Energy Dissipation in Gigahertz Oscillators from Multiwalled Carbon Nanotubes

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Using atomistic models and molecular dynamics simulations, interlayer corrugation and resistant force in a biwalled carbon nanotube are shown to be strongly dependent upon the morphology combination of the bitube. Consequently, energy dissipation in a commensurate (e.g., armchair/armchair or zigzag/zigzag) bitube oscillator is found to be much larger than that in an incommensurate (e.g., zigzag/armchair) oscillator, resulting in a decay of oscillation amplitude within a few nanoseconds in the commensurate bitube and several tens of nanoseconds in the incommensurate bitube.

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Multiwalled carbon nanotubes (MWNTs) have broad prospects as components in nanomechanical devices due to many exceptional electrical and mechanical properties, including wear-free interlayer sliding and ultralow friction [1–6] . Cumings and Zettl [1] have managed to open one end of a MWNT and telescope a core tube out of an outer-housing shell. The interlayer telescoping friction force was estimated to be less than 1.5×10^{-14} N per atom. As the core tube is pulled out and set free, Cumings and Zettl [1] found that it spontaneously retracts into the housing shell [Fig. 1(a)] within one video frame time of 33 ms. Zheng and colleagues [7,8] proposed that MWNTs may be used to create gigahertz nano-oscillators. When both ends of the outer shell are opened, the retracted core will not stop at its original position but oscillates with gigahertz frequency with respect to the outer housing according to the law of energy conservation [Fig. 1(b)]. The gigahertz oscillator from MWNTs may have potential applications [9–11].

Currently, efforts are being made to bring the theoretical prediction of a MWNT nano-oscillator into reality in the laboratory, yet no success has been reported. An important step in experimentally detecting a MWNT oscillator would be to develop methods to maintain oscillation for long enough time duration. Reducing energy dissipation or applying an external field are two possible approaches.

A single carbon nanotube can be indexed by (n, m) which define the circumference in the lattice coordinates of a reference graphite sheet. A biwalled system denoted by $(n_o, m_o)/(n_c, m_c)$ is typically axially commensurate when $n_o/m_o = n_c/m_c$; otherwise it is said to be axially incommensurate. For a short outer shell sliding along a long inner tube, the energy is not constant but fluctuating with corrugation dependent upon both the commensuration and tube size [12]. In a bitube oscillator, the overlapping section changes with time with different interlayer interaction in sliding. Energy corrugation and interlayer force fluctuation have been found to change with the axial commensuration of the tube pair [13]. Although the interlayer sliding resistance force is small in comparison with the van der Waals force [2] and has negligible effect on the oscillating frequency [8], friction induced energy dissipation is nevertheless inevitable. The issue of energy dissipation [14] can be a key obstacle to sustained oscillation. This issue has not been addressed in previous studies [7,8,15].

In this Letter, we perform molecular dynamics (MD) simulations of a bitube oscillator to show that the rate of energy dissipation depends upon the commensuration and relative morphology of the bitube. Temperature control is an important issue in such simulations. A nonequilibrium finite temperature system is difficult to treat by MD simulations and currently there are no reliable simulation tools for such problems. As an initial effort to understand the complex problem of energy dissipation in nanomechanical systems, we have simply chosen a model problem involving a bitube nano-oscillator which is equilibrated at an initial temperature and then simulated under constant energy condition (i.e., a microcanonical NVE ensemble in which the total energy E, volume V, and number of molecules N are fixed and unchanging). Our aim is to investigate energy dissipation under these

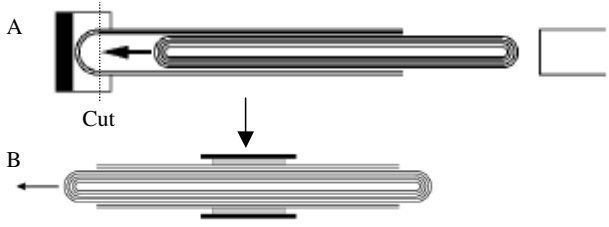

FIG. 1. Schematic representation of MWNT oscillators. (a) The telescoped inner core is released and then retracts back into the outer shell by the interlayer van der Waals interaction in a MWNT system (experiments of Cumings and Zettl [1]). (b) With both ends of the outer shell opened, the core, when released at a telescoping distance, should oscillate back and forth with respect to the other shell (proposed theoretically by Zheng and Jiang [7]).

assumptions in order to gain a first insight into the behavior of such systems.

The smoothest oscillator we studied sustains oscillation up to 60 ns at a low initial temperature. As the temperature rises, the energy dissipation rate sharply increases and the oscillation decays into thermal motion more quickly. Incommensurate tube systems such as an armchair/zigzag pair are found to be much smoother than commensurate systems such as armchair/armchair and zigzag/zigzag pairs. Among the systems we studied, the zigzag/zigzag bitube shows the largest dissipation rate. There are significant size effects on the duration and frequency of oscillation.

Telescoping motion within a MWNT tends to occur between two layers with the smoothest van der Waals interaction [1]. We focus on a bitube system with lengths of the outer and core tubes denoted as L_o and L_c. Most simulations are based on the standard AMBER molecular force field [16] which includes van der Waals, bond stretch, bond angle bending, and torsional interactions. The Tersoff-Brenner potential [17] and the 12-6 Lennard-Jones potential are also used for comparison.

Typical results are presented in Fig. 2, where the abscissa ξ is the center-of-distance separation between the core and the outer shell and $L_o \approx L_c$ is taken to be 3.5 nm. The force in the figure is obtained as the partial differential of the total interlayer van der Walls energy with ξ. When ξ is greater than a certain value, the average telescoping force tends to be constant and independent of the tube length. However, the telescoping force fluctuates around an average value with fluctuation amplitude dependent upon the registry combination of the two tubes.

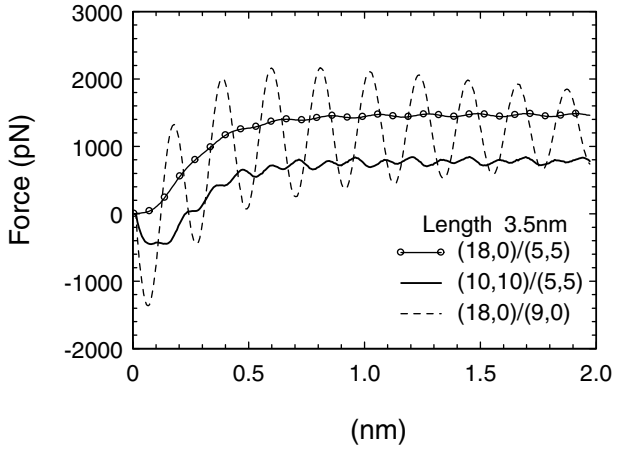

FIG. 2. Atomistic calculations of the telescoping force vs displacement for three typical bitube systems. The force fluctuation range of the armchair/zigzag system is 35 pN and is independent of the tube length; in the armchair/armchair and zigzag/zigzag systems, the fluctuation range increases with tube length and is about 152 and 1926 pN, respectively, for the tube length indicated in the figure.

Figure 2 shows that the force fluctuations in the $(18, 0)/(5, 5)$ armchair/zigzag, $(10, 10)/(5, 5)$ armchair/armchair, and $(18, 0)/(9, 0)$ zigzag/zigzag systems are proportional to the ratio of 1.0:4.4:55.1 for the given tube length. The fluctuation is smallest in an armchair/zigzag system and highest in a zigzag/zigzag system. Interestingly, the force fluctuation is independent of the tube length in the armchair/zigzag system, but increases with the tube length in the armchair/armchair and zigzag/zigzag systems. These properties do not change with tube diameters provided the interlayer gap is close to 0.34 nm. In typical experiments, the length of tubes may be up to several hundreds of nanometers and only the smoothest tube layer can be pulled out [1]. This means that systems with the properties of an armchair/zigzag system are most important. Generally speaking, incommensurate systems have similar properties as the armchair/zigzag system while commensurate systems have similar properties as the armchair/armchair or the zigzag/zigzag system. Legoas *et al.* [15] investigated tubes with more walls and concluded that the bitube system already contains the most relevant effects. Without loss of generality, we will limit our discussions to the armchair/zigzag, armchair/armchair, and zigzag/zigzag bitube systems.

In order to investigate the energy dissipation mechanism and the duration of sustained oscillation in different systems, we have performed MD simulations up to 3 ns at two different initial temperatures [18]. The AMBER standard force fields and the microcanonical dynamics are adopted without temperature control in the simulations. After initial energy minimization under a given temperature, the core tube is telescoped out to certain displacement of ξ_0 (about $1/4$ to $1/2$ of the tube length) and then released to start the oscillation. The outer shell is fixed at three atoms (two at one end and one at the other end). The time step is 1 fs and total energy variation remains within 0.03% in the simulations.

When the core is released at $\xi = \xi_0$, the potential energy is the highest and the kinetic energy is the lowest with zero center-of-mass velocity. As the core is retracted back into the outer shell by the van der Waals force, the potential energy decreases and the kinetic energy increases until the core returns to $\xi = 0$. At this point, the kinetic energy reaches the maximum and the core continues to move out of the other end of the outer shell, after which the potential energy increases and the kinetic energy decreases, and so on. The interlayer friction causes the oscillating amplitude and the peak of the potential energy to decrease over time. Figures 3(a) and 3(b) show the variation of the potential energy in $(18, 0)/(5, 5)$ and $(10, 10)/(5, 5)$ systems at an initial temperature of about 8 K. In the inset, the potential energy variations near the beginning and the end (3 ns) of simulation are presented. At such a low temperature, the oscillation is regular and relatively stable up to 3 ns but its amplitude decreases with time, as is explicitly plotted in Fig. 3(c).

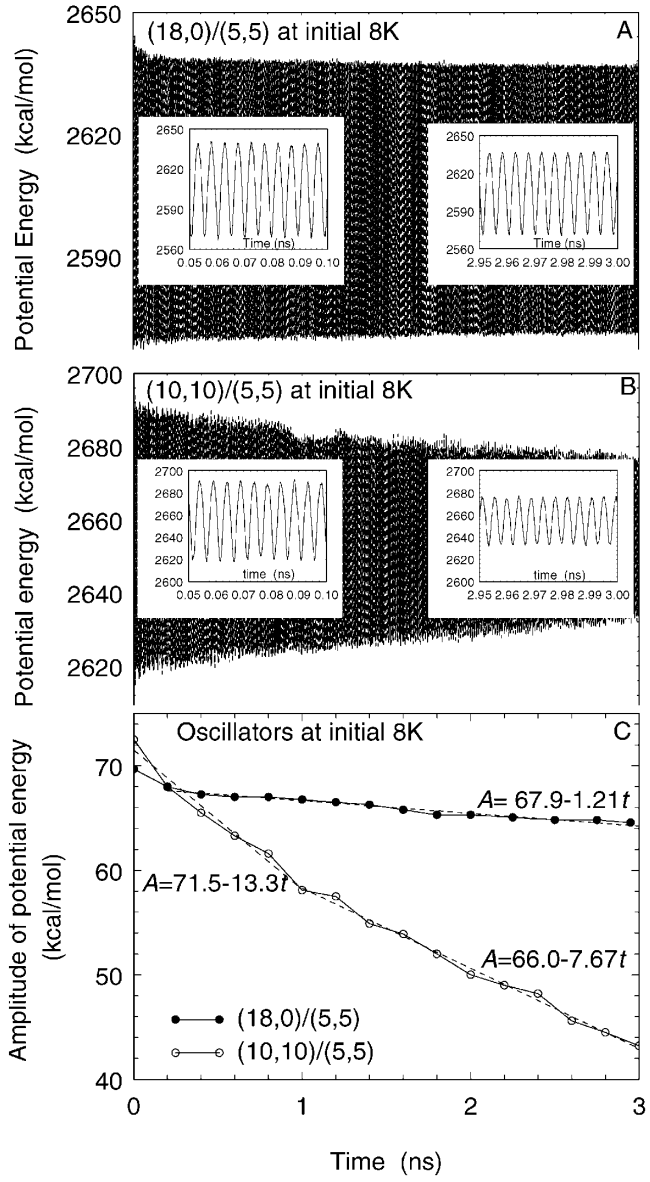

FIG. 3. Variation of potential energy with time at an initial temperature of about 8 K. (a) $(18, 0)/(5, 5)$ tube system; (b) $(10, 10)/(5, 5)$ tube system; the insets show variation of potential energy around 50 and 3000 ps. (c) Reduction of amplitude of potential energy with oscillating time. The tube length is about 3 nm and the initial extrusion displacement is about 1/4 of the tube length. The dashed linear lines and equations in (c) are corresponding fitting curves of the molecular dynamic simulation results.

The energy dissipation rate in the armchair/zigzag system is 1.21 kcal/mol/ns, much lower than that (about ~7.7–13 kcal/mol/ns) in the armchair/armchair system. The energy dissipation rate in a zigzag/zigzag system with the same tube length is much higher and can reach up to 22 kcal/mol/ns. In Fig. 3, the initial amplitude of the potential energy is about 70 kcal/mol, and the oscillating durations of the armchair/zigzag, armchair/armchair, and zigzag/zigzag systems are estimated to be about 58, 9, and 3 ns, respectively.

When the initial temperature increases to about 150 K, the rate of energy dissipation increases significantly, and the oscillating cycles become less regular even in the armchair/zigzag system as shown by Fig. 4. In this case, regular oscillation can be sustained only for several nanoseconds. At the room temperature of 300 K, regular oscillatory motion is expected to be more difficult.

In the armchair/zigzag and other incommensurate systems, the maximum telescoping force fluctuation does not increase with the tube length [13]. For longer incommensurate systems with larger initial oscillating amplitude ξ_0, larger initial amplitudes of potential energy can be achieved which should be beneficial for maintaining regular oscillation over a longer time period. Molecular dynamics simulations for the armchair/zigzag oscillator with ξ_0 2 times larger than that in Fig. 3 show that the oscillating duration is about 62 ns, slightly longer than 58 ns. In commensurate systems, increasing tube length leads to higher interlayer friction.

Because of the energy dissipation, the oscillating amplitude $\xi_{\max}$ decreases while the oscillating frequency increases with time. The higher the energy dissipation rate, the faster the frequency increases. Figure 5 shows the variation of oscillating frequency of the $(18, 0)/(5, 5)$ system with time. For such a bitube system of 3 nm in length, the oscillating frequency is over 100 GHz. At an initial temperature of 8 K, energy dissipation is relatively low and the frequency increases slowly; at an initial temperature of 150 K, increase of frequency is much faster. For longer tubes with larger $\xi_{\max}$ or a heavier core, the oscillating frequency will decrease [8]. Increasing the weight of the core without change in the interlayer interaction and friction might further decrease the energy dissipation rate. Such issues will be addressed in future studies.

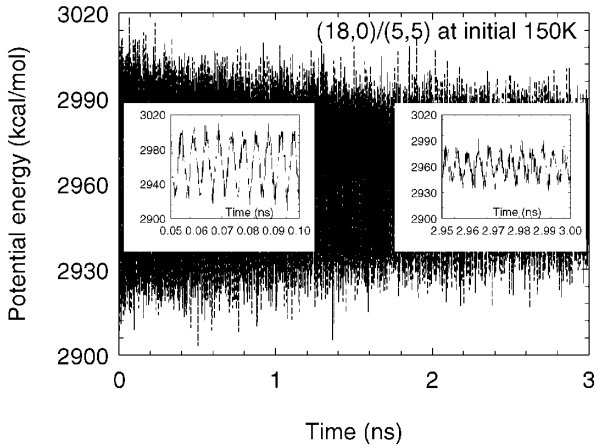

FIG. 4. Variation of potential energy with time in the $(18, 0)/(5, 5)$ bitube system at an initial temperature of 150 K.

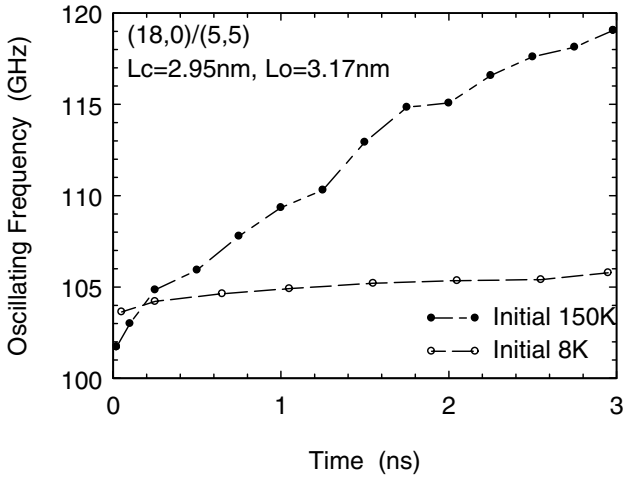

FIG. 5. Variation of oscillating frequency with time in the (18, 0)/(5, 5) system at different temperatures.

In the above analyses, the 12-6 Lennard-Jones potential is used to simulate the interlayer interaction. Using L-J potential leads to slightly smaller fluctuations in interlayer telescoping force than the Kolmogorov-Crespi potential [13], but both potentials confirm that the force fluctuation is independent of tube length in the incommensurate system we consider.

The intrashell potential is found to have much less effect on the telescoping force fluctuation. Even a rigid tube system yields nearly the same telescoping force fluctuation as a flexible tube system but, when the torsion term in the AMBER potential is removed, the predicted duration of regular oscillation decreases significantly as the intrashell stiffness becomes too weak.

The effective dynamic interlayer friction of the MWNT nano-oscillators can be estimated from the potential energy dissipation rate dE_{pot}/dt and the oscillating frequency f as

$$F_{\mathrm{eff}} = \frac{1}{4\xi_{\max} f}\frac{dE_{\mathrm{pot}}}{dt}. \quad (1)$$

Using the results of Fig. 3, F_{eff} is estimated to be 0.085, 0.400, and 0.612 pN/atom for the armchair/zigzag, armchair/armchair, and zigzag/zigzag systems, respectively. In comparison, the corresponding static amplitudes of force fluctuation are 0.053, 0.865, and 9.795 pN/atom, respectively. Therefore, the effective dynamic friction correlates well with the static force fluctuation, but is not in linear proportion to it. As the telescoping length is increased by 50%, F_{eff} in the armchair/zigzag system decreases from 0.085 to 0.040 pN/atom. This is on the same order as predicted by Cumings and Zettl [1].

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