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Oscillatory Behavior of Nested Spherical Fullerenes in the Vicinity of a Single Layer Graphene Sheet

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Abstract

This paper deals with the van der Waals (vdW) interactions and oscillatory motion of nested spherical fullerenes, known as carbon onions, in the vicinity of a single layer graphene (SLG) sheet. The carbon onions are of I_h symmetries and the graphene sheet is modeled as a fully constrained flat surface. Utilizing the continuum approximation in conjunction with the 6-12 Lennard-Jones (LJ) potential function, explicit analytical expressions are derived to evaluate the vdW potential energy and interaction force. The equation of motion is solved numerically to attain the time histories of displacement and velocity of the carbon onion. On the basis of conservation of mechanical energy principle, a semi-analytical expression is also derived to accurately evaluate the oscillation frequency. Numerical results are presented to examine the influences of size of carbon onion and initial conditions (initial separation distance and initial velocity) on the operating frequency of carbon onion-SLG sheet oscillators. It is shown that carbon onion performs oscillatory motion above the graphene sheet with frequencies in the gigahertz (GHz) range. It is further observed that smaller structures of carbon onions produce greater frequencies. The presented results in this study would contribute to the development of new generation of nano-oscillators.

Keywords: graphene sheet; carbon onion; continuum approximation; oscillation frequency.

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1. Introduction

Nano-materials such as carbon nanotubes (CNTs), C_{60} fullerene, nanorods and graphene sheet have received considerable attention in recent years. This is mainly due to their unique physical, chemical and mechanical properties that make them as main components for fabricating a wide range of nanoelectromechanical systems (NEMSs) [1]. One of the most interesting applications of nanoscopic materials is the creation of nano-oscillators with frequencies in the gigahertz (GHz)

range or beyond [2]. Such high-frequency nano-oscillators have been proposed for use in ultrafast optical filters, nanoantennae, ultrasensitive mass detection and nonvolatile memory devices [3].

The idea of GHz oscillators was first proposed in the experiment of Cumings and Zettl [4] who observed that if the inner tube of a multi-walled carbon nanotube (MWCNT) is pulled out and released, it quickly and fully retracts into the outer tube. They reported that the van der Waals (vdW) interaction force acting on the extruded core is the main cause of its retraction. Zheng *et al.* [5] then exhibited that the effect of frictional forces on the oscillation frequency of CNT oscillators is almost negligible. Using the molecular dynamics (MD) simulations, Legoas *et al.* [6] and Rivera *et al.* [7] also confirmed the GHz frequency of such systems. It was shown that stable oscillatory motion is achievable when the interwall spacing between the nanotubes is almost 3.4 Å [8]. The MD simulations were also extensively employed to explore the energy dissipation between two contacting parties [9, 10]. For MWCNT oscillators, Guo *et al.* [11] showed that the commensuration and relative morphology of the bitube are the two essential factors which affect the rate of energy dissipation. In addition, Zhao *et al.* [12] using the chemistry at Harvard macromolecular mechanics force field method reported that frictional forces in CNT oscillators change from 10^{-17} to 10^{-14} N per atom for different dissipative mechanisms. The capability of MWCNT oscillators in generating high frequencies stimulated the interest of many researchers to create new configurations of nanoscale oscillators [13-15].

Recently, nano-oscillators based on graphene sheets have attracted a considerable attention from researchers. Graphene sheets were discovered in 2004 and are two-dimensional nanostructures which are considered as one of the allotropes of carbon. These materials are comprised of sp^2-bonded carbon atoms with a molecular bond length of 0.142 nanometers. The current interest in graphene can be attributed to its exceptional properties such as high surface area, ultralow mass density, tuneable band gap, excellent electrical conductivity, extremely high electron mobility and strong mechanical strength [16]. These unique properties make graphene and its related derivatives as promising candidates for a broad range of technological and scientific applications including composite materials, energy storage, nanoelectronics, catalysis, biosensors and drug/gene delivery [17]. Numerous experimental and theoretical studies have demonstrated the feasibility of single-layered graphene (SLG)-based hybrid structures such as fullerene-graphene, CNT-graphene, nano-rod-graphene and nanoparticle-graphene. The graphene-fullerene hybrid systems can be potentially used in photovoltaic devices, solar cells and Li-ion batteries [18]. The vdW interactions which play a key role in such systems have been explored through experiment or computational methods in the open literature. Grimme *et al.* [19] employed density functional theory to determine the noncovalent interactions between graphene sheets and multilayer fullerenes. Employing the MD simulations, Ma *et al.* [20] discovered that the interfacial thermal conductance between C_{60} fullerene and graphene sheet improves by increasing the vdW interactions between them.

This paper aims to investigate the oscillations of nested spherical fullerenes or carbon onions interacting with a SLG sheet. To this end, the continuum approximation and the 6-12 Lennard-Jones (LJ) potential function are used to model the vdW interactions. Neglecting the frictional effects and also thermally-induced motion of the carbon onion molecule, the equation of motion is numerically solved to attain the time histories of displacement and velocity of the core. The conservation of mechanical energy principle is also used to derive a new semi-analytical expression for the precise evaluation of oscillation frequency. Numerical results are presented to study the vdW interactions and dynamic behavior of carbon onion-SLG sheet oscillators under various system parameters.

2. Potential energy and interaction force

The geometry of a carbon onion situated above a fully constrained graphene sheet is presented in Fig. 1. The origin of the Cartesian coordinate system (x, y, z) is assumed to be located at the center of graphene sheet and the perpendicular distance from graphene sheet to the center of carbon

onion molecule is defined by Z . The carbon onion, which is made up of N concentric spherical fullerenes, has n carbon atoms in the kth shell as $n(k)=60k^2$ [21]. Moreover, the mean surface density of graphene sheet and the mean surface density of fullerene of radius R_{F_k} are denoted by η_g and η_{F_k} , respectively.

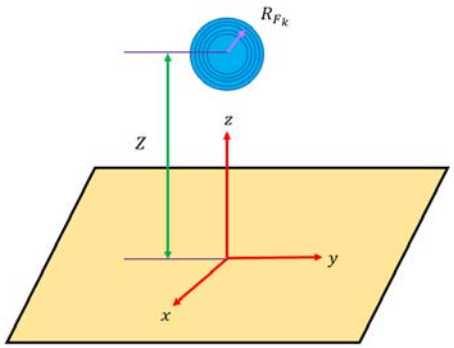

Figure 1. Geometry of carbon onion-SLG sheet oscillators.

Following the continuum approximation in which carbon atoms are assumed to be uniformly distributed over the smeared surfaces of molecules with a constant surface atomic density, the total potential energy between the interacting molecules can be estimated by integrating the LJ potential function over the surfaces of two molecules as

$$E^{tot}=\eta_F\eta_g\int_{\Sigma_G}\int_{\Sigma_F}\left(-\frac{A}{\rho^6}+\frac{B}{\rho^{12}}\right)d\Sigma_F d\Sigma_G \tag{1}$$

in which A and B are the attractive and repulsive constants, respectively and ρ is the distance between two atoms on the molecules.

To determine the total interactions, the interactions between each shell of carbon onion and graphene sheet are first calculated and then the total interactions are obtained through summing the interactions between all layers of carbon onion and graphene sheet. Thus,

$$E^{(tot)}(Z)=\sum_{k=1}^{N}E_k(Z) \tag{2}$$

$$F_z^{(tot)}(Z)=\sum_{k=1}^{N}F_{z_k}(Z) \tag{3}$$

where

$$E_k(Z)=\sum_{n=1}^{2}\sum_{m=3n}^{6n-2}\Gamma_{k,n,m}\frac{1}{\left(Z^2-R_{F_k}^2\right)^{m-1}} \tag{4}$$

$$F_{z_k}(Z)=\sum_{n=1}^{2}\sum_{m=3n}^{6n-2}\lambda_{k,n,m}\frac{Z}{\left(Z^2-R_{F_k}^2\right)^{m}} \tag{5}$$

with parameters as $\Gamma_{k,n,m}=\dfrac{\gamma_k C_n G_k^{(m)}}{m-1}$, $\gamma_k=4\pi^2R_{F_k}^2\eta_{F_k}\eta_g$, $\lambda_{k,n,m}=2(m-1)\Gamma_{k,n,m}$ and

$$\begin{cases} C_1=-A, & C_2=\dfrac{B}{5}, & G_k^{(3)}=1, \\ G_k^{(4)}=2R_{F_k}^2, & G_k^{(6)}=5, & G_k^{(7)}=80R_{F_k}^2, \\ G_k^{(8)}=336R_{F_k}^4, & G_k^{(9)}=512R_{F_k}^6, & G_k^{(10)}=256R_{F_k}^8 \end{cases} \tag{6}$$

3. Equation of motion and the frequency of oscillation

Using the Newton's second law, the equation of motion can be written as

$$m_F \frac{d^2 Z}{dt^2} = F_z^{(tot)}(Z) \tag{7}$$

where $m_F = m_0 \sum_{k=1}^{N} n(k)$ is the total mass of carbon onion molecule and m_0 is the mass of a single carbon atom. The oscillation frequency can be obtained through solving Eq. (7) numerically.

Alternatively, the oscillation frequency of system can be obtained from the energy equation. Since the frictional force can be ignored, the total potential energy is conserved. Thus, conservation of mechanical energy yields

$$\frac{1}{2} m_F \left(\frac{dZ}{dt}\right)^2 + E^{(tot)}(Z) = E^{(tot)}(A_0) \rightarrow \sqrt{\frac{m_F}{2}} \int_{2Z^*-A_0}^{A_0} \frac{dZ}{\sqrt{E^{(tot)}(A_0) - E^{(tot)}(Z)}} = \int_0^{\frac{T}{2}} dt \tag{8}$$

in which Z^* is the equilibrium distance at which total potential energy is minimized, A_0 is the amplitude of motion and T is the period of motion which can be determined from

$$T = \sqrt{2m_F} \int_{2Z^*-A_0}^{A_0} \frac{dZ}{\sqrt{E^{(tot)}(A_0) - E^{(tot)}(Z)}} \tag{9}$$

The period of motion can be obtained by summing T_1 and T_2 as

$$T_1 = \sqrt{2m_F} \int_{2Z^*-A_0}^{\delta A_0} \frac{dZ}{\sqrt{E^{(tot)}(A_0) - E^{(tot)}(Z)}}, \quad T_2 = \sqrt{2m_F} \int_{\delta A_0}^{A_0} \frac{dZ}{\sqrt{E^{(tot)}(A_0) - E^{(tot)}(Z)}}, \quad \delta = 0.99 \tag{10}$$

To remove the singularity from T_2, one can write

$$E^{(tot)}(A_0) - E^{(tot)}(Z) = (A_0 - Z) R(A_0, Z), R(A_0, Z)\big|_{Z=A_0} \neq 0 \tag{11}$$

Performing extensive mathematical manipulations, function $R(A_0, Z)$ can be derived as

$$R(A_0, Z) = -\sum_{k=1}^{N} \sum_{n=1}^{2} \sum_{m=3n}^{6n-2} \sum_{l=1}^{m-1} \Gamma_{k,n,m} \left(\frac{A_0 + Z}{\left(A_0^2 - R_{F_k}^2\right)^{m-l} \left(Z^2 - R_{F_k}^2\right)^{l}} \right) \tag{12}$$

Substituting Eq. (12) into Eq. (11) and letting $Z = A_0 \sin^2\phi$, T_2 is obtained as

$$T_2 = 2\sqrt{2m_F A_0} \int_{\sin^{-1}\sqrt{\delta}}^{\pi/2} \frac{\sin\phi d\phi}{\sqrt{R\left(A_0, A_0 \sin^2\phi\right)}} \tag{13}$$

Note that the oscillation frequency is the reciprocal of amplitude of motion.

4. Numerical results

Based on the proposed formulations, numerical results are presented in this section for the vdW interactions and oscillation frequency of carbon onion-SLG sheet oscillators. The influences of different system parameters such as size of carbon onion and initial conditions on the dynamic behavior of such oscillators are fully examined. For the system under consideration, the values of constant parameters required for numerical evaluations are listed in Table 1.

Table 1. Numerical values of the constant parameters used in the model [21].

Radius of C_{60}	3.55 Å
Radius of C_{240}	7.12 Å
Radius of C_{540}	10.5 Å
Radius of C_{960}	13.8 Å
Radius of C_{1500}	17.5225 Å
Mean surface density of C_{60}	0.3789 $Å^{-2}$
Mean surface density of C_{240}	0.3767 $Å^{-2}$
Mean surface density of C_{540}	0.3898 $Å^{-2}$
Mean surface density of C_{960}	0.4011 $Å^{-2}$
Mean surface density of C_{1500}	0.3888 $Å^{-2}$
Mean surface density of graphene sheet	0.3812 $Å^{-2}$
Attractive constant	17.4 $eVÅ^{6}$
Repulsive constant	29000 $eVÅ^{12}$
Mass of a single carbon atom	1.993×10^{-26} kg

The vdW potential energy versus separation distance is plotted in Fig. 2 for different sizes of carbon onion. As seen, the global shape of total potential energy is not affected by the number of layers of carbon onion molecule.

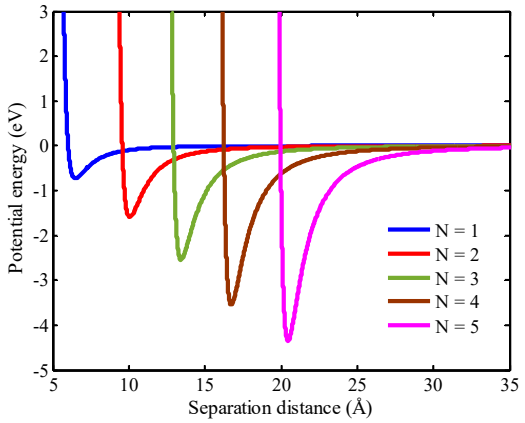

Figure 2. Potential energy profile for different carbon onions interacting with the graphene sheet.

Based on the numerical results, the values of equilibrium distance along with the minimum potential energy are tabulated in Table 2. It can be seen that as the carbon onion gets larger, the equilibrium distance moves further away from the graphene sheet and the magnitude of minimum potential energy increases.

Table 2. Equilibrium distance and minimum potential energy for different sizes of carbon onion.

N	1	2	3	4	5
Z^* (Å)	6.5093	10.0664	13.4401	16.7361	20.4607
E^* (eV)	-0.7290	-1.5936	-2.5473	-3.5457	-4.3517

The effect of initial height of carbon onion above the graphene sheet on the time histories of vdW force, separation distance and velocity along with the phase plot is examined in Figs. 3 (a) to 3 (d), respectively. In this figure, it is assumed that the carbon onion molecule has three shells and is initially at rest. As depicted for all cases, the carbon onion oscillates with respect to the equilibrium distance. The amplitude of motion, velocity and period of motion are shown to be very sensitive to the initial distance. When the initial height shifts towards the equilibrium distance, the amplitude of

motion and the maximum velocity decrease, while the operating frequency increases considerably. In addition, the phase plots are not circular. This is attributed to the quicker growth of velocity compared to the displacement which leads to the egg-shaped curves for all cases.

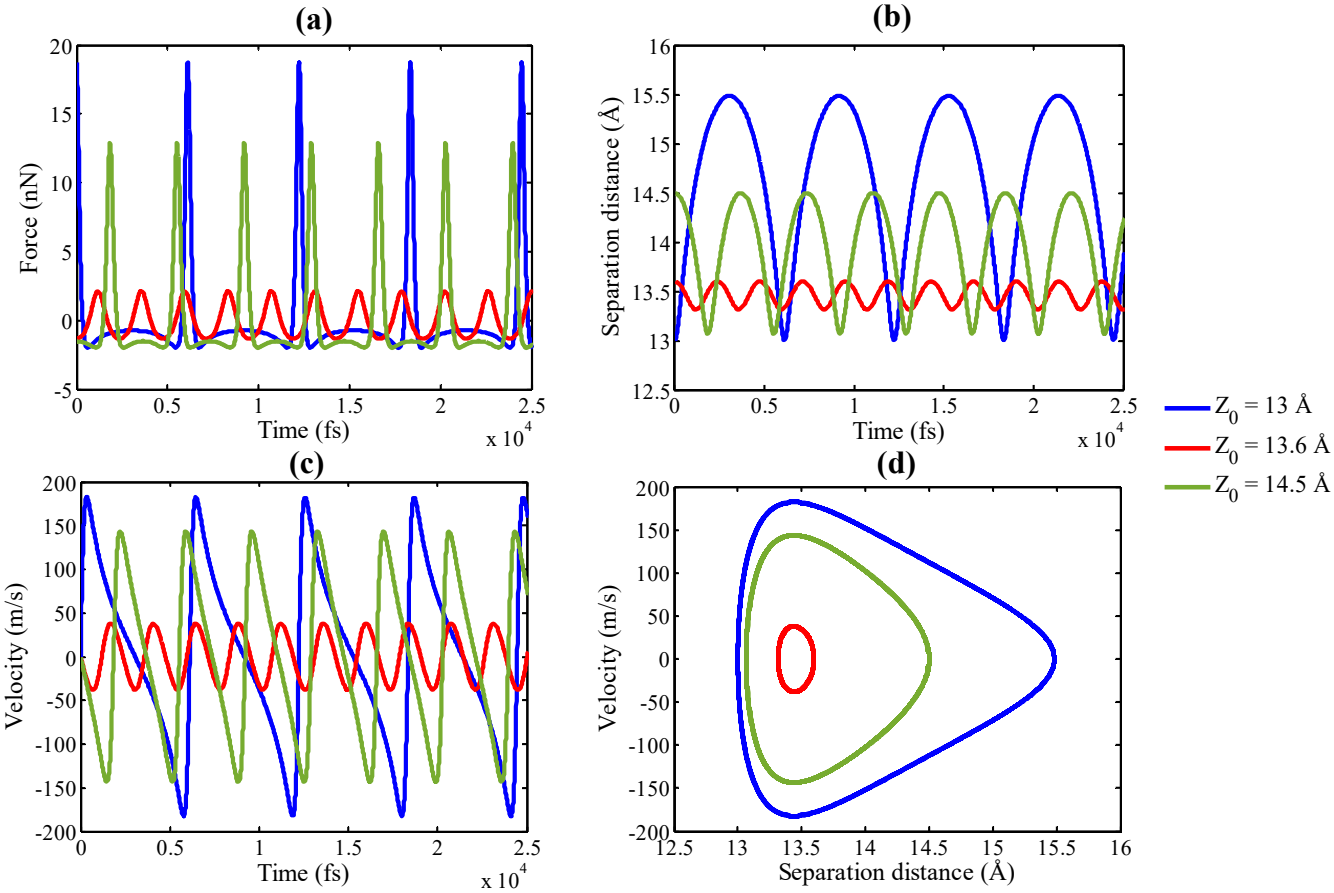

Figure 3. (a) Time history of vdW force **(b)** time history of separation distance **(c)** time history of velocity and **(d)** phase plot for different initial separation distances.

The variation of oscillation frequency obtained from equation of motion and energy equation with the normalized amplitude (the ratio of amplitude to the equilibrium distance) is shown in Fig. 4. Here, the number of shells is three and it is assumed that the carbon onion is initially separated from the graphene sheet and is at rest. As seen, the accuracy of the oscillation frequency formula is verified. In addition, frequency behaves decreasingly as the amplitude of motion increases. Hence, the maximum frequency is obtained when the normalized amplitude is almost one.

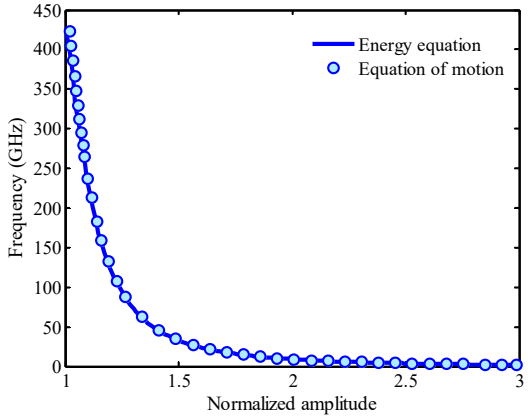

Figure 4. Comparison of oscillation frequency obtained from equation of motion and energy equation.

The effect of size of carbon onion on the variations of oscillation frequency with normalized amplitude and initial velocity is examined in Figs. 5 (a) and 5 (b), respectively. As depicted, the behavior of frequency with both initial separation distance and initial velocity is decreasing.

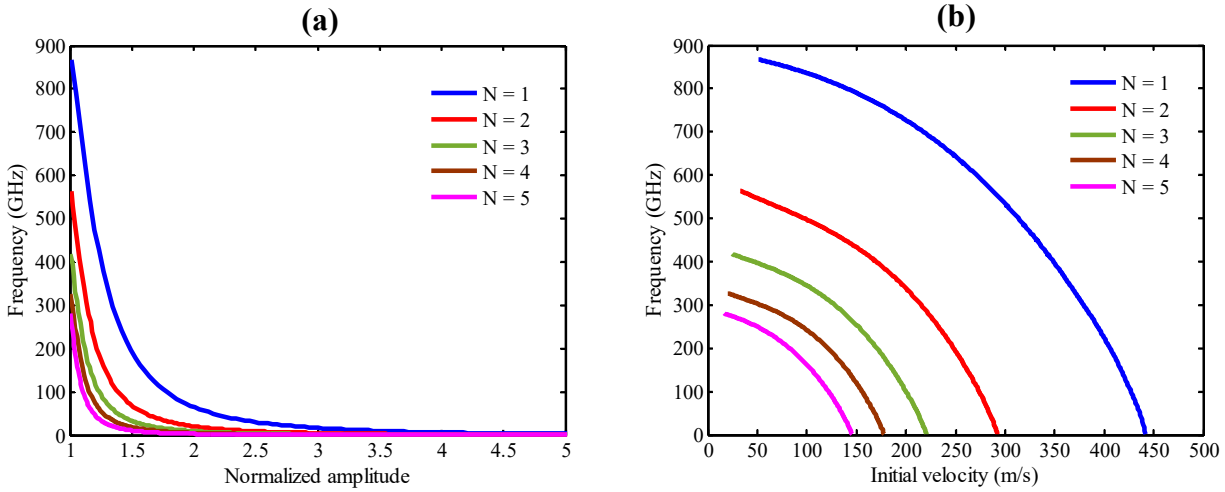

Figure. 5. Oscillation frequency in terms of **(a)** normalized amplitude **(b)** initial velocity for various carbon onions.

The values of maximum frequency associated with different carbon onions are listed in Table 3. It can be observed that heavier carbon onion molecules provide lower maximum frequencies.

Table 3. Maximum frequency for different sizes of carbon onion.

N	1	2	3	4	5
$f_{\max}$ (GHz)	867.8	563.9	417.3	326.9	280.4

5. Conclusion

In this paper, the dynamic behavior of carbon onions near a fully constrained SLG sheet was explored. A continuum model based on the continuum approximation and the 6-12 LJ potential function was constructed to determine the vdW interactions analytically. The equilibrium distance related to this hybrid-nanostructure was predicted by minimizing the total potential energy and it was found that this distance moves further away from the sheet as the carbon onion gets heavier. The magnitude of minimum potential energy was also shown to increase by enlarging the carbon onion. Using the vdW force distribution, the time histories of separation distance and velocity were attained through solving the equation of motion numerically. It was found that amplitude, velocity and period of motion are very sensitive to initial conditions. In addition, oscillation frequencies were accurately determined using a semi-analytical expression derived from the energy equation. It was demonstrated that the operating frequencies are in the GHz range and can be controlled by the size of carbon onion and initial conditions. Numerical results revealed that smaller carbon onion molecules generate higher frequencies. It was also observed that oscillation frequency decreases monotonically as the amplitude of motion or the initial velocity increases.

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