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OSCILLATORY BEHAVIOUR AND NUMERICAL SIMULATION OF A C_{60} FULLERENCE IN SINGLE-WALLED CARBON NANOTUBES

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Abstract

Carbon nanotubes play an important role in nanotechnology worldwide because of their physical and mechanical properties. Their applications can be found in drug delivery, nano-bearings and nano-oscillators. In this paper, we study the oscillatory behaviour of a C_{60} fullerence inside single-walled carbon nanotubes (SWCNTs) by using the finite element method based on the Arbitrary Lagrangian Eulerian (ALE) approach. We apply the continuum assumption and the Lennard-Jones potential for non-bonded interaction potential energy between two molecules. The movement of the C_{60} fullerene described by Newton law of the motion associated with the van der Waals force and a frictional force, is investigated. It is assumed that the C_{60} fullerene is initially set outside the nanotubes.

Key words: Lennard-Jones potential, single-walled carbon nanotubes, C_{60} fullerene, Arbitrary Lagrangian Eulerian.

1 Introduction

Nanotechnology is one of the most important research and development frontiers in modern science. Carbon nanotubes (CNTs) are one of the most interesting branches of nanotechnology, which can be applied to benefit many fields such as drug delivery, therapy techniques, diagnostic and imaging techniques, anti-microbial techniques, cell repair, nano-bearings and nano-oscillators. Iijima [6] discovered the carbon nanotubes in 1991, which are tubular structures like a sheet of graphite rolled up into a cylindrical tube, with high aspect ratio. Iijima *et al.* [7] and Bethune *et al.* [1] discovered the simplest kind of carbon nanotube in 1993. The applications of nanotechnology in the field of health care can be described by Khan [8] consist of disease diagnosis, prevention and treatment of disease, more efficient drug delivery system with minimal side effects and tissue reconstruction. Surendiran *et al.* [11] improved methods of cancer detection based on nanoparticles which can be used as contrast agents, fluorescent materials, molecular research tools and drugs with targeting antibodies. Girifalco *et al.* [5, 4] used the Lennard-Jones potential energy function to determine the universal graphitic system. Zheng and Jiang [13] presented a gigahertz oscillator of multi-walled carbon nanotubes with different core lengths. To analyze the frequency of the core oscillation, they used Newton's second law which is given by

$$M\ddot{x}(t) = F_{vdW} + F_r, \tag{1}$$

where M is the total mass of the core, F_{vdW} and F_r are the van der Waals force and total intershell sliding resistance force, respectively. Zheng *et al.* [14] applied the Lennard-Jones potential energy function to determine the energy for multi-walled carbon nanotubes and proposed gigahertz frequency oscillators.

The objective of this paper is to study the oscillatory behaviour of a C_{60} fullerence inside single-walled carbon nanotubes by using the finite element method based on the Arbitrary Lagrangian Eulerian (ALE) approach. The C_{60} fullerence is chosen in this study because it has unique properties which have draw the interest of many researchers. The governing equation of the C_{60} fullerence motion includes Newton's law of the motion associated with the van der Waals force derived from Lennard-Jones potential [2] and a frictional force.

2 Mathematical Model

To study the oscillatory behaviour of a C_{60} fullerene inside a carbon nanotube, we assume that the system consisting of the nanotube and the nanoparticle occupies a bounded $\bar{\Omega}$ in $\mathbf{R}^2$. At a typical instant of time t, the nanoparticle occupies a closed connected subset $\Omega_q \subset \mathbf{R}^2$. The computational domain is then given by $\bar{\Omega} - \Omega_q$ which is called the area of particle motion. In this study,

we use two coordinate systems: a reference system Ω where the model is drawn and the particle movement is solved, and a moving mesh system Ω_{def} corresponding to the deformed mesh where we simulate the particle motion inside the nanotube. The time evolution of the domain Ω_{def} is determined by means of an Arbitrary Lagrangian Eulerian (ALE) [3] mapping $\mathbf{x} : \Omega \times \mathbf{R}^+ \mapsto \Omega_{def}$ which maps any point $(\mathbf{X}, t)$ in Ω to its image $\mathbf{x}(\mathbf{X}, t)$ in Ω_{def}.

2.1 Arbitrary Lagrangian Eulerian Approach

In this section, we introduce the numerical techniques of continuum mechanics which can be classified into the Lagrangian and the Eulerian methods. In the Eulerian approach, the computational grid is fixed in time. For dynamical systems, the pure Eulerian approach is not suitable for simulations because it is unable to treat moving boundaries. To study fullerene motion inside carbon nanotubes, there exists a mass movement in computational domains, representing the particle movement through the grid. In the Lagrangian approach, the computational grid moves with the particle. However, it can lead to severe grid distortions due to, for example, a shear in velocity, so the mathematical assumptions of the numerical method are violated. To resolve these problems, the Arbitrary Lagrangian Eulerian (ALE) method [3] is used. In the ALE method, several steps of pure Lagrangian computation are performed. Then, a mesh rezoning technique is used to keep the computational grid smooth and convex during the whole computation as shown in Figure 1.

To construct a mesh-based numerical model involving the motion of particles motion, we will introduce the Arbitrary Lagrangian-Eulerian method which will involve the mesh movement of the numerical model. Due to the movement of the coordinate system, the mesh velocity $\mathbf{\Psi} = (\Psi_x, \Psi_z)$ is introduced in the deformed domain Ω_{def}. To guarantee a smoothly varying distribution of the nodes, we know that the nodes on $\partial\Omega_q$ move with the particle and that each component of the mesh velocity in the area of particle motion is governed by a Laplace equation [15]:

$$\nabla^2 \mathbf{\Psi} = \mathbf{0}, \quad \forall \mathbf{x} \in \Omega_{def}. \tag{2}$$

From equation (2), it is to smooth gradient of the mesh velocity over the domain so as to reduce mesh distortion. Once the mesh velocity components are determined, we can determine the smoothed deformed mesh for the area of particle motion at each time instant by updating the coordinates of the nodes according to the following formulae

$$\begin{array}{rcl} x & = & X + \int_0^t \Psi_x \, dt, \\ z & = & Z + \int_0^t \Psi_z \, dt. \end{array} \tag{3}$$

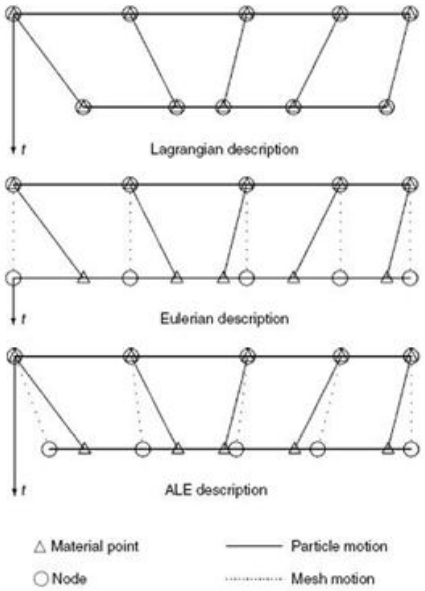

Figure 1: One-dimensional example of Lagrangian, Eulerian and ALE mesh and particle motion [3].

Another condition that needs to be specified is that the particle and mesh all move with the same velocity on the particle boundaries, i.e.,

$$\mathbf{\Psi} = \mathbf{V} \quad \text{on } \partial\Omega_q. \tag{4}$$

The detail of how to determine the mesh velocity,$\mathbf{\Psi}$, using the finite element method can be found in [12].

2.2 Lennard-Jones Potential Function

In the continuum approach, it is assumed that carbon atoms are uniformly distributed over the surface of the molecules and the non-bonded in tradition energy is given by

$$E = n_g n_f \int_{\Sigma_g} \int_{\Sigma_f} \Phi(r) d\Sigma_f d\Sigma_g, \tag{5}$$

where n_g and n_f are the mean surface density of carbon atom on a carbon nanotube and a C_{60} fullerene, respectively, and r denotes the distance between two typical surface elements $d\Sigma_f$ and $d\Sigma_g$ on each molecule.

The Lennard-Jones potential energy which is a function used to describe the interaction between a pair of neutral atoms or molecules [9] is given by

$$\Phi(r) = -Ar^{-6} + Br^{-12}, \tag{6}$$

where A and B are the attractive and the repulsive constants, respectively. By performing an integral of the classical Lennard-Jones potential over the C_{60} fullerene, the potential energy is in the form

$$P(\rho) = -Q_6(\rho) + Q_{12}(\rho), \tag{7}$$

where ρ is the distance between an atom and the wall of the carbon nanotube and the determination of Q_n is detailed in [2, 10] with coefficients $C_6 = A$ and $C_{12} = B$. Then we have

$$P(\rho) = \frac{n_f \pi b}{\rho} \left\{ \frac{A}{2} \left(\frac{1}{(\rho+b)^4} - \frac{1}{(\rho-b)^4} \right) - \frac{B}{5} \left(\frac{1}{(\rho+b)^{10}} - \frac{1}{(\rho-b)^{10}} \right) \right\}, \tag{8}$$

where b is the radius of C_{60} fullerene. The geometry of a C_{60} fullerene oscillation in a single-walled carbon nanotube is shown in Figure 2.

The van der Waals interaction force between the fullerene molecule and the carbon nanotube is given by

$$\mathbf{F}_{vdW} = -\nabla P. \tag{9}$$

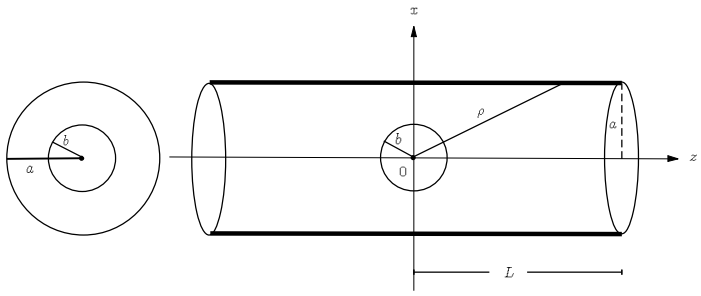

Figure 2: A Geometry of a C_{60} fullerene oscillation in a single-walled carbon nanotube.

Assuming the forces are in the axial direction, the van der Waals force is given by

$$F_Z = -\frac{(Z-z)}{\rho}\frac{dP}{dZ}. \tag{10}$$

Thus, the total axial force between the C_{60} molecule and the entire carbon nanotube is obtained by performing the surface integral of equation (9) over the carbon nanotube, therefore we may deduce

$$F_z^{tot}(Z) = -2a\pi\eta_g \int_0^\infty -\frac{(Z-z)}{\rho}\frac{dP}{dZ}\,dz, \tag{11}$$

and letting $\rho^2 = a^2 + (Z-z)^2$, we have $d\rho = -[(Z-z)/\rho]dz$. Thus, equation (11) becomes

$$\begin{aligned} F_z^{tot}(Z) &= -2a\pi\eta_g \int_{\sqrt{a^2+z^2}}^\infty \frac{dP}{dZ}\,dz \\ &= -2ab^2\pi^2\eta_g\eta_f \left\{ \frac{A}{2b\rho}\left(\frac{1}{(\rho+b)^4} - \frac{1}{(\rho-b)^4} \right) \right. \\ &\quad \left. -\frac{B}{5b\rho}\left(\frac{1}{(\rho+b)^{10}} - \frac{1}{(\rho-b)^{10}} \right) \right\}_{\rho=\sqrt{a^2+z^2}}. \end{aligned} \tag{12}$$

Using the partial fractions in the terms of $(\rho^2 - b^2)$ and we rewrite equation (12) as follows

$$F_z^{tot}(Z) = \frac{8a\eta_f\eta_g\pi^2}{b^4\lambda^3}\left\{ A\left(1+\frac{2}{\lambda}\right) - \frac{B}{5b^6\lambda^3}\left(5+\frac{80}{\lambda}+\frac{336}{\lambda^2}+\frac{512}{\lambda^3}+\frac{256}{\lambda^4}\right)\right\}, \tag{13}$$

where $\lambda = (a^2 - b^2 + Z^2/)b^2$.

2.3 Movement of a C_{60} fullerene in the Deformed Mesh System

In this section, the mathematical model for the particle motion inside a nanotube is presented. The governing equation for particle motion is Newton's second law of motion. It is assumed that a carbon nanotube is of length $2L = 129$Å of radius a and the C_{60} fullerene is set to move along the z-axis to avoid the rocking the motion. Now consider the mathematical model for particle motion. We assume that the gravitational force can be neglected and the particle motion is governed by Newton's second law:

$$\begin{aligned} m\frac{d\mathbf{V}}{dt} &= \mathbf{F}^{tot} - \mathbf{F}_r, \\ \mathbf{V}(t_0) &= \mathbf{0}, \end{aligned} \tag{14}$$

where m is the total mass of the nanoparticle, $\mathbf{V}$ is the velocity of the particle, $\mathbf{F}^{tot}$ is the total van der Waals interaction force between the nanoparticle and the carbon nanotube and $\mathbf{F}_r$ is a friction force. The position $\mathbf{X}$ of the center of the nanoparticle can be defined as

$$\begin{aligned} m\frac{d\mathbf{X}}{dt} &= \mathbf{V}, \\ \mathbf{X}(t_0) &= \mathbf{X}_0. \end{aligned} \tag{15}$$

As a preliminary study, we are only concerned with the forces in an axial direction. Therefore, the total axial van der Waals interaction force between the nanoparticle and the carbon nanotube, $\mathbf{F}_z^{tot}$, is given by equation(13).

The frictional force $F_r(Z)$ is a periodic inter-atomic locking force defined by [14]

$$F_r(Z) = \kappa_0 \sin(\frac{2\pi Z}{\ell}), \tag{16}$$

where $\ell = \sqrt{3}\sigma$ is the spatial period of the inter-atomic locking, σ is the carbon - carbon bond length. Typically for an armchair carbon nanotube, $\kappa_0 = \tau_s\alpha$, where τ_s is the resistance strength and α is the area of contact ring of certain prescribed length of the sphere, given by $\alpha = 4\pi b^2 \sin(\theta_0/2)$, for a certain angle θ_0.

3 Numerical Results and Discussions

In this section, we present the numerical results of the movement of a C_{60} fullerene inside the carbon nanotubes of different radii which are (11,11), (12,12) and (13,13). Their radii and all other constants used in calculations

throughout this study are given in Table 1. In our simulations, the C_{60} fullerene is initially set outside the nanotubes at $Z(t_0) = -80$Å. The finite element method based on the Arbitrary Lagrangian Eulerian approach is used to simulate the oscillatory behaviour of the nanoparticle in the nanotubes and we set $\triangle t = 1 \times 10^{-18}$s or 1 attosecond. Note that the finite element mesh changes as the nanoparticle moves down the tube. In Figure 10, we plot the z components of the total force acting on the C_{60} fullerene for the three carbon nanotubes: (11,11), (12,12), (13,13) of radius a=7.463, a=8.141, and a=8.820$\mathring{A}$, respectively. For the three cases of the offset distance, we assume that the potential energy is negatively minimum. For the (11,11) nanotube, the total force near both open ends of the tube are attractive forces. After the encapsulation, the particle moves very fast inside the nanotube at constant speeds $v = 798$ m/s as shown in Figure **??**, yielding an oscillatory motion with a gigahertz frequency $f = 30.93$ GHz. For the (12,12) and (13,13) nanotubes, the similar oscillatory motion is found with $v = 638, 513$ m/s and $f = 24.73, 19.88$ GHz, respectively.

Note that the offset distance is set for the minimum energy of the (11,11) nanotube, (12,12) nanotube, and (13,13) nanotube. It is seen that the C_{60} fullerene will move faster inside the the (11,11) nanotube, compared to the larger (12,12) and (13,13) nanotubes. Comparing the nanoparticle inside the (11,11) nanotube, it shows that the nanoparticle located at minimun energy generates a higher total force, and higher frequency than for the other two cases. It is noted that the suction of the nanoparticle into the nanotubes depends on the size of the nanoparticle and the radius of nanotubes.

Table 1: Parameters used in our simulation.

Parameter	*Value*
radius of (11,11) (Å)	$a = 7.463$
radius of (12,12) (Å)	$a = 8.141$
radius of (13,13) (Å)	$a = 8.820$
radius of C_{60} fullerene (Å)	$b = 3.55$
carbon - carbon bond length (Å)	$\sigma = 1.421$
mean surface density-graphen $[4\sqrt{3}/(9\sigma^2)](\text{Å}^{-2})$	$n_g = 0.3812$
mean surface density-fullerene $[60/(4\pi b^2)](\text{Å}^{-2})$	$n_f = 0.3789$
mass of the nanoparticles (kg)	$m = 1.196\times10^{-24}$
length of carbon nanotube (Å)	$2L = 129$
attractive constant (eV×Å^6)	$A = 17.4$
repulsive constant (eV×Å^{12})	$B = 29\times10^3$
spatial period of the interatomic locking (Å)	$\ell = \sqrt{3}\sigma$
sliding resistance strength (MPa)	$\tau_s = 0.48$

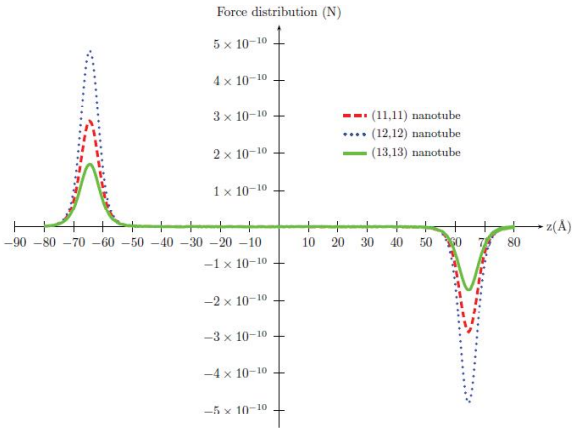

Figure 3: The numerical results of the z component of the total force acting on the C_{60} fullerene for the three carbon nanotubes: (11,11), (12,12), (13,13). The ends of the tube are at $z = -64.5$ and $z = 64.5$Å.

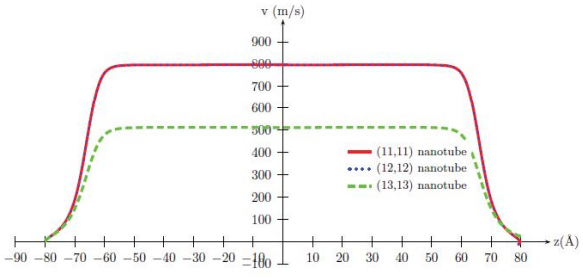

Figure 4: The numerical results of the z component of the velocity of the C_{60} fullerene for the three carbon nanotubes: (11,11), (12,12), (13,13). The ends of the tube are at $z = -64.5$ and $z = 64.5$Å.

4 Conclusions

In this paper, we have studied the oscillatory behaviour of a C_{60} fullerene inside single-walled carbon nanotubes of three different sizes: (11,11), (12,12) and (13,13), with radii of 7.463Å, 8.141Å and 8.820Å, respectively. A finite element method, based on the Arbitrary Lagrangian Eulerian approach, is used in simulation of the nanoparticle motion. The Lennards-Jones potential is used to calculate the van der Waal force. The nanoparticles are initially set at rest outside the carbon nanotubes and the potential energy is negative. The results show that the nanoparticles enter into the tubes if the potential energy is negative. After the encapsulation, the nanoparticles oscillate inside the tube with highest frequency when the nanoparticles are set at the offset distances of minimum energy for the (11,11), (12,12) and (13,13) nanotubes. From our numerical study, it might be able to predict the oscillation of a nanoparticle in a carbon nanotube, which will become an important issue for a applications such as drug delivery and nano-oscillator devices.

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