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Self-Excited Oscillation of Rotating Double-Walled Carbon Nanotubes

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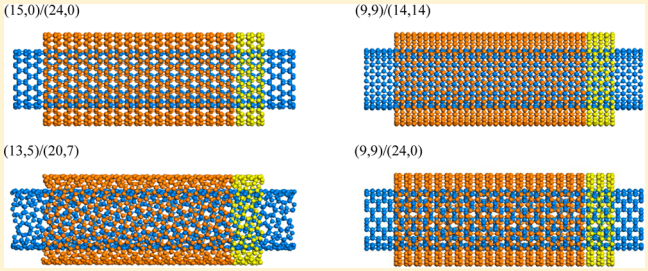
ABSTRACT: The oscillatory behavior of a double-walled carbon nanotubes with a rotating inner tube is investigated using molecular dynamics simulation. In the simulation, one end of the outer tube is assumed to be fixed and the other is free. Without any prepullout of the rotating inner tube, it is interesting to observe that self-excited oscillation can be triggered by nonequilibrium attraction of the ends of two tubes. The oscillation amplitude increases until it reaches its maximum with decrease of the rotating speed of the inner tube. The oscillation of a bitube is sensitive to the gap between two walls. Numerical results also indicate that the zigzag/zigzag commensurate model with a larger gap of >0.335 nm can act as a terahertz oscillator, and the armchair/zigzag incommensurate model plays the role of a high amplitude oscillator with the frequency of 1 GHz. An oblique chiral model with a smaller gap of <0.335 nm is unsuitable for the oscillator because of the steep damping of oscillation.



KEYWORDS: *Carbon nanotube, self-excited oscillation, gigahertz oscillator, molecular dynamics*

Carbon nanotubes (CNTs) are of chirality characterized by the helical angle, at which a graphene sheet is rolled to produce a given CNT. The chirality can be described by two chiral indices (n,m) which define the circumference in the lattice coordinates of a reference graphite sheet. A double-walled system is denoted by $(n_i,m_i)/(n_o,m_o)$, where (n_i,m_i) and (n_o,m_o) are the chiral indices of the inner and outer tubes. The system is typically axially commensurate when $n_i/m_i = n_o/m_o$; otherwise, it is said to be axially incommensurate.[1] The unique property of chirality creates the CNTs' excellent mechanical, electrical, and thermal properties, which have made them a candidate in the application of micro-electromechanical systems (MEMS) and nano-electromechanical systems (NEMS).[2] In 2000, several experiments[3−5] demonstrated the low interactions between adjacent walls of multiwalled carbon nanotubes (MWCNTs) by pulling the inner tubes out of MWCNTs. For example, Cumings and Zettl[3] opened the caps of MWCNTs at one end of the outer shells and pulled core tubes out of the shells by a nanomanipulator. After the core tubes were released, van der Waals force drew them back into the outer shells. Since that time, much attention has been paid to this phenomenon for fabricating molecular oscillators or resonators.

It is noted that Zheng and colleagues[6,7] initially proposed a continuum mechanics-based static model of a gigahertz MWCNT oscillator. In their simulations, both ends of outer shells of a MWCNT were assumed to be open, and the inner tubes were pulled out of the fixed outer tubes, after which they found that oscillation would occur under the van der Waals restoring force. To further understand the functional abilities of the MWCNT, Legoas et al.[8] presented a molecular dynamics study of the nano-oscillation with respect to dynamical aspects such as temperature, force, and energy temporal fluctuations and showed that telescopic extension and retraction movements are possible for a large combination of tube diameters and types (armchair, zigzag, and chiral). However, sustained oscillations are possible only when the radii difference values between inner and outer tubes are about 3.4 Å. Guo et al.[1] stated that the interlayer sliding resistance force is small in comparison with the van der Waals force[9] and has negligible effect on the oscillating frequency,[10] but friction-induced energy dissipation is nevertheless inevitable. The issue of energy dissipation is a key obstacle to sustained oscillation. They performed 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, and temperature control is an important issue in such simulations. In considering the effects of initial temperature on oscillatory behavior, Damnjanovic et al.[11] simulated the behaviors of both commensurate and incommensurate double-walled carbon nanotubes (DWCNTs). They found that the incommensurate model with an armchair inner tube and a zigzag outer tube would yield the smallest energy dissipation rate compared with other commensurate tubes. Rivera et al.[12,13] simulated the oscillation of incommensurate DWCNTs of various lengths between 12.21 and 98.24 nm.

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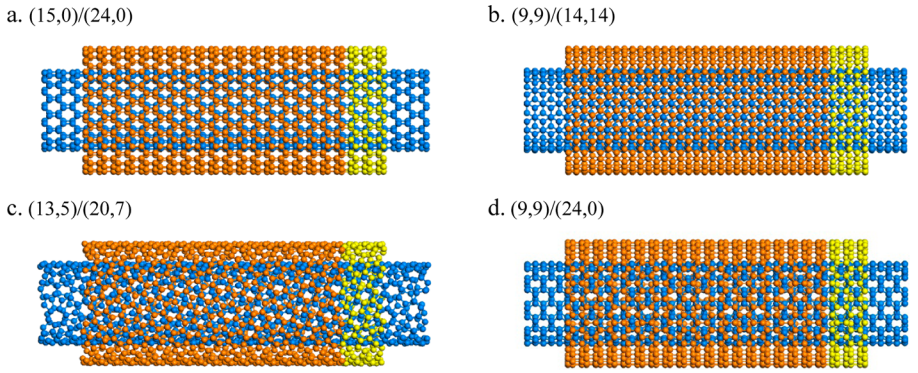

Figure 1. Simulation modes of four different DWCNTs with open ends, the atoms (yellow parts) near the right-hand end of the outer tube being fixed.

Table 1. Geometric Parameters of DWCNTs in Figure 1

bitube	length (nm)	radius (nm)	gap between walls (nm)	atoms in walls	helical angle
(15,0)/(24,0) (Figure 1a)	6.048/5.040	0.596/0.953	0.357	870/1152	0°/0°
(9,9)/(14,14) (Figure 1b)	5.986/4.864	0.619/0.963	0.344	882/1120	30°/30°
(13,5)/(20,7) (Figure 1c)	5.995/5.050	0.639/0.964	0.325	886/1118	15.6°/14.5°
(9,9)/(24,0) (Figure 1d)	5.986/5.040	0.619/0.953	0.334	882/1152	30°/0°

Their results implied that the inner wall of the incommensurate DWCNTs had damped oscillatory behavior at gigahertz frequencies, and their corresponding amplitude showed linear dependence on the tube length. Zhao et al.[14] found that a steep rocking motion of the walls in short DWCNTs oscillators (with length no greater than 3 nm) would appear if the inner wall was pulled about one-third out of the outer wall. In recent years, several research reports have focused on stability and tunability of the oscillators. For instance, Neild et al.[15] suggested applying periodic forces on the sliding core tube to overcome the damping, which would decrease the amplitude of the oscillators along with an increase in the time. Kang et al.[16] proposed a MD simulation model for a bitube oscillator. In their model, the inner tube oscillated between two outer tubes. The frequency of CNT oscillators can be adjusted by changing the gap between the two outer walls.

In all the simulations mentioned above, the inner tube was in a translational motion in oscillation, and the oscillation of the MWCNT was assumed to be triggered initially by pulling the inner tube partly out of the outer wall(s). Considering the inner tube as a component of the system (i.e., DWCNT), the motion of the inner tube may be either a translational motion (oscillation) or a rotating motion along its axis in the outer tube. Recently, Král and Sadeghpour[17] proposed to spin nanotubes with circularly polarized light, further motivating the use of nanotubes as possible axes of rotation at the nanoscale. Zhang et al.[18] carried out nonequilibrium molecular dynamics simulations to calculate the energy dissipation rate during the rotational motion in the presence of an external thermostat. These methods have been widely used to study various mechanical behaviors of CNTs.[2,18−21] When the inner tube rotates within a fixed outer tube, it is still an open question whether the speed of rotation will decrease because of the friction between the two walls. It is known that any asymmetrical boundary condition may lead to a difference in van der Waals force at the two ends. It is expected that if the outer tube remains partly or wholly fixed, the inner tube will oscillate while rotating. Especially, the oscillator with the stable amplitude attracts our attention. Considering the work reported by Fennimore et al.[22] and Bourlon et al.,[23] this kind of rotation of the tubes can be achieved experimentally. Such an oscillator therefore has great potential for future applications in NEMS. Determining the rules of axial oscillation of a bitube with a rotating inner tube is the motivation of this study.

Molecular Dynamics Simulation. In this study, we use a MD simulation approach to investigate rotational DWCNT-bearing systems. For the model of CNTs, the adaptive intermolecular reactive empirical bond order (AIREBO) potential[24] is used, which can describe both the covalent bond between carbon atoms and the long-range van der Waals interactions for the force field. The time step for all simulations is chosen to be 1 fs.

The internal conformation of each tube in the DWCNT system is characterized by the chiral indices (n,m). To keep the radii of both the inner and outer tubes approximately equal among the cases, the four simulation modes shown in Figure 1 are adopted, namely, two commensurate systems, (15,0)/(24,0) and (9,9)/(14,14), and two incommensurate systems, (13,5)/(20,7) and (9,9)/(24,0). The distance between the inner and outer walls in all four DWCNTs is nearly 0.335 nm, which is the common equilibrium distance between adjacent sheets in graphite. The geometric parameters are given in Table 1.

In all four modes, the length of the inner tube is around 6 nm and the length of the outer tube is around 5 nm (see Table 1). Initially, the two tubes in each mode have a symmetrical layout along the axis. At the beginning of the simulation, the whole system, except for the atoms within 0.5 nm near the right-hand end of the outer tube (yellow atoms along the axis in Figure 1), which is fixed, is specified by a Nosé−Hoover heat bath (the Nosé−Hoover thermostat is a deterministic method used in molecular dynamics to maintain temperature around an average, while the heat bath is made an integral part of the system by adding an artificial variable associated with an artificial mass) at 300 K for relaxation. After 20 ps of relaxation, the whole inner tube is set to rotate with a period of 4 ps under constant temperature (the microcanonical *NVT* ensemble, where N is the total number of particles in the system, V is the

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system's volume, and T is the absolute temperature; T = 300 K in this analysis). After 20 ps rotation (i.e., 5 rounds), we vary the whole system under a constant energy condition (the microcanonical *NVE* ensemble, where *NV* is the same as above and E is the total energy in the system) and let the inner tube rotate freely. The right-hand end of the outer tube remains fixed. Under this condition, the axial oscillatory behavior of the inner tube is investigated. The simulation is carried out on a computer with 6-core (2.0 GHz) and 32 GB RAM.

Results and Discussion. Figure 2 shows the angular velocities of the inner tubes in the four modes shown in Figure 1. The rotational speeds decrease monotonously with time. For example, for the oblique chiral bitube mode [(13,5)/(20,7)], the rotational speed of inner tube decreases nonlinearly with respect to time. For the remaining modes, the rotational speeds decrease linearly at different dissipation rates. For a model under *NVE* ensemble, the total energy remains constant. Therefore, part of the variation of the kinetic energy of its inner tube has been transferred into the phonon kinetic energy of two tubes because of friction between the two tubes in the DWCNT.[2,18,25] It implies that temperature increases (Figure 3) with the decrease of rotating speed of inner tubes. Besides, Guo et al.[25] discovered that the friction force between walls depends on both contact area and contact edge length. In fact, the gap between two walls in a bitube system has a significant effect on friction. Table 1 shows that the gap (0.357 nm) between (15,0) and (24,0) is the largest one among the four different DWCNTs. Small gap means stronger interaction between two walls in a bitube system. Therefore, the energy dissipation for the (15,0)/(24,0) zigzag/zigzag system is the smallest one. We can also find that the rotating speed of (13,5)/(20,7) (blue line in Figure 2) or the energy dissipation of the inner tube (Figure 3c) is very steep because of the smallest gap (0.325 nm) between two walls.

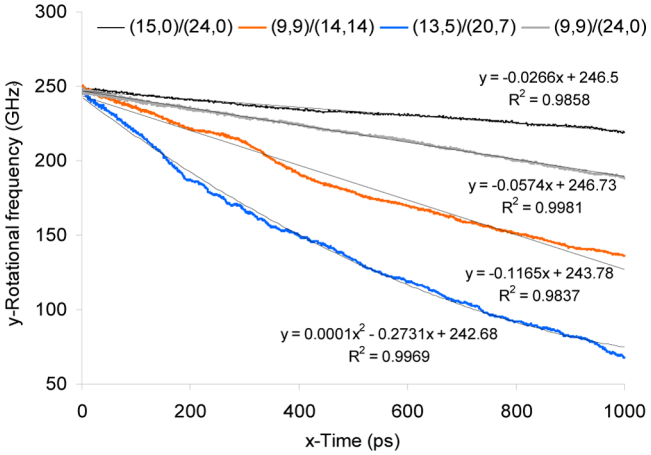

Figure 2. Histories of rotational frequencies of the inner tubes in simulation.

Checking the energy transition and temperature variation process (see Figure 3), we find that the energy of the inner tube in each mode decreases continuously after around 100 ps. The result shown in Figure 3 also implies that the energy dissipation rate of the inner tube in the commensurate model [(15,0)/(24,0)] is the lowest among the four cases. This is not identical to the result reported by Guo et al.,[1] in which the incommensurate system (5,5)/(18,0) was found to have a

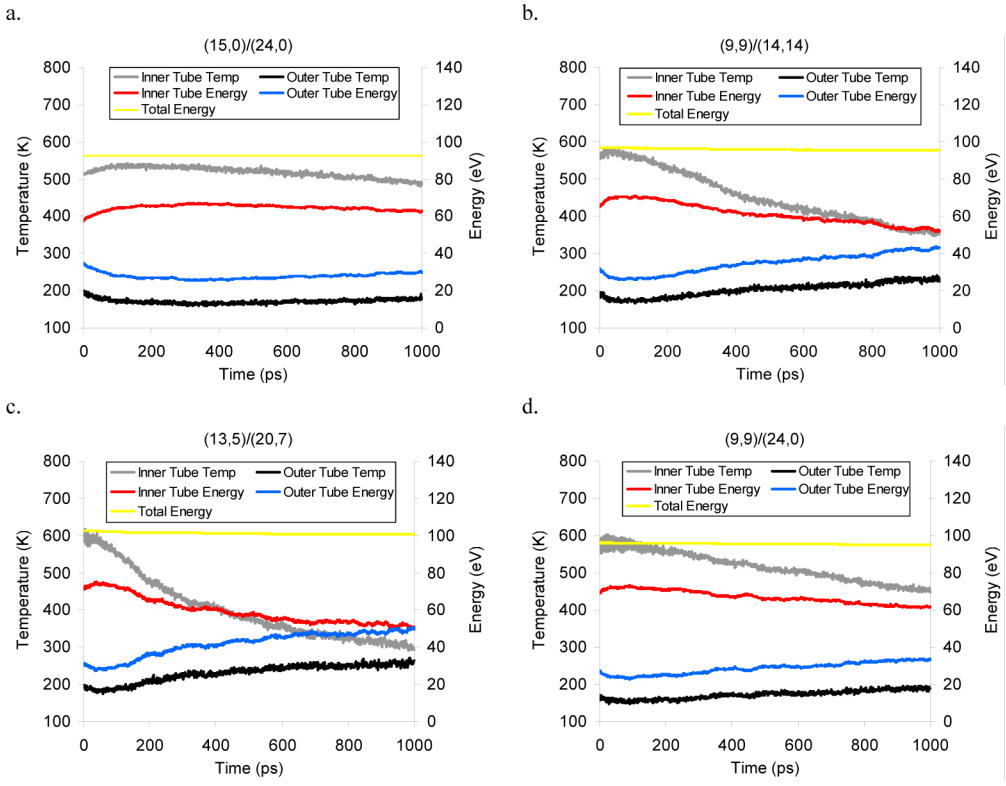

Figure 3. Transition histories of temperature and energy (including kinetic energy and potential energy) of bitubes; gray line represents the temperature of the inner tube; black line the temperature of the outer tube; red line the energy of the inner tube; blue line the energy of the outer tube; and yellow line the total energy of the system.

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lower rate of energy dissipation than the commensurate model with translational motion. The contradictory conclusions may be mainly caused by four reasons besides chirality difference. First, the potential function used is different: AMBER force field is employed by Guo et al.,[1] while AIREBO is adopted in the present work. Second, boundary conditions are different. For example, there are only 3 atoms on the outer tube that are fixed in Guo's simulation, while there are tens of atoms on the right-hand end of the outer tube that are fixed in the present simulations. Third, the motion types are different; for example, there is only axial translational motion of inner tubes in Guo's simulations. The final reason is that the length of the bitube system in Guo's work is 3 nm, which is only half the length of the inner tube in the present modes.

For the first, second, and fourth cases, both the temperature and the energy of the inner tubes decay linearly with time. This conclusion corresponds well with that reported by Zhu et al.[19] For the inner tube in the (13,5)/(20,7) mode, however, either its energy or its temperature decays nonlinearly with respect to time. At the same time, its decay rate is the highest among the four cases. Therefore, an oblique chiral bitube is unsuitable as an oscillator because of its steep energy dissipation with smaller gap between two tubes.

The translational motion causes oscillation if the velocity direction of the mass center of the inner tube (MCIT) varies with time. Figure 4 shows the oscillation histories of the

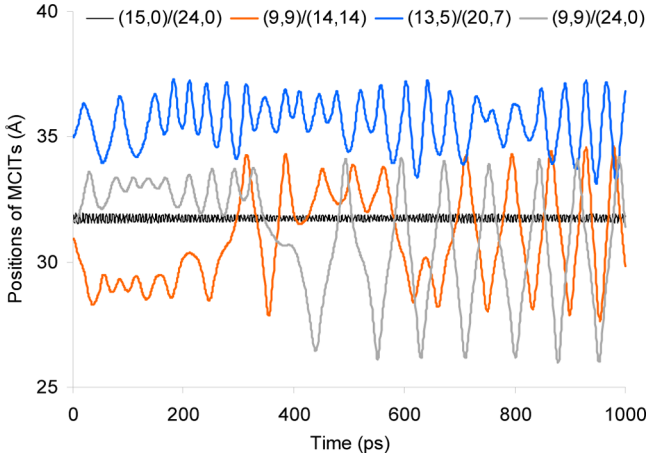

Figure 4. Histories of position of mass center of inner tubes (MCITs) in simulation.

MCITs. Although there is no initial axial load on the inner tubes, the MCITs move under asymmetrical absorption of the ends of tubes (van der Waals force) as the inner tubes rotate. The amplitude of the oscillation of the MCITs increases in the first 300 ps. Subsequently, the amplitude tends to be stable. Therefore, the oscillation of the inner tube found here is different from that in previous studies[1,6,7,12,13] using gigahertz oscillators, in which an external force was applied to pull the inner tube partly out of the outer tube to trigger oscillation. We describe the present oscillation of the inner tube as *a self-excited oscillation*.

For the (13,5)/(20,7) mode, the oscillation is still unstable after 650 ps, and the equilibrium position of the inner tube is not identical to its initial position. In the remaining three cases, the oscillation tends to be metastable and the initial and equilibrium positions of the inner tube match well after 650 ps (Figure 5). Figure 6 shows five configurations of the (9,9)/

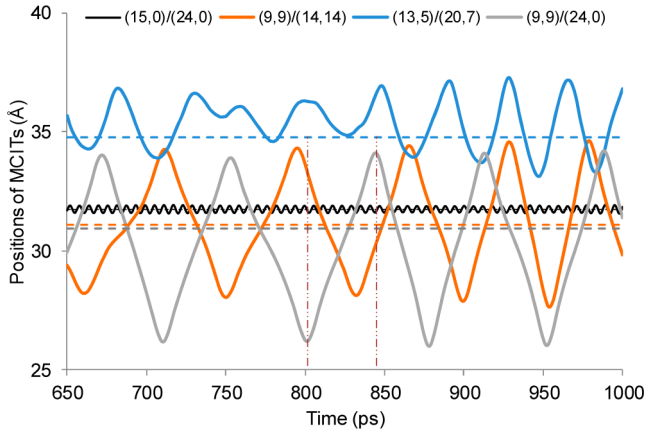

Figure 5. Histories of position of MCITs from 650 to 1000 ps; the broken lines show the MCIT initial positions (broken lines represent the equilibrium position of MCITs).

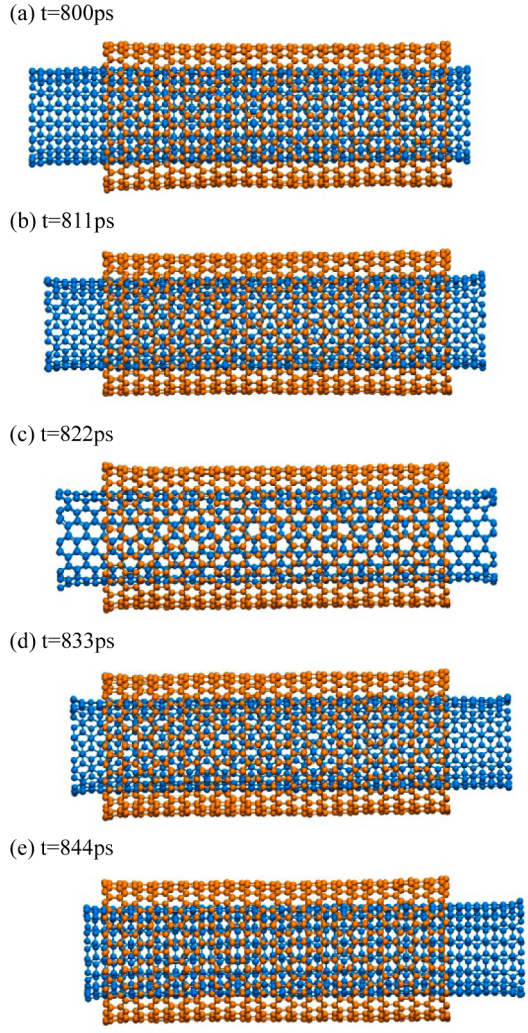

Figure 6. Configurations of model (9,9)/(24,0) from 800 to 844 ps (the ends of both walls protrude outward).

(24,0) mode from the valley (t = 800 ps) to the peak (t = 844 ps) with a rotating inner tube. It is found that the maximum amplitude does not exceed half the length difference between the two tubes. The reason is that the repulsive interaction exists when the distance between the two left-hand (Figure 6e) or

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two right-hand ends (Figure 6a) of the two tubes is too small. Simultaneously, the other ends attract each other. As the repulsion is not identical to the attraction between the two tubes, the motion of the inner tube is driven by the higher one.

Considering the amplitudes and frequencies of the inner tubes, we find that the amplitude of the (15,0)/(24,0) mode is the lowest (0.148 Å in Table 2) but the frequency (166.67 GHz = 0.16667 THz) is the highest among the four modes. For the fourth (9,9)/(24,0) mode, the amplitude (i.e., 3.970 Å) is the highest, but the frequency (12.66 GHz) is the lowest among the four cases. Obviously, the major difference between the first and the fourth case is the chirality of the inner tubes. We conclude that a zigzag/zigzag bitube with larger gap will produce a high frequency (THz) self-excited oscillator, and an armchair/zigzag bitube with smaller gap will behave like a self-excited oscillator with high amplitude.

Table 2. Average Amplitudes and Frequencies of MCITs after 650 ps

bitube	gap between walls (nm)	average amplitude (Å)	frequency (GHz)
(15,0)/(24,0) (Figure 1a)	0.357	0.148	166.67
(9,9)/(14,14) (Figure 1b)	0.344	3.232	14.98
(13,5)/(20,7) (Figure 1c)	0.325	1.764	25.42
(9,9)/(24,0) (Figure 1d)	0.334	3.970	12.66

Concluding Remarks. Using four modes of DWCNTs with the appropriate geometrical configurations, their self-excited oscillatory behavior is founded, and some conclusions are drawn as follows:

(a) Due to triggers by the attraction at the ends of the outer tube as well as high-speed rotating of the tubes, the oscillation of the inner tube of a DWCNT is present.

(b) After several rounds of rotation, the amplitude of the oscillation of a MCIT increases until it reaches a maximum value and then it remains stable. It means that the damping effect caused by friction is removed by absorption of rotating kinetic energy of the inner tube. The maximum amplitude does not exceed half the length difference between the inner and outer tube.

(c) The energy dissipation rate of the inner tube is sensitive to the gap between two walls in a bitube system. Smaller gap leads to larger energy dissipation rate.

(d) A zigzag/zigzag bitube can act as a high-frequency (terahertz) self-excited oscillator, and an armchair/zigzag bitube can act as a self-excited oscillator with high amplitude. A bitube with smaller gap between two tubes is unsuitable as an oscillator because of its steep energy dissipation.

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Notes

The authors declare no competing financial interest.

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