

The Oscillatory Characteristics of a $2C_{60}$ /CNT Oscillator System

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The authors have studied, using molecular dynamic (MD) simulations, the oscillatory characteristics of a $2C_{60}$ /CNT oscillator system, in which two C_{60} fullerenes oscillate inside a single walled carbon nanotube (CNT) in two basic modes, i.e., the symmetric and non-symmetric motions. In the symmetric mode, with each oscillation the two fullerenes move symmetrically from the CNT ends towards the CNT center where they bounce off each other and head back towards the ends. In the non-symmetric mode, the two fullerenes move back and forth inside the CNT crossing the center point of the CNT together with each oscillation. The simulations show that the non-symmetric oscillation mode is stable for the prescribed initial (maximum) velocities up to 300 m/s, while the symmetric oscillation mode however, experiences dynamic instabilities for a prescribed initial (maximum) velocity larger than 250 m/s. The instability takes place as a result of the transfer of energy from the translational to the rotational motion of the fullerenes. This characteristic differentiates $2C_{60}$ /CNT oscillators from double-walled CNT oscillators. The rotation is primarily caused by the inter-colliding of the two fullerenes, which subjects the fullerenes to large van der Waals repelling forces. These repelling forces are not necessarily aligned perfectly along the CNT axis nor precisely pointing towards the mass centers of the fullerenes. These misalignments cause the fullerenes to rock around the CNT's axis, while their offsets from the mass centers cause the fullerenes to rotate. The rocking motion, being severely confined by the CNT, does not gain much energy itself, but instead, channels energy from translational to rotational motion. The energy channeling is found to be reversed in some very short time intervals, but the rotational motion always gains energies from the translational motion over a time interval that is long enough at the MD time scale. This feature, contrary to our experiences in the macroscopic world, appears to be unique for such nanoscopic mechanical systems.

Keywords: Fullerene, Carbon Nanotube, GHz Oscillator, Energy Dissipation, Nano System, MD Simulations.

1. INTRODUCTION

Since the first proposal¹ to use the relative motion of different walls of a multiwalled carbon nanotube (CNT) for nanoscopic mechanical oscillators of frequencies in the GHz range, there have been many investigations^{2–14, 18–29} on CNT-based oscillators. With their detailed calculations of the excess van der Waals (vdW) energy due to the extrusion of the oscillating core from the outer tube and the corresponding restoring force, Zheng et al.³ have shown that the oscillation frequency can be significantly increased

by shortening the oscillation core. This finding, in part, has motivated the study of the C_{60} /CNT oscillator system, in work such as that by Liu et al.,⁴ in which one C_{60} oscillates inside a CNT. Prior to this work, Qian et al.⁵ investigated firing a C_{60} into a CNT with high velocities and studied its subsequent motion forward and backward. The various issues related to energy dissipation of CNT oscillators have been investigated by many researchers, including Guo et al.,⁶ Zhao et al.,⁷ Ma et al.,⁸ and Tangney et al.⁹ One of the common features reported in these studies is that some of the energies in the translation motion are always irreversibly transferred to thermal energies, resulting in an increase of system temperature. This is typical

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in nano-sized systems, where useful motions gradually diminish when energies unidirectionally and continuously flow to thermal bath if systems are isolated without energies being supplied.

Multiple fullerenes are found encapsulated in CNTs and, in many cases, they are aligned along the CNT axis. This eccentric structure, called a carbon peapod, exhibits unprecedented properties.^{10,11} Here we present our studies, using MD simulations, of the oscillatory behaviors of an oscillator system consisting of two fullerenes and a hosting CNT and their characteristics, particularly those that differentiate this $2C_{60}$ /CNT oscillator from the multi-walled and double-walled CNT oscillators, which have been intensively investigated. The two basic oscillation modes of this system, i.e., the symmetric and non-symmetric modes, are found to be of frequencies in the range of one hundred to several hundreds GHz. The symmetric oscillation exhibits instability behaviors as the maximum oscillation velocity (that is, the prescribed initial velocity of the fullerenes) is increased from 200 m/s to 250 m/s. The non-symmetric oscillation, on the other hand, remains stable with the maximum oscillation velocity up to 300 m/s. Our MD simulations reveal that the instability takes place through transferring the energy of the fullerene from translational to rotational motion of the fullerenes, and that this characteristic differentiates the $2C_{60}$ /CNT oscillators from the double-walled CNT oscillators. The inter-colliding of the two fullerenes, which subjects the fullerenes to extremely large van der Waals repelling forces, is primarily responsible for the onset of rotation. These repelling forces are not necessarily aligned perfectly along the CNT axis nor precisely pointing towards the mass centers of the fullerenes, and their misalignments drive the fullerenes to rock around the CNT's axis, while their offsets from the mass centers lead to the tendency of rotating the fullerenes. The rocking motion, being severely confined by the CNT, does not gain much energy itself, but instead, it channels the energy from the translation motion into the rotation motion. The energy channeling is found to be reversed in some very short time intervals, but the rotation motion always gains energies from the translation motion over a time interval that is long enough at the MD time scale. This feature, contrary to our experiences in the macroscopic world, appears to be unique for such nanoscopic mechanical systems.

2. METHOD AND MODELING

Molecular dynamics (MD) has been broadly used to study the properties of nano devices. Legoas et al.^{12,13} have performed the MD simulations on a number of CNT-oscillator systems and have shown that these nano-oscillators are dynamically stable when the radii difference values between inner and outer tube are ~ 3.4 Å. Guo et al.⁶ have thermally equilibrated double-walled CNT oscillators with a bath to reach an initial temperature, and then switched to a microcanonical ensemble for

simulations. A similar approach is used by Zhao et al.⁷ on nano-oscillator setups to study their energy dissipation mechanisms. In this study,⁷ the geometry of the oscillator is optimized first, and the system is then thermally heated up to a desired temperature prior to a microcanonical ensemble run, before which the inner tube is initially displaced by giving it an extrusion such that an initial potential energy is assigned to the system to start dynamics. Legoas et al.^{12,13} have discussed the possibilities that initialization of these systems could be achieved by applying external electric field, through charge injection, or applying variable magnetic fields, using an internal metallic (or filled with metallic materials) and an external semi-conducting nanotube. Using MD simulations, Kang et al.¹⁴ reported that encapsulated potassium ions accelerated by an applied external electric field could initialize a gigahertz actuator composed of a $7K^+$ @CNT oscillator, in which a CNT encapsulates seven potassium ions.

Our model system consists of a (10,10) CNT with a length of 2.46 nm and two encapsulated C_{60} fullerenes, as illustrated in Figure 1. We first optimize the ($2C_{60}$ /CNT) system configuration by performing minimization. The system is then heated to a pre-assigned temperature, 3 K unless stated otherwise, by performing a canonical run with Andersen thermostat, and this is the initial temperature of the microcanonical ensembles. For the subsequent MD simulations, we have used essentially the same approach as Zhao et al., except that because we would like to initiate oscillations at an optimized state,¹⁵ we have prescribed the initial kinetic energies (or the initial velocities equivalently) for the oscillating C_{60} s, while Zhao et al. assigned the initial potential energies (or the initial displacement equivalently) for the oscillating core tube. The MD simulations are carried out using Discover Dynamic Engine in MS Modeling¹⁶ with a second generation force field, named COMPASS.¹⁷ A time step of 1 fs is used for all simulations. However, much smaller time steps,

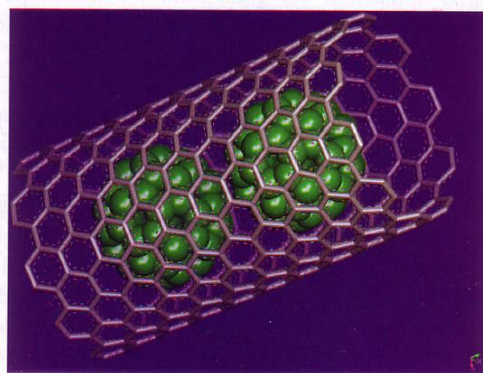


Fig. 1. The model of $2C_{60}$ /CNT oscillator system: initially, the centers of two fullerenes are positioned rightly on the CNT axis and are symmetric about the CNT's mid-section. The center-center distance between the two fullerenes is about 1 nm, and the (10,10) CNT is 2.46 nm in length. The origin of coordinate is set at the center point of the CNT's one of the two open ends.

such as 0.1 fs have been tested to ensure qualitative features from simulations are robust. This system has two basic oscillation modes, the symmetric mode, in which the two C_{60} s move towards or away from each other, and the non-symmetric mode, in which the two C_{60} s always move towards the same direction. We have generated the oscillations in the symmetric and non-symmetric modes by assigning the two C_{60} s initial velocities of the same magnitude and, respectively, with the opposite and the same directions.

3. RESULTS AND DISCUSSION

The trajectories of the oscillating fullerenes for the symmetric and the non-symmetric modes are shown in Figure 2, for which the initial velocities are the same, 200 m/s. The oscillations are shown to be very stable, and the frequencies are, approximately, 667 GHz for the symmetric mode and 115 GHz for the non-symmetric mode. In the symmetric mode, each of the two C_{60} molecules oscillates within the corresponding half of the CNT, because they collide at the mid section of the CNT and thus are repelled away with large accelerations. Consequently, the symmetric mode has a smaller oscillation magnitude and a higher oscillation frequency. The oscillation frequencies of this system, especially for the symmetric mode, are substantially higher than those of the double-walled and multi-walled oscillators reported in a number of previous investigations.^{6-9,12,13} The symmetric oscillation exhibits instability behaviors as the maximum oscillation velocity (i.e., the initial velocity) is increased from 200 m/s to 250 m/s, while the non-symmetric oscillation remains stable with the maximum oscillation velocity up to 300 m/s. The center-center distance between the two fullerenes in

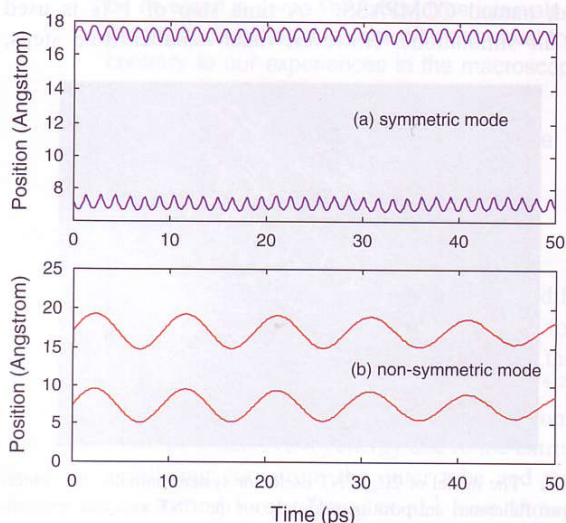


Fig. 2. The mass-center trajectories of the two fullerenes for the initial system temperature 3 K, and the initial velocity 200 m/s: Upper panel (a) for the symmetric mode; Lower panel (b) for the non-symmetric mode.

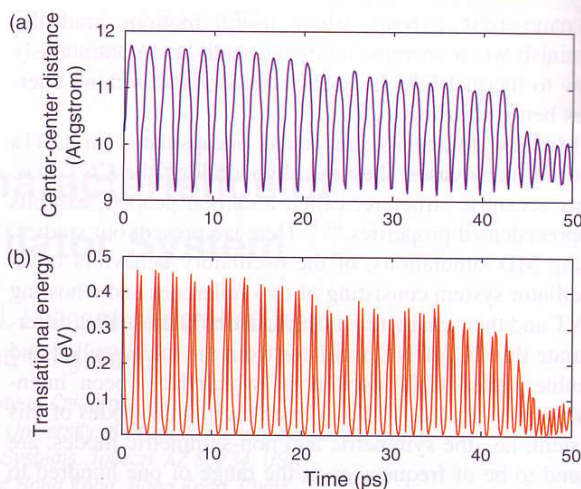


Fig. 3. Upper panel (a): the center-center distance between the two fullerenes versus the MD simulation time; Lower panel (b): the translational energy versus the MD simulation time, for symmetric oscillation with the initial temperature is 3 K and the initial velocity 250 m/s.

the symmetric oscillation with a prescribed initial velocity, 250 m/s, plotted versus the simulation time in Figure 3(a),[†] indicates that the oscillation amplitudes of the fullerenes decrease gradually during the first 42 ps, approximately, and subsequently experience a sudden drop of about 50%. The translation energy of the system, defined as the total kinetic energies of the two fullerenes in translational oscillation, exhibits corresponding instability behavior, as shown in Figure 3(b), indicating severe loss of translational energy. In their investigations of double-walled and triple-walled CNT oscillators, Zhao et al.⁷ have shown the energy transfers from the translation motion of the inner tube into the motions, ordered and disordered, of the outer tube, and this has led us to wonder if the same process has taken place in our ($2C_{60}$ /CNT) oscillator and is responsible for the energy loss shown in Figure 3. We have thus plotted in Figure 4 the energies of the hosting CNT of our ($2C_{60}$ /CNT) oscillator, both for the ordered translation motion and disordered thermal fluctuation, with the results from the same MD simulation as those shown in Figure 3. The plots indicate, however, that the gain in the translation energy of the hosting CNT is negligibly small and the thermal fluctuation energy is one order smaller than the loss of the translation energy of the fullerenes. We have therefore been challenged by the questions: where has the

[†]We have noted that the oscillation with the initial velocity of 200 m/s shown in Figure 2(a) has a slightly higher frequency than the oscillation with initial velocity of 250 m/s shown in Figure 3(a). The larger maximum velocity leads to a larger oscillation magnitude, indicating that the C_{60} molecules travel a longer distance in each oscillation period. Looking into the detailed numerical results, we find that the average velocities are nearly the same in these two cases, although the second case has a larger maximum velocity, 250 m/s. This leads to a longer oscillation period and thus a lower frequency.

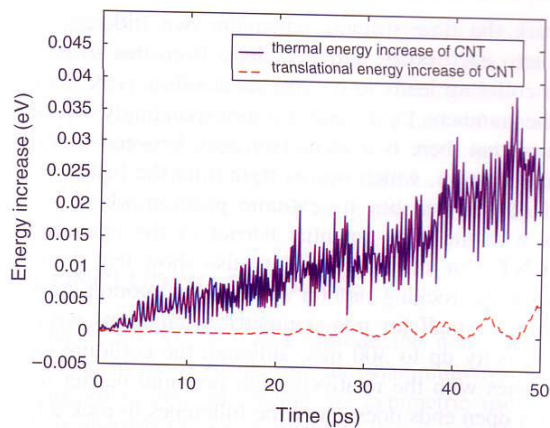


Fig. 4. The energy components of the hosting CNT for symmetric oscillations with the initial temperature 3 K and the initial velocity 250 m/s: blue solid line—the disorderly thermal energy; red dashed line—the translational energy.

translation energy of the fullerenes been leaked and how does the leakage occur in this oscillating system?

Zhao et al.⁷ have shown that the rocking motion of the inner tube is primarily responsible for the loss in the translation energy for double-walled CNT oscillators with short inner tubes, versus oscillators with long inner tubes, for which the rocking motion is suppressed by the confinement of the large “close-contact surfaces.” While, our ($2C_{60}/CNT$) system has the smallest “close-contact surface” and thus the confinement on other motions, the rotation motion in particular, is far less significant. We have therefore calculated the total rotation energy of the two fullerenes of the same system with two different initial velocities, $v = 200$ m/s, and 250 m/s, respectively, and have plotted them in Figure 5(a). For the case $v = 200$ m/s,

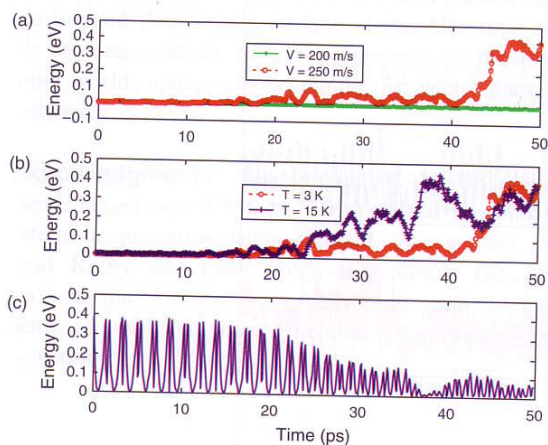


Fig. 5. The rotational energies for symmetric oscillations: (a) with initial velocities 200 m/s and 250 m/s respectively, and with the same initial temperature 3 K; (b) with initial temperatures 3 K and 15 K, respectively, and with the same initial velocity 250 m/s; and (c) the translational energy for the symmetric oscillation with initial velocity 250 m/s and initial temperature 15 K.

the rotation energy remains zero, practically, during the entire MD simulation, and this is consistent with the trajectory plot shown in Figure 2(a), indicating that the oscillation is stable during the entire simulation. The rotation energy for the case $v = 250$ m/s is seen increasing gradually and very moderately during the first 42 ps and subsequently, it suddenly rises to nearly 0.4 eV. This corresponds amazingly well with the translation energy plot in Figure 3(b), which shows that the translation energy magnitude decreases gradually and moderately until about 42 ps, when it suffers a sudden loss of about the same magnitude. We have also investigated the thermal effect on the instability by running the MD simulations for the identical system with the same initial velocity but with different initial temperatures, and the simulations indicate that a higher initial temperature leads to not only a larger loss of the translation energy into the rotation energy but also an earlier onset of the rotation motion, as shown in Figure 5(b) where the rotation energies for the initial temperatures 3 K and 15 K, respectively, are plotted versus the simulation time. We have plotted in Figure 5(c) the translational energy versus the simulation time for the initial temperature 15 K and initial velocity 250 m/s. The plot in Figure 5(c) shows the periodic exchange between the translational energy and the potential energy, as expected for such an oscillating system, and more interestingly, it also makes apparent the exchange between the translational energy and the rotational energy with the decrease and increase in its magnitude being well coordinated with the increase and decrease of the rotational energy plotted in Figure 5(b). We have noted that this energy exchange phenomenon persists as we increase the temperature, even at the room temperature, but the energy plots, however, become much jagged possibly due to relatively large thermal effects, which are of a disordered nature. Our choices of 3 K and 15 K are subjective for the clarity of presentations.

As discussed previously, the rocking motion is primarily responsible for dissipating the translation energy for double-walled CNT oscillators with short inner tubes. For our ($2C_{60}/CNT$) oscillator, the fullerenes do bounce around the CNT axis, along which they oscillate, and we refer to this motion as the rocking motion because of its analogy to the rocking motion of the inner tube for the double-walled CNT oscillators. We have plotted this rocking motion energy together with the translation and rotation energies in Figures 6(a) and (b) for initial velocities 250 m/s and 300 m/s, respectively. It is seen that the rotation energy climbs up consistently with the MD time, and the crest of the rotation energy amplitude corresponds to the trough of the translational energy amplitude rightly, which illuminates that the energy transfer takes place between the translation motion and the rotation motion. To our surprise, however, the rocking energy is negligibly small compared to the translation and the rotation energies, and this has prompted us to further investigate the role of the rocking motion.

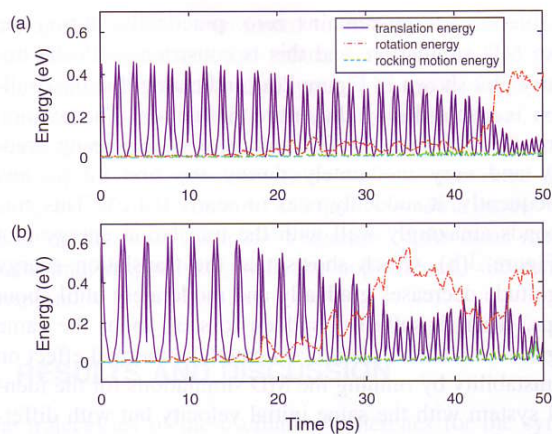


Fig. 6. The translation, rotation, and rocking energies for symmetric oscillations with initial temperature 3 K, and with (a) initial velocity 250 m/s; (b) initial velocity 300 m/s.

We have thus plotted, in Figures 7(a–d), the trajectory, the offset (the distance of the mass center to the CNT axis), the offset velocity (the velocity component perpendicular to the CNT axis) and the angular velocity magnitude versus the simulation time for one of the two fullerenes in oscillation and again for the initial velocity 250 m/s. It is seen that the fullerene has essentially no angular velocity during the first 20 ps, and its angular velocity increases very moderately until after the 40th ps when it rises sharply. We note that the offset velocity and the offset itself augment significantly near the 40th ps, indicating that the rocking motion of the fullerene is apparently responsible for the rotation motion. The insert of Figure 7 shows the detailed trajectory and offset profiles during the simulation time: 26 ps to 32 ps. The numbers 1, 2, and

3 mark the time instants when the two fullerenes collide into each other, and it is seen from this insert that each colliding leads to a subsequent offset peak, marked by the numbers 1', 2', and 3', correspondingly. We have noticed that there is a secondary peak between each pair of major peaks, which occurs right after the instant when the fullerene reaches its extreme position where it collides with the high potential barrier of the open end of the CNT. Our simulation results also show that the energies for the rocking motion and rotation motion are both negligibly small for non-symmetric oscillations with initial velocity up to 300 m/s, although the colliding of the fullerenes with the relatively high potential barrier of the CNT's open ends does cause the fullerenes to rock a little. We have thus concluded that the rocking motion (i.e., the off-axis motion) is primarily caused by the inter-colliding of the two fullerenes, which subjects the fullerenes to very large van der Waals repelling forces. These repelling forces should not be expected to be aligned perfectly along the CNT axis or precisely pointing towards the mass centers of the fullerenes, because of the discrete distribution of the carbon atoms over the surface of each fullerene. The rocking motion is severely confined by the CNT and correspondingly, the rocking motion does not gain much energy itself. Instead, it channels the energy from the translational motion into the rotational motion. The energy profiles for the initial velocity 300 m/s, plotted in Figure 6(b), shows that the rotational energy climbs up to a peak at $t = 35$ ps, and then falls down, followed by a second peak at about $t = 47$ ps. The translational energy goes down and up in the opposite direction. We note the recent report of Ma et al.³⁰ on a DWNT oscillator. Their simulations apparently suggest that the inner tube may exhibit a behavior of a somewhat similar nature to the phenomenon we report

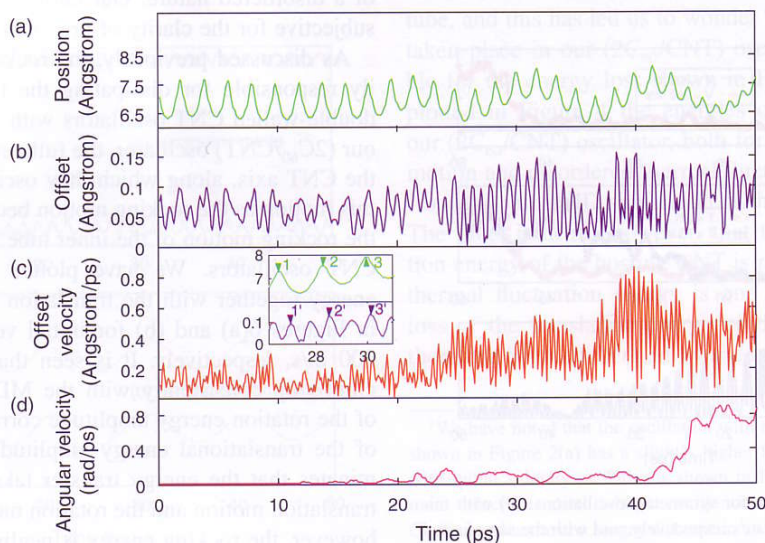


Fig. 7. The trajectory (a), offset (b), offset velocity (c), and angular velocity (d) of one of the two fullerenes in symmetric oscillation with initial velocity 250 m/s and initial temperature 3 K. Inserts: the time correlation between the trajectory (upper green line) and the offset (lower blue line) of the C_{60} during 26 ps ~ 32 ps: number 1, 2, and 3 mark the collision instants, and number 1', 2', and 3' mark the corresponding peaks in the offset.

here. The phenomenon that the energy transfers back to the translation motion from the rotation motion is apparently contrary to our experiences in the macroscopic world, where the energy transfer from the translational motion to the rotational is irreversible, such as is the case in bowling, in which a ball makes an irreversible transition from sliding to rotating as it moves along the bowling track.

4. CONCLUDING REMARKS

For the $2C_{60}$ /CNT oscillator system, the non-symmetric oscillation mode is stable for the maximum oscillation velocity up to 300 m/s, while the symmetric oscillation mode is unstable for a maximum oscillation velocity larger than 250 m/s. The instability takes place through transferring the energy of translational motion to the rotational motion of the fullerenes, and this characteristic differentiates $2C_{60}$ /CNT oscillators from double-walled CNT oscillators for which instability occurs primarily through transferring energy from translational motion to the ordered and disordered motions of the outer tube. The $2C_{60}$ /CNT rotation is primarily caused by the inter-colliding of the two fullerenes, which subjects the fullerenes to large van der Waals repelling forces. These repelling force are not necessarily aligned perfectly along the CNT axis nor precisely pointing towards the mass centers of the fullerenes. Their misalignments drive the fullerenes to rock around the CNT's axis, and their offsets from the mass centers cause the fullerenes to rotate. The rocking motion is severely confined by the CNT and correspondingly, the rocking motion does not gain much energy itself. Instead, it channels the energy from the translational motion into the rotational motion. The energy channeling is found to be reversed in some very short time intervals, but the rotation motion always gains energies from the translation motion over a time interval that is long enough at the MD time scale. This feature, contrary to our experiences in the macroscopic world, appears to be unique for such nanoscopic mechanical systems.

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15. Assigning an initial velocity to a structure before a dynamic run is not a natural feature of Discover in MS Modeling 3.2. As a development, we realize this by handling the BTCL language by which the simulations are organized.
16. MS Modeling 3.2, Materials Studio's premier modeling and simulation product suite, is licensed to Accelrys Inc. (2005).
17. COMPASS stands for Condensed-phase Optimized Molecular Potentials for Atomistic Simulation Studies. It is the first *ab initio* force field that has been parameterized and validated using condensed-phase properties in addition to various *ab initio* and empirical data for molecules in isolation. It is licensed to Accelrys Inc., (2005).
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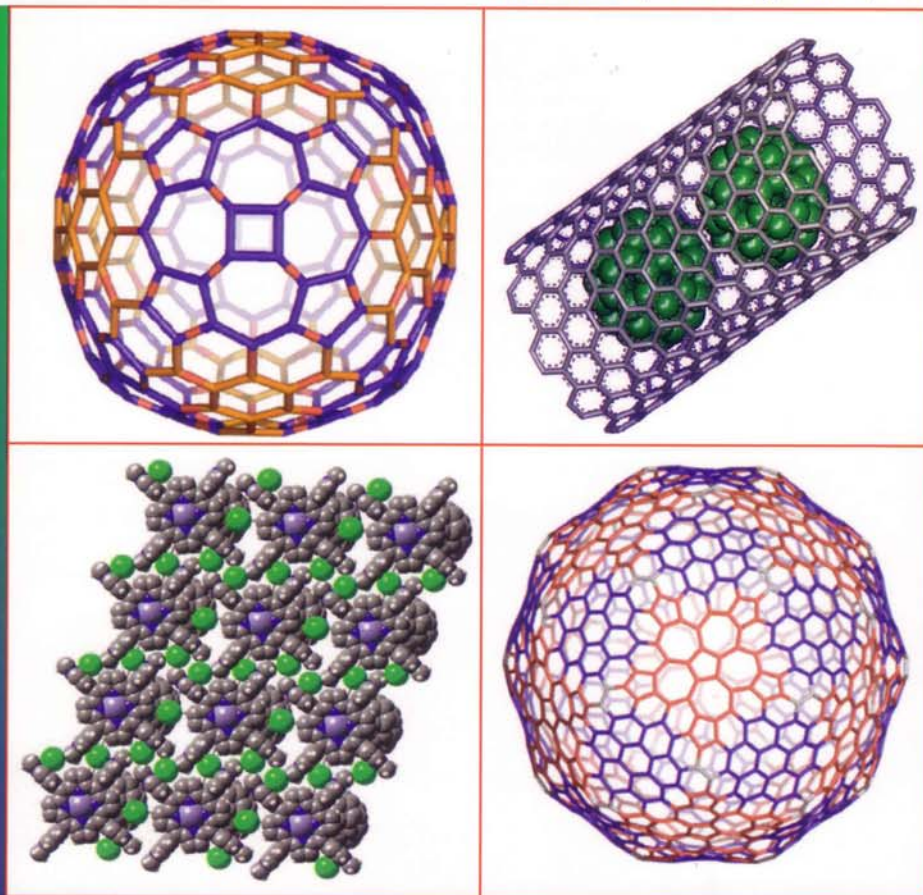
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