} \\ w_i = 0, \quad M_{yi} = 0, \quad \frac{\partial^2 \varphi_i}{\partial x \partial y} = 0, \quad \int_{-a/2}^{a/2} \frac{\partial^2 \varphi_i}{\partial x^2} dx = 0 \quad \text{at } y = \pm \frac{b}{2} \end{aligned} \tag{19a}$$

Clamped:

$$\begin{aligned} w_i = \frac{\partial w_i}{\partial x} = \frac{\partial^2 \varphi_i}{\partial x \partial y} = \int_{-b/2}^{b/2} \frac{\partial^2 \varphi_i}{\partial y^2} dy = 0 \quad \text{at } x = \pm \frac{a}{2} \\ w_i = \frac{\partial w_i}{\partial y} = \frac{\partial^2 \varphi_i}{\partial x \partial y} = \int_{-a/2}^{a/2} \frac{\partial^2 \varphi_i}{\partial x^2} dx = 0 \quad \text{at } y = \pm \frac{b}{2} \end{aligned} \tag{19b}$$

In view of the boundary conditions given in Eq. (19), the vibration mode of each layer in transverse direction is assumed to be

For simply supported graphene sheet:

$$w_i = h W_i(t) \cos\left(\frac{n\pi x}{a}\right) \cos\left(\frac{m\pi y}{b}\right) \tag{20a}$$

For clamped graphene sheet:

$$w_i = h W_i(t) \cos^2\left(\frac{n\pi x}{a}\right) \cos^2\left(\frac{m\pi y}{b}\right) \tag{20b}$$

where  $n$  and  $m$  are the numbers of half waves in  $x$  and  $y$  directions, respectively and  $W_i(t)$  is the nondimensional transverse displacement (amplitude). It can be found that the admissible function (20) satisfies the first two boundary conditions in Eq. (19). Substituting Eq. (20) into the right side of Eqs. (17b and d), the general solutions for the stress function  $\varphi_i$  of simply supported bilayer graphene can be obtained as

$$\varphi_1 = \frac{h^2}{32n^2 m^2 a^2 b^2} \left( \frac{m^4 a^4 \cos(2n\pi x/a)}{A_{22}} + \frac{n^4 b^4 \cos(2m\pi y/b)}{A_{11}} \right) W_1^2(t) \tag{21a}$$

$$\varphi_2 = \frac{h^2}{32n^2 m^2 a^2 b^2} \left( \frac{m^4 a^4 \cos(2n\pi x/a)}{A_{22}} + \frac{n^4 b^4 \cos(2m\pi y/b)}{A_{11}} \right) W_2^2(t) \tag{21b}$$

These functions exactly satisfy two last boundary conditions in Eq. (19) (the in-plane boundary conditions). Substituting the transverse displacement  $w_i$  for  $i=1$  and  $2$  and the stress function from Eqs. (20) and (21) into the basic Eqs. (17a and c) and then using the Galerkin method, two modal equations are obtained as

$$\begin{aligned} \frac{d^2 W_1(t)}{dt^2} + \alpha_1 W_1(t) + \beta_1 W_1^3(t) + \gamma W_2(t) \\ + \lambda W_2^3(t) + \eta W_1(t) W_2^2(t) + \delta W_1^2(t) W_2(t) = 0 \end{aligned} \tag{22a}$$

$$\begin{aligned} \frac{d^2 W_2(t)}{dt^2} + \alpha_2 W_2(t) + \beta_2 W_2^3(t) + \gamma W_1(t) + \lambda W_1^3(t) + \eta W_1^2(t) W_2(t) \\ + \delta W_1(t) W_2^2(t) = 0 \end{aligned} \tag{22b}$$

where the modal coefficients for a bilayer graphene are defined as

$$\alpha_1 = -\frac{12a^2b^2[a^2b^2 + \pi^2(e_0l)^2a^2 + \pi^2(e_0l)^2b^2](c_1 - k_{11}) - \pi^4[2a^2b^2(Q_{12} + 2Q_{33}) + Q_{22}a^4 + Q_{11}b^4]h^3}{(12a^2b^2 + a^2h^2\pi^2 + b^2h^2\pi^2)(a^2b^2 + a^2\mu\pi^2 + b^2\mu\pi^2)\rho h} \quad (23a)$$

$$\beta_1 = \frac{3\pi^4h^2(a^4Q_{22} + b^4Q_{11})(Q_{11}Q_{22} - Q_{12}^2) - 9a^4b^4Q_{11}Q_{22}(c_3 - k_{31})}{4\rho Q_{11}Q_{22}a^2b^2(12a^2b^2 + a^2h^2\pi^2 + b^2h^2\pi^2)} \quad (23c)$$

$$\beta_2 = \frac{3\pi^4h^2(a^4Q_{22} + b^4Q_{11})(Q_{11}Q_{22} - Q_{12}^2) - 9a^4b^4Q_{11}Q_{22}(c_3 - k_{32})}{4\rho Q_{11}Q_{22}a^2b^2(12a^2b^2 + a^2h^2\pi^2 + b^2h^2\pi^2)} \quad (23d)$$

$$\gamma = \frac{12c_1a^2b^2}{\rho h(12a^2b^2 + a^2h^2\pi^2 + b^2h^2\pi^2)} \quad (23e)$$

$$\lambda = \frac{27c_3a^2b^2h}{4\rho(12a^2b^2 + a^2h^2\pi^2 + b^2h^2\pi^2)} \quad (23f)$$

$$\eta = -\delta = -\frac{81c_3a^2b^2h}{4\rho(12a^2b^2 + a^2h^2\pi^2 + b^2h^2\pi^2)} \quad (23g)$$

It is noted that when the nonlinear terms and elastic foundation are neglected, i.e.,  $c_3 = k_{11} = k_{31} = k_{21} = k_{32} = 0$ , all of the above relations can be reduced to the corresponding equations in Ref. [20].

For linear vibration of the graphene sheets in which the nonlinear terms are neglected, two distinct linear natural frequencies are obtained as

$$\omega_{L,1}^2 = \frac{\alpha_1 + \alpha_2 - \sqrt{\alpha_1^2 + \alpha_2^2 - 2\alpha_1\alpha_2 + 4\gamma^2}}{2} \quad (24a)$$

$$\omega_{L,2}^2 = \frac{\alpha_1 + \alpha_2 + \sqrt{\alpha_1^2 + \alpha_2^2 - 2\alpha_1\alpha_2 + 4\gamma^2}}{2} \quad (24b)$$

where  $\omega_{L,1}$  and  $\omega_{L,2}$  stand for the lowest linear natural frequencies corresponding to the in-phase and out of phase vibrational modes, respectively. It can be found that the linear vibration analysis cannot consider the amplitude of the free vibrational modes. In fact, the deflection amplitude depends on the frequency which is determined by nonlinear vibration analysis.

In order to find the nonlinear natural frequencies of DLGS's, the Harmonic Balance (HB) method is employed. The HB method is an analytical approach for solving nonlinear oscillators, in which the initial conditions are generally simplified by setting velocity or displacement equal to zero [33].

Here, the periodic solutions are considered for Eq. (22), and the following two algebraic equations are obtained by applying the HB method

$$\omega_{NL}^2 = \alpha_1 + \frac{3\beta_1}{4}W_1^2 + \gamma\frac{W_2}{W_1} + \frac{3\lambda}{4}\frac{W_1^3}{W_1} + \frac{3\eta}{4}W_2^2 + \frac{3\delta}{4}W_1W_2 \quad (25a)$$

$$\omega_{NL}^2 = \alpha_2 + \frac{3\beta_2}{4}W_2^2 + \gamma\frac{W_1}{W_2} + \frac{3\lambda}{4}\frac{W_1^3}{W_2} + \frac{3\eta}{4}W_1^2 + \frac{3\delta}{4}W_1W_2 \quad (25b)$$

From the above two equations, the nonlinear frequency ( $\omega_{NL}$ ) and amplitude ratio ( $W_1/W_2$ ) of the double layered graphene sheet embedded in a nonlinear elastic medium can be determined.

## 5. Forced vibration analysis

Consider now the motion which results when a periodic external force  $p_2 = p_0 \cos(\omega t)$  is applied to the top layer of graphene sheet. In this case, the modal equations can be obtained in the following nonlinear forms:

$$\frac{d^2W_1(t)}{dt^2} + \alpha_1W_1(t) + \beta_1W_1^3(t) + \gamma W_2(t) + \lambda W_2^3(t) + \eta W_1(t)W_2^2(t) + \delta W_1^2(t)W_2(t) = 0 \quad (26a)$$

$$\frac{d^2W_2(t)}{dt^2} + \alpha_2W_2(t) + \beta_2W_2^3(t) + \gamma W_1(t) + \lambda W_1^3(t) + \eta W_1^2(t)W_2(t) + \delta W_1(t)W_2^2(t) = P \cos(\omega t) \quad (26b)$$

As it can be seen, because of the external force on the top layer, the second modal equation is a non-homogeneous equation. The coefficient  $P$  in this equation is defined as

$$P = \frac{192a^4b^4}{\rho h^2\pi^2(12a^2b^2 + \pi^2h^2b^2 + \pi^2h^2a^2)(\pi^2\mu b^2 + \pi^2\mu a^2 + a^2b^2)}p_0 \quad (27)$$

We consider the harmonic oscillations in which the period is the same as the period  $2\pi/\omega$  of the external force  $p_0 \cos(\omega t)$ . Using the HB method, the relation between amplitude and oscillation frequency of a double layered graphene sheet can be determined.

## 6. Numerical results

In the following section, numerical results are presented for the vibrational frequencies of double layered graphene sheets (DLGS's). The material properties of graphene sheets are presented in Table 1. They were obtained by Shen et al. [18] by comparing the continuum and molecular dynamic results.

In order to verify the accuracy of the results, the obtained natural frequencies for a clamped zigzag single layer graphene sheet are compared with molecular simulation using the REBO potential in Table 2. It can be seen that the results have a good agreement with ones in literature especially in the nonlinear case.

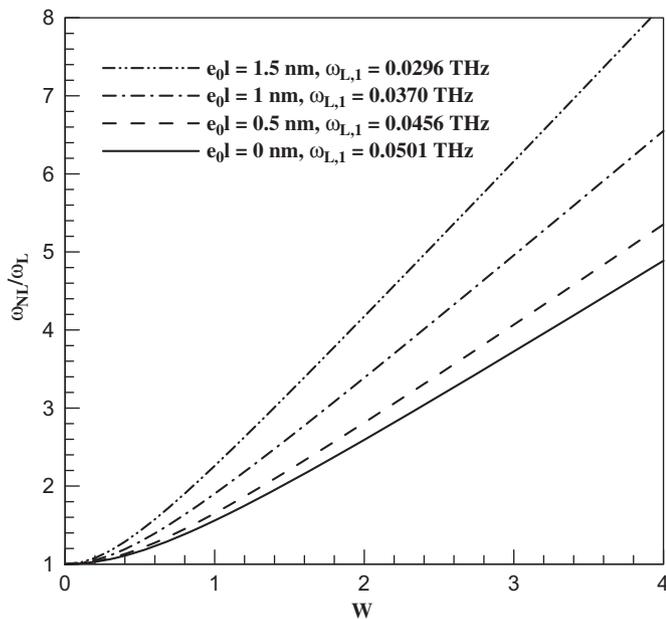
The numerical results are presented for simply supported graphene sheets and dimensions of the layers are assumed to be 4.888 nm  $\times$  4.855 nm for armchair and 1.987 nm  $\times$  1.974 nm for

**Table 1**  
Material properties of graphene layers.

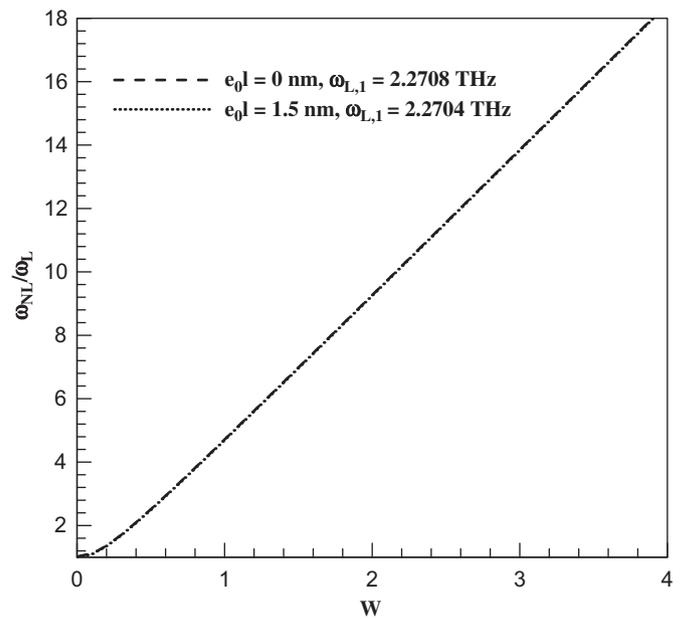
Type of graphene sheet	Small scale $e_0l$ (nm)	Thickness $h$ (nm)	Density $\rho$ (Kg/m <sup>3</sup> )	$-x$ Young modulus $E_{11}$ (TPa)	$-y$ Young modulus $E_{22}$ (TPa)	Shear modulus $G_{12}$ (TPa)	Poisson's ratio $\nu_{12}$
Armchair	0.27	0.156	5295	1.949	1.962	0.846	0.201
Zigzag	0.22	0.154	5363	1.987	1.974	0.857	0.205

**Table 2**  
Comparison of the natural frequencies (GHz) of a clamped single layer graphene sheet.

		Dimensions			
		10 nm × 10 nm	20 nm × 10 nm	10 nm × 20 nm	20 nm × 20 nm
<b>Molecular Simulation [34]</b>		27.28	18.86	18.47	6.94
<b>Present</b>	$\omega_{NL}$	27.52, $W=0$ nm	18.86, $W=0.75$ nm	18.47, $W=0.66$ nm	6.94, $W=0.2$ nm
	$\omega_L$	27.52	17.20	17.16	6.90



**Fig. 4.** In-phase nonlinear frequency ratio versus the amplitude of the top graphene sheet for various small scale parameters.



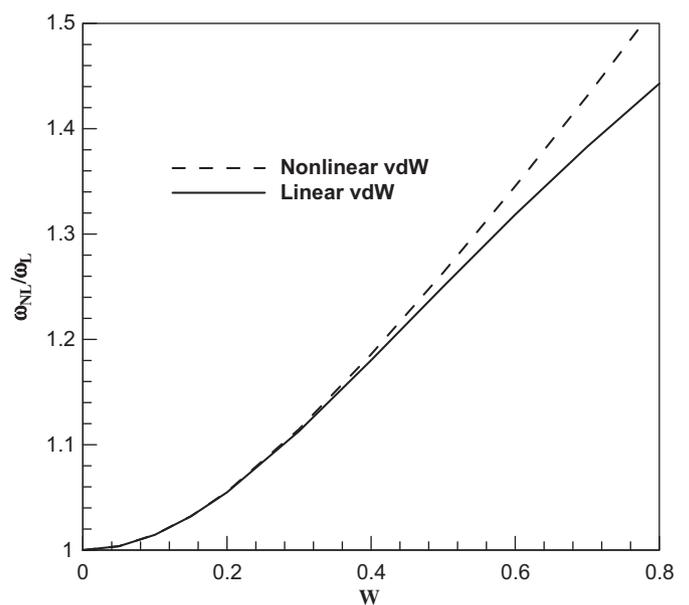
**Fig. 5.** Out of phase nonlinear frequency ratio versus the amplitude of the top graphene sheet for various small scale parameters.

zigzag grapheme sheets. In all figures, the amplitude indicates the nondimensional displacement of the top graphene sheet ( $W = W_2(t)$ ).

In order to study the effect of the small scale parameter ( $e_0l$ ) on the nonlinear frequency ratio  $\omega_{NL}/\omega_L$  (nonlinear to linear frequencies), the backbone curves are depicted in Fig. 4 for a double layered zigzag graphene sheet without polymer medium for various small scale parameters. The nonlinear frequency ratio in this figure is in-phase frequency. It can be seen that the nonlinear frequency ratio increases by increasing the small scale parameter while the linear frequency will decrease. This effect is more significant for higher values of amplitude.

The out of phase nonlinear frequency is depicted in Fig. 5 for the above case. It can be concluded that the small scale parameter does not have a significant effect on the out of phase frequency of the double layered graphene sheet. Also, since there is no polymer matrix in this case, the amplitude ratio ( $W_1/W_2$ ) is 1 and  $-1$  for in-phase and out of phase frequencies, respectively.

To study the effect of nonlinear coefficient of the van der Waals (vdW) interaction between two layers of graphene sheet ( $c_3$ ), the backbone curves are shown in Fig. 6 for double layered zigzag graphene sheets embedded in polymer matrix for both nonlinear and linear vdW cases. The values are obtained for in-phase frequencies. It can be seen that the effect of nonlinear vdW pressure is significant for higher amplitude and the nonlinear coefficient of the vdW pressure should be considered for amplitudes more than 0.4. Also, the backbone curves are shown for in-phase and out of phase modes in Figs. 7 and 8, respectively, for various values of  $c_3$ . It can be concluded that the effect of



**Fig. 6.** Backbone curves for DLGSs with linear and nonlinear vdW interaction.

nonlinear vdW coefficient for out of phase mode is higher than that for in-phase mode. Therefore, in order to obtain accurate results, the nonlinear vdW coefficient should be considered especially for out of phase modes. Besides, it can be seen that

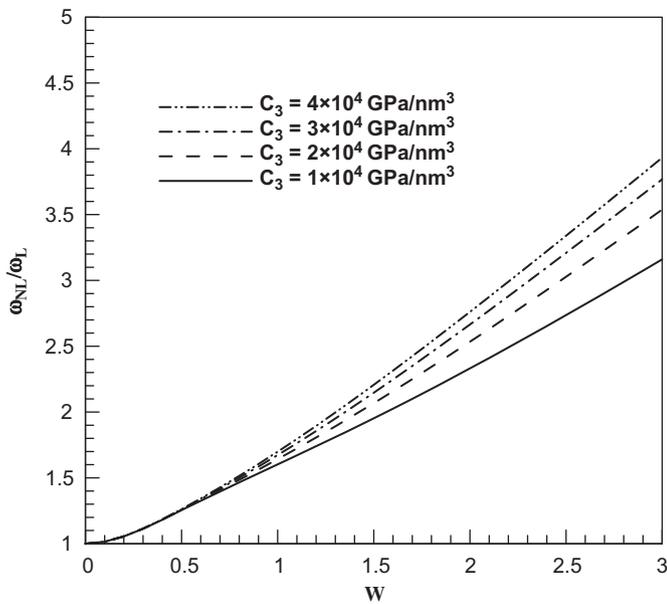


Fig. 7. Effect of nonlinear coefficient of van der Waals pressure on in-phase frequency.

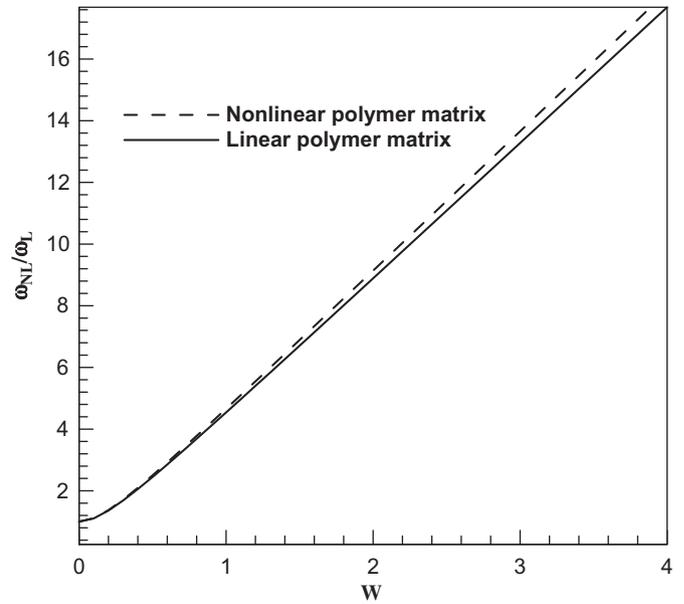


Fig. 9. Backbone curves for out of phase modes of DLGSs with linear and nonlinear polymer interaction.

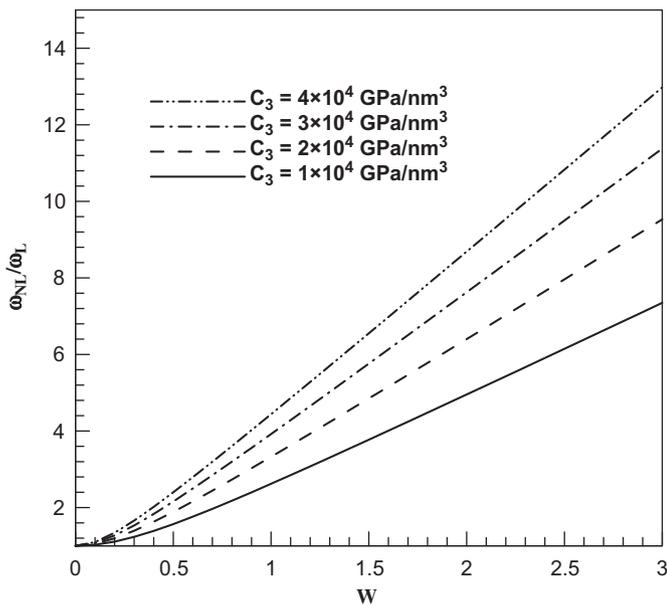


Fig. 8. Effect of nonlinear coefficient of van der Waals pressure on out of phase frequency.

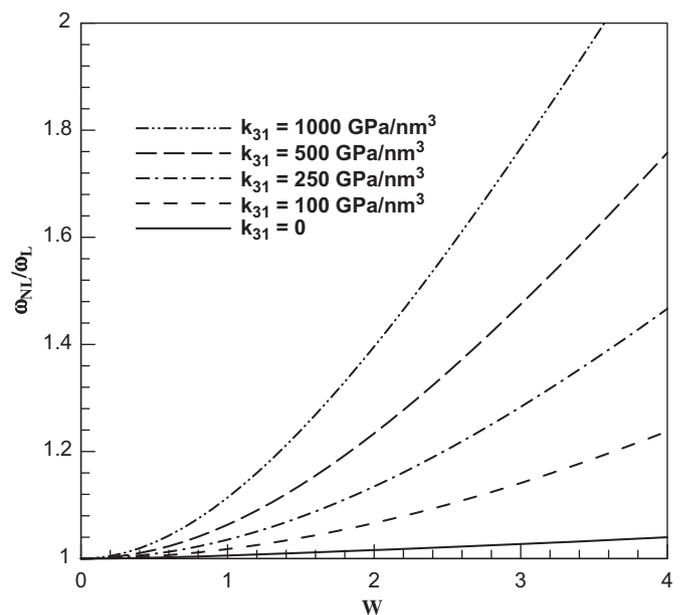


Fig. 10. Effect of nonlinear coefficient of polymer interaction on in-phase frequency.

this coefficient increases the nonlinear frequencies of the embedded DLGS.

To study the effects of nonlinear coefficient of the polymer matrix on natural frequency of double layered graphene sheets embedded in a polymer matrix, the backbone curves are depicted for out of phase natural frequencies in Fig. 9. It can be seen that unlike the vdW interaction, the nonlinear coefficient of polymer matrix has an inconsiderable influence on out of phase frequencies. If the amplitude ratios of two layers ( $W_1/W_2$ ) are obtained for both linear and nonlinear polymer interactions, it can be found that they are very close to each other for out of phase modes (for linear polymer foundation:  $-1.15 < W_1/W_2 < -1$  and for nonlinear polymer foundation:  $-1.16 < W_1/W_2 < -1.04$ ).

Also, the backbone curves of in-phase frequencies are plotted for double layered armchair graphene sheets with different nonlinear polymer coefficients in Fig. 10. It can be seen that the effect

of nonlinear polymer interaction on in-phase frequencies is considerable even for small amplitudes.

Variation of nonlinear frequency of a single layered armchair graphene sheet versus the aspect ratio ( $a/b$ ) is shown in Fig. 11. Although the small scale parameter may depend on the aspect ratio, it has been considered constant ( $e_0l=0.2$  nm) in this figure to study the effect of aspect ratio. It can be seen that nonlinear frequency has a sharp variation for aspect ratios close to unity. This effect is the same for a graphene sheet with linear and nonlinear polymer medium.

For forced vibration results, the response curves (oscillation amplitude of top layer versus the frequency ratio) of a zigzag DLGS without a polymer matrix are shown in Fig. 12 for different values of nonlocal parameters. The value of nondimensional external loads ( $\bar{P} = p_0 a^4 / E_{11} h^4$ ) is assumed to be unit. It can be

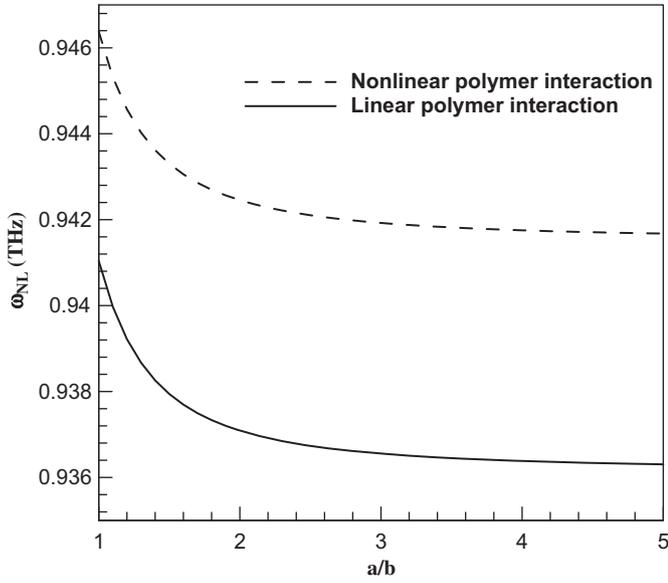


Fig. 11. Nonlinear frequency of a single layered armchair graphene sheet versus the aspect ratio for both linear and nonlinear polymer matrix ( $W_1=0.05$ ).

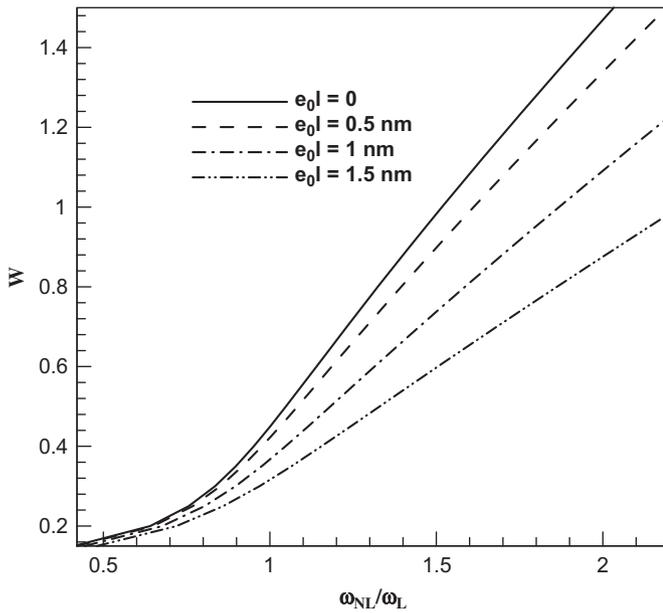


Fig. 12. Response curve of a double layered graphene sheet without the polymer matrix.

seen that the effect of nonlinearity increases as the nonlocal parameter increases. Also, the response curves of a DLGS on a polymer matrix are depicted in Fig. 13 for several values of external loads. It can be concluded that because of the existence of polymer matrix, the changes of vibrational behavior with respect to external force is not considerable.

### 7. Conclusion

The nonlinear free and forced vibration analyses of double layered orthotropic graphene sheets embedded in a polymer medium have been presented. Explicit formulations have been defined for linear and nonlinear coefficients of van der Waals interactions between graphene layers and polymer matrix. Considering the small scale effect, the nonlinear governing equations

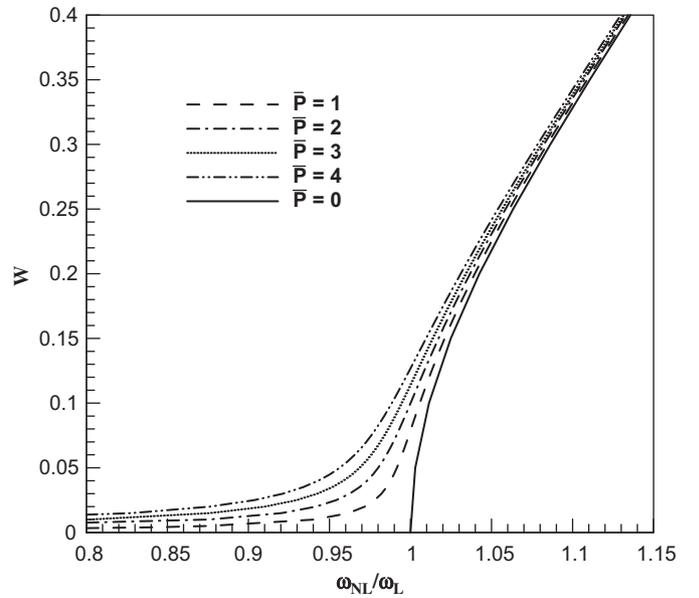


Fig. 13. Response curve of a double layered graphene sheet on a polymer matrix.

of motion have been obtained based on the von Karman hypothesis. The harmonic balance method has been employed to study the large amplitude vibration of a double layered graphene sheet with nonlinear vdW interactions.

It is seen that the nonlinear coefficient of vdW interaction between two layers has significant effect on out of phase nonlinear frequencies while the nonlinear coefficient of polymer matrix has considerable influence on in-phase frequencies. Also, the small scale parameter does not change the nonlinear out of phase frequencies whereas it increases the in-phase nonlinear frequency ratio.

### Acknowledgment

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### Appendix A

The coefficients of the governing equations of motion are obtained in terms of mechanical properties of graphene sheets as

$$\begin{aligned}
 A_{11} &= \frac{Q_{22}}{h(Q_{11}Q_{22}-Q_{12}^2)} & A_{12} &= -\frac{Q_{12}}{h(Q_{11}Q_{22}-Q_{12}^2)} \\
 A_{22} &= \frac{Q_{11}}{h(Q_{11}Q_{22}-Q_{12}^2)} & A_{33} &= \frac{1}{hQ_{33}} \\
 D_{11} &= \frac{h^3Q_{11}}{12} & D_{12} &= \frac{h^3Q_{12}}{12} & D_{22} &= \frac{h^3Q_{22}}{12} & D_{33} &= \frac{h^3Q_{33}}{12} \quad (A.1)
 \end{aligned}$$

where

$$\begin{aligned}
 Q_{11} &= \frac{E_{11}(\cos^4\theta + 2\nu_{21}\sin^2\theta\cos^2\theta) + E_{22}\sin^4\theta}{1-\nu_{12}\nu_{21}} + 4G_{12}\sin^2\theta\cos^2\theta \\
 Q_{12} &= \frac{E_{11}(\sin^2\theta\cos^2\theta + \nu_{21}\sin^4\theta + \nu_{21}\cos^4\theta) + E_{22}\sin^2\theta\cos^2\theta}{1} \\
 &\quad - \nu_{12}\nu_{21} - 4G_{12}\sin^2\theta\cos^2\theta \\
 Q_{22} &= \frac{E_{11}(\sin^4\theta + 2\nu_{21}\sin^2\theta\cos^2\theta) + E_{22}\cos^4\theta}{1-\nu_{12}\nu_{21}} \\
 &\quad + 4G_{12}\sin^2\theta\cos^2\theta
 \end{aligned}$$

$$Q_{33} = \frac{E_{11}(\sin^2 \theta \cos^2 \theta - 2\nu_{21} \sin^2 \theta \cos^2 \theta) + E_{22} \sin^2 \theta \cos^2 \theta}{1 - \nu_{12}\nu_{21}} + G_{12}(\cos^2 \theta - \sin^2 \theta). \quad (\text{A.2})$$

where  $\theta$  denotes the chiral angle (i.e.  $\theta=0$  and  $90^\circ$  represent armchair and zigzag configurations, respectively),  $E_{11}$  and  $E_{22}$  are Young's modulus in the direction and perpendicular of chiral vector, respectively. Also,  $G_{12}$  and  $\nu$  are the shear modulus and Poisson's ratio of the graphene sheet, respectively.

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