Molecular Motor of Double-Walled Carbon Nanotube Driven by Temperature Variation

Zhan-chun Tu\textsuperscript{1,2,} and Zhong-can Ou-Yang\textsuperscript{1,3}

\textsuperscript{1}Institute of Theoretical Physics, The Chinese Academy of Sciences, P.O.Box 2735 Beijing 100080, China
\textsuperscript{2}Graduate School, The Chinese Academy of Sciences, Beijing, China
\textsuperscript{3}Center for Advanced Study, Tsinghua University, Beijing 100084, China

Abstract

An elegant formula for coordinates of carbon atoms in a unit cell of a single-walled nanotube (SWNT) is presented and a new molecular motor of double-walled carbon nanotube whose inner tube is a long (8,4) SWNT and outer tube a short (14,8) SWNT is constructed. The interaction between inner and outer tubes is analytically derived by summing the Lennard-Jones potentials between atoms in inner and outer tubes. It is proved that the molecular motor in a thermal bath exhibits a directional motion with the temperature variation of the bath.

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It is well known that a pollen in water exhibits Brownian motion. The forces on the pollen stem from two components [1]: A fluctuating force that averages to zero over time, and a viscous force that slows the motion. These two kinds of forces are related by temperature so the fluctuation is often called thermal noise. The second law of thermodynamics suggests that biased Brownian motion requires two conditions [2]: (1) breaking of thermal equilibrium and (2) breaking of spatial inversion symmetry. In order to illustrate the second law of thermodynamics can not be violated, Feynman devised an imaginary ratchet system with a pawl in his famous lecture [3].

To explain the matter transport in biological systems, the concept of molecular motor is introduced [4, 5]. From the statistical viewpoint, many models [2] of molecular motor were put forward, such as on-off ratchets [6, 7], fluctuating potential ratchets [1, 8], fluctuating force ratchets [9, 10], temperature ratchets [11, 12, 13] and so on. All those models satisfy the two conditions required by the second law of thermodynamics.

With the development of nanotechnology, especially the discovery of carbon nanotubes [14], people are putting their dream of manufacturing nanodevice [15] into practice. Here a natural question arises: Can we construct molecular motors from carbon nanotubes? In this Letter, we will prove that it is possible to construct a molecular motor from a double-walled carbon nanotube (DWNT). The molecular motor in a thermal bath exhibits a directional motion with the temperature variation of the bath.

A DWNT consists of two single-walled carbon nanotubes (SWNT’s) with a common axis. The layer distance between the two tubes is about 3.4 Å. A SWNT without two end caps can be constructed by wrapping up a single sheet of graphite such that two equivalent sites of hexagonal lattice coincide [16]. To describe the SWNT, some characteristic vectors need to be introduced. As shown in Fig.1, the chiral vector \( \mathbf{C}_h \), which defines the relative location of two carbon atoms, is specified by a pair of integers \((n_1, n_2)\) which is called the index of the SWNT. We have \( \mathbf{C}_h = n_1\mathbf{a}_1 + n_2\mathbf{a}_2 \) with \( \mathbf{a}_1 \) and \( \mathbf{a}_2 \) being two unit vectors of graphite. The translational vector \( \mathbf{T} \) corresponds to the first lattice point of 2D graphitic sheet through which the line normal to the chiral vector \( \mathbf{C}_h \) passes. The unit cell of the SWNT is the rectangle defined by vectors \( \mathbf{C}_h \) and \( \mathbf{T} \), while vectors \( \mathbf{a}_1 \) and \( \mathbf{a}_2 \) define the area of the unit cell of 2D graphite. The number \( N \) of hexagons per unit cell of the SWNT is obtained as a function of \( n_1 \) and \( n_2 \) as \( N = 2(n_1^2 + n_2^2 + n_1n_2)/d_R \), where \( d_R \) is the greatest common divisor of \((2n_2 + n_1)\) and \((2n_1 + n_2)\). There are \( 2N \) carbon atoms in each unit cell of the SWNT.
because each hexagon contains two atoms. To denote the $2N$ atoms, we use a symmetry vector $\mathbf{R}$ to generate coordinates of carbon atoms in the nanotube; it is defined as the site vector having the smallest component in the direction of $\mathbf{C}_h$. From a geometric standpoint, vector $\mathbf{R}$ consists of a rotation around the nanotube axis by an angle $\Psi = 2\pi/N$ combined with a translation $\tau$ in the direction of $\mathbf{T}$; therefore, $\mathbf{R}$ can be denoted by $\mathbf{R} = (\Psi |\tau)$. Using the symmetry vector $\mathbf{R}$, we can divide the $2N$ carbon atoms in the unit cell of SWNT into two classes: one includes $N$ atoms whose site vectors satisfy:

$$A_l = l\mathbf{R} - [l\mathbf{R} \cdot \mathbf{T}/T^2]\mathbf{T} \quad (l = 0, 1, 2, \ldots, N - 1),$$

another includes the remainder $N$ atoms whose site vectors satisfy:

$$B_l = l\mathbf{R} + \mathbf{B}_0 - \left[\frac{(l\mathbf{R} + \mathbf{B}_0) \cdot \mathbf{T}}{T^2}\right]\mathbf{T}$$

$$-\left[\frac{(l\mathbf{R} + \mathbf{B}_0) \cdot \mathbf{C}_h}{C_h^2}\right]\mathbf{C}_h \quad (l = 0, 1, 2, \ldots, N - 1),$$

where $\mathbf{B}_0$ represents one of the nearest-neighboring atoms of $\mathbf{A}_0$. In and only in the above two equations, $[\cdots]$ denotes the Gaussian function, e.g., $[5.3] = 5$.

We construct the molecular motor of double-walled carbon nanotube as shown in Fig.2. The inner tube’s index is (8, 4) with a length long enough to be regarded as infinite. The outer tube is set to be a (14, 8) tube with just a single unit cell [18]. Obviously, they are both chiral nanotubes and their layer distance is about 3.4 Å. If we prohibit the motion of the outer tube in the direction of nanotube axis, it will be proved that this system in a thermal bath exhibits a directional rotation when the temperature of the system varies with time. Thus it could serve as a thermal ratchet.

To see this, we first select an orthogonal coordinate system shown in Fig.2 whose $z$-axis is the tube axis and $x$-axis passes through carbon atom $A_0$ of the inner tube. We fix the inner tube and forbid the $z$-directional motion of the outer tube, that is, carbon atom $A_0$ of outer tube is always in the $xy$-plane. We denote the angle rotated by the outer tube around the inner tube to be $\theta$.

We take the interaction between atoms in the outer tube and the inner tube as the Lennard-Jones potential $u(r_{ij}) = 4\epsilon [(\sigma/r_{ij})^{12} - (\sigma/r_{ij})^6]$, where $r_{ij}$ is the distance between atom $i$ in the inner tube and atom $j$ in the outer tube, $\epsilon = 28$ K, and $\sigma = 3.4\text{Å}$ [13]. We calculate the potential $V(\theta)$ [17] when the outer tube rotates around the inner tube with angle $\theta$ and plot it in Fig.3. We find that $V(\theta)$ is periodic (with period $\pi/2$) and violates
the spatial inversion symmetry. Thus the second condition to make a molecular motor is satisfied. Up to now we can explain why we select (8,4) tube and (14,8) tube. The following three criteria are considered: (1) layer distance is about 3.4 Å; (2) the shape of $V(\theta)$ is not too complicated; (3) the difference of maximum and minimum of $V(\theta)$ is remarkable. We find that only (8,4) and (14,8) tube satisfy these criteria for $0 < n_1, n_2 < 30$ through our calculation.

The easiest way to satisfy the first condition is to put our system into a thermal bath full of He gas whose temperature varies with time. We will show that the outer tube will exhibit a directional rotation.

We can write the Langevin equation [20] for the outer tube

$$m\rho^2\ddot{\theta} = -V'(\theta) - \eta\dot{\theta} + \xi(t),$$

where $m$ and $\rho$ are respectively the mass and the radius of the outer tube, $\eta$ is the rotating viscous coefficient [21], and dot and prime indicate, respectively, differentiations with respect to time $t$ and angle $\theta$. $\xi(t)$ is thermal noise which satisfies $\langle \xi(t) \rangle = 0$ and the fluctuation-dissipation relation $\langle \xi(t)\xi(s) \rangle = 2\eta T(t)\delta(t-s)$ [22], where $T(t)$ is temperature and the Boltzmann factor is set to 1. Let us consider the overdamped case that the inertial term $m\rho^2\ddot{\theta}$ is much less than thermal fluctuations and can be neglected. Thus we arrive at

$$\eta\dot{\theta} = -V'(\theta) + \xi(t)$$

and the corresponding Fokker-Planck equation [2, 23]

$$\frac{\partial P(\theta,t)}{\partial t} = \frac{\partial}{\partial \theta} \left[ \frac{V'(\theta)P(\theta,t)}{\eta} \right] + \frac{T(t)}{\eta} \frac{\partial^2 P(\theta,t)}{\partial \theta^2},$$

where $P(\theta,t)$ represents the probability of finding the outer tube in angle $\theta$ at time $t$ which satisfies $P(\theta + \pi/2, t) = P(\theta, t)$. If the period of temperature variation is $T$, we arrive at the average angular velocity in the long-time limit [2]

$$\langle \dot{\theta} \rangle = \lim_{t \to \infty} \frac{1}{T} \int_t^{t+T} dt \int_0^{\pi/2} d\theta \left[ \frac{V'(\theta)P(\theta,t)}{\eta} \right].$$

For example, set $T(t) = \bar{T}[1 + A\sin(2\pi t/\bar{T})]$ with $\bar{T} = 50$ K and $|A| \ll 1$. Let $D = \eta/\bar{T}$, $t = D\tau$, $U(\theta) = V(\theta)/\bar{T}$, $T = D\mathcal{J}$ and $\bar{P}(\theta,\tau) = P(\theta,D\tau)$, We arrive at the dimensionless equations of Eqs.(3) and (4)

$$\frac{\partial \bar{P}(\theta,\tau)}{\partial \tau} = \frac{\partial}{\partial \theta} \left[ \frac{U'(\theta)\bar{P}}{\mathcal{J}} \right] + (1 + A\sin \frac{2\pi \tau}{\mathcal{J}}) \frac{\partial^2 \bar{P}}{\partial \theta^2},$$

$$\langle \frac{d\theta}{d\tau} \rangle = \lim_{\tau \to \infty} \frac{1}{\mathcal{J}} \int_\tau^{\tau+\mathcal{J}} d\tau \int_0^{\pi/2} d\theta \left[ -U'(\theta)\bar{P} \right].$$

Let $A = 0.01$, we can numerically solve Eq.(5) and calculate Eq.(6) with different $\mathcal{J}$. The solid line in Fig.4 shows the relation between the average dimensionless angular velocity $\langle \frac{d\theta}{d\tau} \rangle$
and the dimensionless period $J$ of temperature variation. We find that $\langle \frac{d\theta}{d\tau} \rangle \simeq 0$ for very small and large $J$, and $\langle \frac{d\theta}{d\tau} \rangle \neq 0$ for middle $J$, which implies that the outer tube has a evident directional rotation in this period range. Thus we have constructed a temperature ratchet.

For He gas at temperature $\bar{T} = 50$ K, we can calculate $\eta = 861$ Kns from its value at $273$ K [21, 24] and $D = 17.2$ ns. Based on these data, we obtain $\langle \frac{d\theta}{d\tau} \rangle = -43$ nrad when $J = 0.17$, i.e. $\langle \dot{\theta} \rangle = \frac{1}{T} \langle \frac{d\theta}{d\tau} \rangle = -2.5$ nrad/ns when $T = 2.9$ ns. Here the minus sign of average angular velocity means that the rotation of the outer tube around $z$-axis is left-handed. We notice that $\langle \dot{\theta} \rangle = -2.5$ nrad/ns is a remarkable value (-2.5 rad/s) that is easy to be observed from experiment. If consider the inertial effect of the outer tube, this value may be changed. But we believe that it is still an observable quantity.

Furthermore, from Fig. 4 we observe that the sign of $\langle \frac{d\theta}{d\tau} \rangle$ changes from “+” to “−” and back to “+” with $J$ increasing, which suggests that the outer tube’s rotation around the inner tube is the right-handed for small and large period $J$, and the left-handed for middle period $J$. Through detailed analysis, we find that $\langle \frac{d\theta}{d\tau} \rangle$ is proportional to $J^3$ for very small period $J$ and proportional to $J^{-2}$ for period $J$ large enough, which agrees with the results of asymptotic analysis [2, 25].

To summarize, we have theoretically constructed a temperature ratchet using a DWNT, but some practical difficulties are remained: How to synthesize the proper DWNT? How to forbid the axial translation of outer tube? If these obstacles are ruled out, molecular motor of DWNT must be reality.

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* Email address: tzc@itp.ac.cn

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FIG. 1: The unrolled honeycomb lattice of a SWNT. By rolling up the sheet such that the point $A_0$ coincides with the point corresponding to chiral vector $C_h$, a nanotube is formed. Vectors $a_1$ and $a_2$ are real space unit vectors of the hexagonal lattice. The translational vector $T$ is perpendicular to $C_h$ and runs in the direction of tube axis. The vector $R$ is the symmetry vector. $A_0$, $B_0$ and $A_l, B_l (l = 1, 2, \ldots, N)$ are used to denote the sites of carbon atoms.

FIG. 2: A double-walled carbon nanotube with the inner tube’s index being (8, 4) and the outer tube’s index being (14, 8). $z$-axis is the tube axis, $x$-axis perpendicular to $z$ passes through carbon atom $A_0$ (see Fig[I]) of the inner tube and $y$-axis is perpendicular to the $xz$-plane. There is no obviously relative motion along radial direction between the inner and the outer tubes at low temperature. If we forbid the motion of the outer tube in the direction of $z$-axis, only the rotation of the outer tube around the inner tube is permitted.
FIG. 3: The potentials $V(\theta)$ between the outer and the inner tubes with the outer tube rotating around the inner tube. $\theta$ is the rotating angle. Here we have set $V(0) = 0$. The squares are the numerical results which can be well fitted by $V(\theta) = 15.7 - 0.6 \cos 4\theta - 2.2 \sin 4\theta - 12.7 \cos 8\theta - 6 \sin 8\theta - 1.7 \cos 12\theta + 10.8 \sin 12\theta$ (solid curve).

FIG. 4: The average dimensionless angular velocity $\langle d\theta/d\tau \rangle$ of the outer tube rotating around the inner tube in thermal bath whose temperature changes with the dimensionless period $\mathcal{J}$. The minus sign means the rotation around $z$-axis is the left-handed.