How can Encapsulated C_{60} Fullerenes Escape from a Carbon Nanotube? A Molecular Dynamics Simulation Answer

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This work aim is to determine how a C_{60} fullerene, encapsulated into a (10,10) carbon nanotube, can be ballistically expelled from it by using a colliding capsule. Initially, the C_{60} fullerene is positioned at rest inside the nanotube. The capsule, also starting from rest but outside of the nanotube, is put in a position such that it can be trapped towards the interior of the nanotube by attraction forces between their atoms. The energy gain associated to the capsule penetration is kinetic energy, giving rise to a high velocity for it. When the capsule reaches the C_{60} fullerene, it transfers energy to it in an amount that enables the fullerene to escape from the nanotube. The mechanical behavior was simulated by classical molecular dynamics. The intermolecular interactions are described by a van der Waals potential while the intramolecular interactions are described by an empirical Tersoff-Brenner potential for the carbon system.

Keywords: Carbon nanotubes; Fullerenes; Molecular dynamics

I. INTRODUCTION

Since the discovery of new carbon allotropic varieties, other than graphite and diamond [1, 2], a completely new and still increasingly area of research has been developed. A great amount of experimental effort has been put for controlling the production of these new allotropic carbon varieties, in pace with the design and tests of theoretical models and corresponding computational work. This activity, designed to interpret experimental data and giving hints for additional experimental research, has given space even for suggestions of prospective and heuristically conceived materials [3–7], with several possible characteristics being inferred from calculations of theoretical models. The controlled production of individual sheets of graphene has also added curiosity and additional work, either theoretical as experimental [8, 9]. Recently, a great theoretical and experimental research effort has been carried out in order to investigate the encapsulation processes and oscillatory mechanisms of C_{60} and C_{20} fullerenes into nanotubes [10–14]. Zig-zag and helical configurations of nanotubes filled with C_{60} fullerenes were first theoretically predicted [15, 16] and followed by experimental observation [17].

In this paper it is presented simulations of the entrance/encapsulation and oscillation processes of a capsule into a single walled armchair (10,10) carbon nanotube with a C_{60} fullerene being expelled from the interior of the nanotube as a result of collision with the penetrating capsule.

II. METHODOLOGY

Classical molecular dynamics calculations are used to simulate the interaction mechanism involving the three considered subsystems: nanotube, capsule and fullerene. This is done in two levels: internal, i. e. among atoms of each subsystem and external, i. e. involving atoms of different subsystems. In doing this, it is explored the entrance/encapsulation and subsequent oscillatory mechanism of the capsule within the nanotube, determination of the equilibrium position of the fullerene inside the nanotube and also the mechanism of a previously encapsulated C_{60} fullerene being ejected from the single walled carbon nanotube as a result of a collision with the penetrating capsule.

The position, velocity and acceleration of both, fullerene and capsule, were calculated using the Nordsieck fourth order method [18]. The *intramolecular* interaction, i. e. among atoms constituting each subsystem, is described by using the reactive empirical bond-order potential (REBO), as developed by Brenner to study carbon and hydrocarbon systems [19]. This model uses the Tersoff-style [20] potential to describe the covalent bonding interaction between carbon atoms. The *intermolecular* interaction, i. e. among atoms of the capsule and the ones of the nanotube, among the fullerene atoms and the ones of the nanotube and, similarly, among those from capsule and the ones of the fullerene, was described by the well known Lennard-Jones (12-6) potential (LJ). This potential includes dispersion and short-range repulsion effects that are responsible for many of the carbon properties. Here it was adopted, for the LJ parameters, the values used by Kang et al. [21], i. e. ε equal to 2.635×10^{-3} eV and σ being 3.369 Å. To switch off the LJ interaction, three different criteria were chosen: i) the value of the separating distance between the pair atoms in question (two carbon atoms interact directly only if they are separated by a distance less than 2.0 Å); ii) the strength of any bonding interaction between them, and iii) the network of bonds connecting them. This new potential model is the adaptive intermolecular REBO potential (AIREBO), as developed by Stuart et al. [22].

III. CALCULATIONS AND RESULTS

A. The nanotube and an oscillatory capsule.

The calculations were performed considering intra and intermolecular interactions for each two units. The first one is a

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capsule formed by a part of a (5x5) nanotude with 150 atoms and a radius of 3.39 Å, linked at the ends to two halves of a C_{60} fullerene, each one with 30 atoms behaving as a "closing" device for the nanotube. This corresponds to a total of 210 carbon atoms. The nanotube, inside of which the capsule is going to be inserted by intermolecular attraction, is a (10x10) nanotube with 3,200 atoms and a radius of 6.78 Å. This nanotube consists of 80 cells, each one with 40 atoms, corresponding to a total length of 195.97 Å. The difference in radius between the capsule and the nanotube is 3.39 Å. Legoas et al. [23, 24] have already showed that this is the optimum radii difference to make nano-oscillators dynamically stable. Initially, the capsule is abandoned near the nanotube entrance cavity from rest, with nanotube and capsule symmetry axis aligned each other. The initial position of the capsule was chosen such that it can be attracted from the nanotube and subsequently be encapsulated into the nanotube. Fig. 1a shows the initial configuration of the system. The initial distance between capsule and nanotube was chosen so that the initial attracting force is very faint. This procedure was adopted in order to maximize the trapping/penetration energy. Figs. 1b and c indicate, respectively, intermediary and extremal positions of the capsule in its oscillatory movement.

Figure 2 shows the center of mass (CM) position and the CM velocity of the capsule as a function of time. In the left axis it is shown the CM position (black line) in units of Angstrom (Å) while in the right axis it is represented the CM velocity (grey line, red in color) in units of Å/fs (1 fs corresponds to 10^{-15} s and 1 Å/fs corresponds to 10^5 m/s). It is possible to observe in Fig. 2 that the capsule acquires a velocity of 250 m/s when it is totally inside of the nanotube (see Fig. 1b). When the capsule reaches the opposite nanotube end, its velocity vanishes (see Fig. 1c) and the capsule returns to the nanotube interior, forming then a nano-oscillator. In Fig. 3 it is shown the total potential energy in the left axis (black line) while the total kinetic energy is in the right axis (grey line, red color) as a function of time. The system has an encapsulating energy of 10.77 eV and this energy is transformed into kinetic energy, which corresponds a velocity of ca. 250 m/s for the capsule (see Fig. 2).

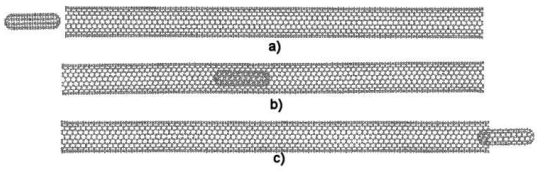

FIG. 1: Schematic representation of the capsule and nanotube nano-oscillator system. Figure a is the initial configuration.

B. A fullerene encapsulated into the nanotube and a penetrating capsule.

The problem to be considered now refers to the collision process between a penetrating capsule, as described in the previous section, with a C_{60} fullerene at rest in an equilibrium position inside the nanotube. Before simulating the molecule being expelled from the nanotube, it was performed a simulation of the behavior of the C_{60} molecule inside the nanotube, in order to determine its equilibrium position. Then the capsule was abandoned near the nanotube entrance cavity, as in the previous section. Fig. 4 shows four different stages of

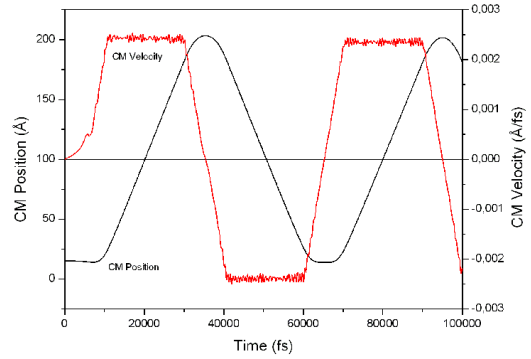

FIG. 2: The center of mass (CM) position (black line) and the CM velocity (red line).

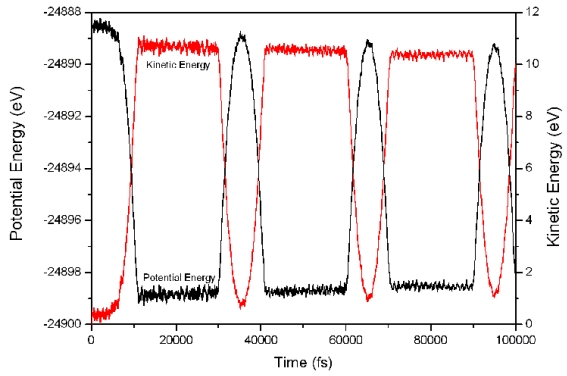

FIG. 3: Total potential (black line) and kinetic (red line) energies of the capsule and nanotube, forming a nano-oscillator system.

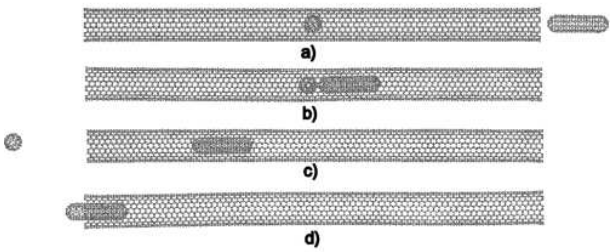

FIG. 4: Schematic representation of the capsule, nanotube and C_{60} fullerene. Figure a) is the initial configuration.

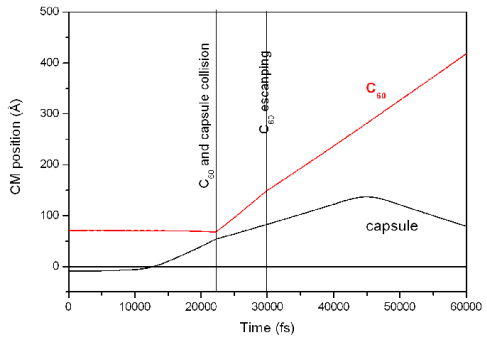

FIG. 5: The CM position for the capsule (black line) and fullerene (red line).

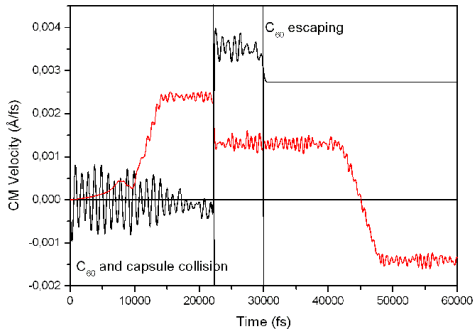

FIG. 6: The CM velocity for the capsule (red line) and fullerene (black line).

the penetration process. Fig. 4a shows the initial configuration, Fig. 4b corresponds to the moment of collision between the capsule and the fullerene, while Fig. 4c shows the moment just after the one at which the fullerene escapes from the nanotube. Fig. 4d shows a moment of returning for the capsule. The penetrating capsule, when reaching the fullerene, transfers to it enough kinetic energy so that the C_{60} can escape from the interior of the nanotube. The CM positions for the capsule and fullerene are represented in Fig. 5. The grey (red in color) line shows the fullerene CM position while the black one indicates the CM position of the capsule. At a time value indicated as 22,500 fs, occurs the collision between capsule and fullerene. Just before this, the capsule had a velocity of 270 m/s, as can be seen in Fig. 6. Just after collision the fullerene acquires a velocity of 350 m/s and escapes from the nanotube at a time value corresponding to about 30,000 fs. Far way from the nanotube end, the fullerene velocity is 275 m/s as a result of the interaction fullerene-nanotube during the time between collision and when it is expelled. The capsule velocity goes down to 125 m/s just after the collision and remains encapsulated inside the nanotube.

IV. SUMMARY

A Molecular Dynamics simulation was performed for the interaction process among a C_{60} fullerene, a carbon capsule and a carbon nanotube. The system was studied using a van der Waals potential for interaction between atoms of each unit and an empirical Tersoff-Brenner potential for interactions inter-units. We have discussed the encapsulation process of a carbon capsule with 210 atoms inside a (10,10) carbon nanotube with 3,200 atoms. The encapsulation energy was 10.77 eV and the oscillatory frequency was ca. 17.18 GHz. After determining the equilibrium position of a C_{60} fullerene inside the nanotube, it was considered the collision process of the penetrating capsule into the nanotube and its interaction with the fullerene. This has resulted in the expulsion of the fullerene while the capsule resulted trapped in the interior of the nanotube. After collision, the C_{60} fullerene acquired a velocity of 350.0 m/s which was enough for it to escape out of the nanotube. When getting free from the interaction with the nanotube, the velocity of the C_{60} fullerene is reduced to 275 m/s. We have so demonstrated that is possible to expell a C_{60} fullerene from a single walled carbon nanotube using a carbon capsule.

V. ACKNOWLEDGEMENTS

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