Periodic one-dimensional hopping model with transitions between nonadjacent states

Yunxin Zhang*

Shanghai Key Laboratory for Contemporary Applied Mathematics, Centre for Computational Systems Biology, School of Mathematical Sciences, Fudan University, Shanghai 200433, China

(Received 24 April 2011; published 1 September 2011)

A one-dimensional hopping model is useful for describing the motion of microscopic particles in a thermal noise environment. Recent experiments on the new generation of light-driven rotary molecular motors found that a motor in state i can jump forward to state $i+1$ or $i+2$ or backward to state $i-1$ or $i-2$ directly. In this paper, inspired by these experiments, such a modified periodic one-dimensional hopping model with arbitrary period N is studied theoretically. The mean velocity, effective diffusion constant, and mean dwell time in one single mechanochemical cycle are obtained. The corresponding results are illustrated and verified by being applied to the synthetic rotary molecular motors.

DOI: 10.1103/PhysRevE.84.031104 PACS number(s): 02.50.Ey, 82.20.−w, 87.16.Nn

I. INTRODUCTION

Many physical [1,2] and biochemical processes, for example, the motion of motor proteins kinesin, dynein, and myosin [3–5], can be well described by periodic one-dimensional hopping models [6,7]. So far, these models have been extensively studied, and the mean velocity, effective diffusion constant, and mean first passage time have been obtained [8–11]. In these models, a particle in state i can jump forward to state $i+1$ with rate u_i or backward to state $i-1$ with rate w_i. Where the forward and backward transition rates u_i and w_i satisfy periodic conditions $u_{N+i}=u_i$ and $w_{N+i}=w_i$, where N is the number of states in one cycle. Meanwhile, for some special problems, this simple hopping model has been generalized in some aspects [12].

Recently, experimental data of a type of synthetic light-driven rotary molecular motor, which was devised by Feringa and coworkers, found that the motor in state i can jump forward to state $i+2$ or backward to state $i-2$ directly [13–16]. In fact, it also has been found that, for many molecular motors [17–21], there are usually more than two ways for the motor to leave its present state. Therefore, it is necessary to study more general one-dimensional hopping models, in which the particle in state i is allowed to jump to other states besides $i-1$ and $i+1$.

In this paper, one of the simple cases, which we call the *modified* periodic one-dimensional hopping model, during which, in addition to states $i-1$ and $i+1$, the particle in state i also can jump forward to state $i+2$ with rate u_i', or backward to state $i-2$ with rate w_i', is theoretically analyzed. Using an idea similar to Derrida's [8], the mean velocity V and effective diffusion constant D are obtained. Meanwhile, by a method similar to the one in [9], the mean dwell time in one single cycle is also obtained. The reason that we discuss this simple case here is not only because it is theoretically convenient but also because the corresponding results can be verified by the experimental data [16]. Compared with the more general discussions in [22], the expressions of the effective diffusion constant D and mean dwell time $T_{\rm dwell}$ in one mechanochemical cycle are also provided, and then the effective forward and backward transition rates, $u_{\rm eff}$ and $w_{\rm eff}$, are obtained using the recent results [23], which are then employed to get the *rotational excess* of this modified one-dimensional hopping model, as discussed in detail in Sec. V. Moreover, the results obtained here can be applied directly to the recent experimental studies about synthetic rotary molecular motors [16].

This paper is organized as follows. The modified model is briefly introduced in the next section, and then in Secs. III and IV, the formulations of mean velocity, effective diffusion constant, and mean dwell time in one mechanochemical cycle are obtained. In Sec. V, the theoretical results are illustrated numerically and verified by experimental data of the rotary molecular motor. Finally, this paper is summarized in the last section.

II. ONE-DIMENSIONAL HOPPING MODEL WITH NONADJACENT STATE TRANSITION

Our modified one-dimensional hopping model of period N is schematically depicted in Fig. 1, in which u_i is the forward transition rate from states i to $i+1$, while u_i' is the forward transition rate from states i to $i+2$, and likewise for the backward transition rates w_i $(i\to i-1)$ and w_i' $(i\to i-2)$. Periodicity requires $u_{i+N}=u_i, u'_{i+N}=u_i', w_{i+N}=w_i, w'_{i+N}=w_i'$.

Similar to the analysis in [6,8], if $\tilde{p}_i(t)$ is the probability of finding the particle in state i at time t, then the master equation reads

$$\frac{d\tilde{p}_i(t)}{dt}=(u'_{i-2}\tilde{p}_{i-2}+u_{i-1}\tilde{p}_{i-1}+w_{i+1}\tilde{p}_{i+1}+w'_{i+2}\tilde{p}_{i+2})-(u_i+u_i'+w_i+w_i')\tilde{p}_i. \quad (1)$$

Let

$$\bar{p}_i=\sum_{k=-\infty}^{+\infty}\tilde{p}_{kN+i},\ \bar{s}_i=\sum_{k=-\infty}^{+\infty}(kN+i)\tilde{p}_{kN+i}; \quad (2)$$

then

$$\bar{p}_i=\bar{p}_{N+i},\quad \bar{s}_i=\bar{s}_{N+i},\quad \sum_{i=1}^{N}\bar{p}_i=\sum_{i=-\infty}^{+\infty}\tilde{p}_i=1, \quad (3)$$

*xyz@fudan.edu.cn

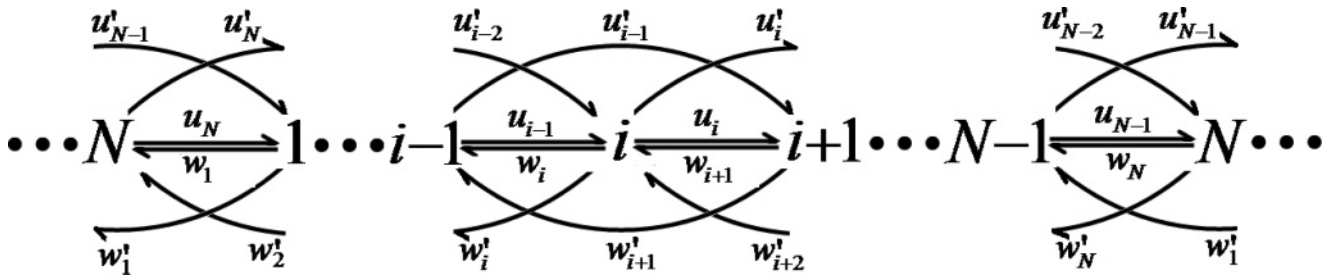

FIG. 1. Schematic depiction of the modified one-dimensional hopping model of period N, in which the particle in state i can jump forward to state $i+1$ with rate u_i or to state $i+2$ with rate u'_i or jump backward to state $i-1$ with rate w_i or to state $i-2$ with rate w'_i. The rates u_i, u'_i, w_i, and w'_i satisfy the periodicity conditions $u_{i+N} = u_i, u'_{i+N} = u'_i, w_{i+N} = w_i$, and $w'_{i+N} = w_i$.

and $\bar{p}_i, \bar{s}_i$ satisfy

$$\frac{d\bar{p}_i(t)}{dt} = (u'_{i-2}\bar{p}_{i-2} + u_{i-1}\bar{p}_{i-1} + w_{i+1}\bar{p}_{i+1} + w'_{i+2}\bar{p}_{i+2}) - (u_i + u'_i + w_i + w'_i)\bar{p}_i, \tag{4}$$

$$\frac{d\bar{s}_i(t)}{dt} = (u'_{i-2}\bar{s}_{i-2} + u_{i-1}\bar{s}_{i-1} + w_{i+1}\bar{s}_{i+1} + w'_{i+2}\bar{s}_{i+2}) - (u_i + u'_i + w_i + w'_i)\bar{s}_i + 2u'_{i-2}\bar{p}_{i-2} + u_{i-1}\bar{p}_{i-1} - w_{i+1}\bar{p}_{i+1} - 2w'_{i+2}\bar{p}_{i+2}. \tag{5}$$

In the long time limit, we assume, as in [8], that

$$\bar{p}_i(t) \to p_i, \qquad \bar{s}_i(t) \to a_i t + b_i, \tag{6}$$

where p_i, a_i, b_i are constants to be determined (see the Appendix).

III. MEAN VELOCITY AND EFFECTIVE DIFFUSION CONSTANT

Mathematically, the definitions of the mean velocity V and effective diffusion constant D are as follows (see [8,24]):

$$V = \lim_{t\to\infty} \frac{d\langle x(t)\rangle}{dt}, \quad D = \frac{1}{2}\lim_{t\to\infty}\frac{d}{dt}[\langle x^2(t)\rangle - \langle x(t)\rangle^2], \tag{7}$$

where $\langle x^k(t)\rangle := \sum_{i=-\infty}^{\infty} i^k \tilde{p}_i(t)$ is the average kth moment of the particle position at time t. One can easily show that

$$\begin{aligned} V &= \lim_{t\to\infty}\frac{d\langle x(t)\rangle}{dt} = \lim_{t\to\infty}\sum_{i=1}^{N}\frac{d\bar{s}_i}{dt} \\ &= \lim_{t\to\infty}\sum_{i=1}^{N}(2u'_{i-2}\bar{p}_{i-2} + u_{i-1}\bar{p}_{i-1} - w_{i+1}\bar{p}_{i+1} - 2w'_{i+2}\bar{p}_{i+2}) \\ &= \sum_{i=1}^{N}(2u'_{i-2}p_{i-2} + u_{i-1}p_{i-1} - w_{i+1}p_{i+1} - 2w'_{i+2}p_{i+2}), \end{aligned} \tag{8}$$

where p_i is the long time limit of $\bar{p}_i$ [see (2) and (6)], and it can be obtained by (A1) or (A5). Meanwhile,

$$\begin{aligned} \frac{d\langle x^2(t)\rangle}{dt} &= \sum_{i=1}^{N}\sum_{k=-\infty}^{\infty}(kN+i)^2[u'_{i-2}\tilde{p}_{kN+i-2} + u_{i-1}\tilde{p}_{kN+i-1} + w_{i+1}\tilde{p}_{kN+i+1} + w'_{i+2}\tilde{p}_{kN+i+2} \\ &\quad - (u_i + u'_i + w_i + w'_i)\tilde{p}_{kN+i}] \\ &= \sum_{i=1}^{N}[(4u'_i + 2u_i - 2w_i - 4w'_i)\bar{s}_i \\ &\quad + (4u'_i + u_i + w_i + 4w'_i)\bar{p}_i]. \end{aligned} \tag{9}$$

Combining Eqs. (6)–(9), we have

$$\begin{aligned} D &= \sum_{i=1}^{N}[(2u'_i + u_i - w_i - 2w'_i)(a_i t + b_i) \\ &\quad + \frac{1}{2}(4u'_i + u_i + w_i + 4w'_i)p_i] - V\sum_{i=1}^{N}(a_i t + b_i) \\ &= \sum_{i=1}^{N}[(2u'_i + u_i - w_i - 2w'_i)b_i \\ &\quad + \frac{1}{2}(4u'_i + u_i + w_i + 4w'_i)p_i] - V\sum_{i=1}^{N} b_i, \end{aligned} \tag{10}$$

where $\vec{b} = (b_1, \ldots, b_N)^T$ satisfies (6). In numerical calculations, $\vec{b}$ can be replaced by any solutions of Eq. (A8) (for details, see the Appendix).

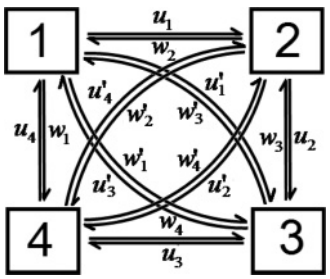

FIG. 2. Kinetic model of the synthetic rotary molecular motors recently devised by Feringa and coworkers [16]. The processes $1 \to 2$ and $3 \to 4$ correspond to photochemical conversions, while $2 \to 3$ and $4 \to 1$ correspond to subsequence thermal conversions. The rates u_2, w_3, u_4, and w_1 depend on the environmental temperature T, and the transition rates u_1, u_3, w_2, and w_4 and all u'_i and w'_i for $i = 1$–4 scale linearly with the light intensity l.

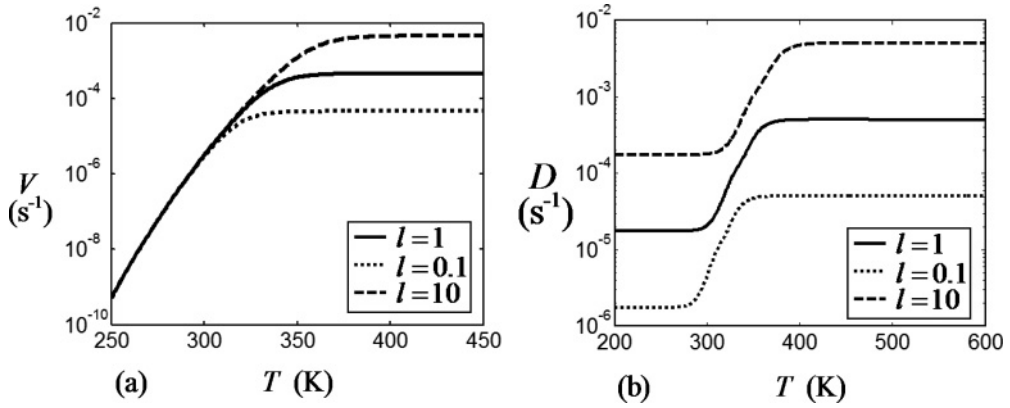

FIG. 3. (a) The mean velocity and (b) the effective diffusion constant of the synthetic rotary molecular motor, as depicted in Fig. 2, as functions of temperature T.

IV. MEAN DWELL TIME IN ONE SINGLE MECHANOCHEMICAL CYCLE

Let T_k be the mean first passage time (MFPT) of the particle starting at mechanochemical state k $(-N+1 \leqslant k \leqslant N-1)$ to reach the next mechanochemical cycle (backward or forward), i.e., to reach any of the states i with $|i-k| \geqslant N$. Then the mean dwell time T_{dwell} of the particle in a single mechanochemical cycle can be obtained as follows:

$$T_{\text{dwell}} = \sum_{k=0}^{N-1} p_k T_k. \tag{11}$$

The mean first passage time T_k can be obtained by the methods presented in [9,25]. For example, T_0 can be obtained as follows: Let $\bar{T}_i$ be the mean first passage time of the particle starting at state i $(-N+1 \leqslant i \leqslant N-1)$ to reach one of the absorbing boundaries $i = -N, -N-1, N, N+1$; then $\bar{T}_i$ is governed by

$$\bar{T}_i = \frac{1 + u_i \bar{T}_{i+1} + u'_i \bar{T}_{i+2} + w_i \bar{T}_{i-1} + w'_i \bar{T}_{i-2}}{u''_i + w''_i}, \tag{12}$$

in which $u''_i = u_i + u'_i$, $w''_i = w_i + w'_i$. $\bar{T}_i$ can be obtained by Eq. (12) and the absorbing boundary conditions

$$\bar{T}_i = 0, \quad i = -N, -N-1, N, N+1. \tag{13}$$

Consequently, the mean first passage time T_0 for the particle at state $k = 0$ to reach another mechanochemical cycle can be obtained by $T_0 = \bar{T}_0$. The mean first passage times T_k for $k = 1, \ldots, N-1$ can be obtained similarly. Finally, the mean dwell time of the particle in a single mechanochemical cycle can be obtained by formulation (11).

It can be verified that, if $u'_i = w'_i = 0$ for $1 \leqslant i \leqslant N$, i.e., the model is reduced to the usual periodic one-dimensional hopping model, then $T_{\text{dwell}} = T_0 = \cdots = T_{N-1}$.

V. NUMERICAL RESULTS

To illustrate and verify the methods to calculate the mean velocity V, effective diffusion constant D, and mean dwell time T_{dwell}, we discuss a class of synthetic rotary molecular motors here that was recently devised by Feringa and coworkers [13,16] and that can be schematically depicted by Fig. 2. The rotation of such molecular motors can be described by the modified one-dimensional hopping model with $N = 4$.

In numerical calculations, the parameters obtained by experiments [16] are used. The transition rates are as follows (with units of s^{-1}):

$$u_1 = 27.4 \times 10^{-5}, \quad u'_1 = 0.7 \times 10^{-5},$$
$$u_2 = \frac{k_B T}{h} e^{-\frac{\Delta G_{23}}{RT}}, \quad u'_2 = 0.35 \times 10^{-5},$$

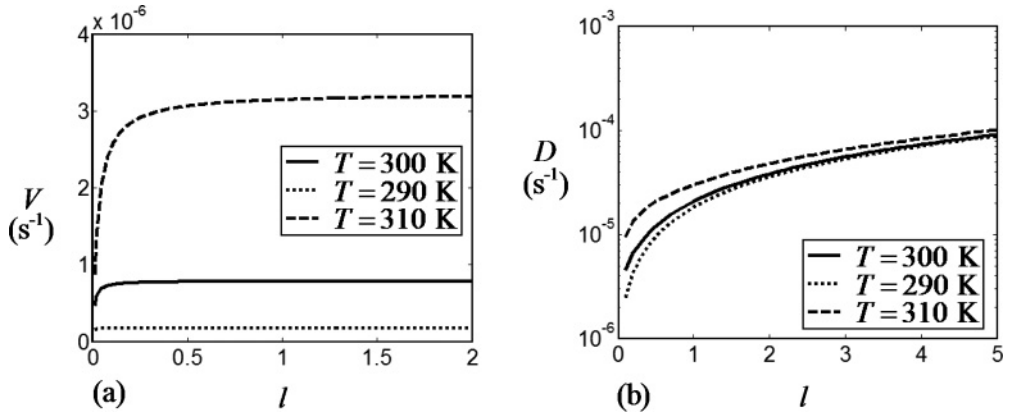

FIG. 4. (a) The mean velocity and (b) the effective diffusion constant of the synthetic rotary molecular motor, as schematically depicted in Fig. 2, as functions of light intensity l.

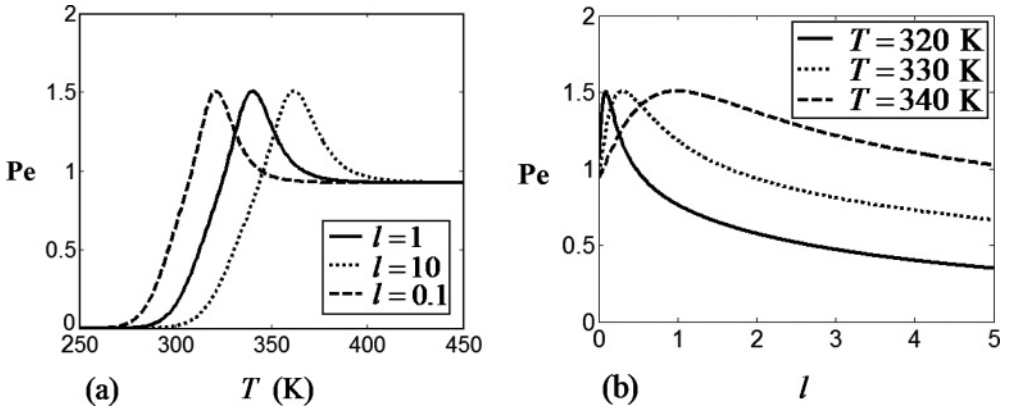

FIG. 5. The Péclet number $\mathrm{Pe} = V/D$ has a maximum as a function of both (a) temperature T and (b) light intensity l.

$$
\begin{aligned}
u_3 &= 20.6 \times 10^{-5}, \quad u_3' = 0.8 \times 10^{-5},\\
u_4 &= \frac{k_B T}{h} e^{-\frac{\Delta G_{41}}{RT}}, \quad u_4' = 0.55 \times 10^{-5},\\
w_1 &= \frac{k_B T}{h} e^{-\frac{\Delta G_{14}}{RT}}, \quad w_1' = 0.7 \times 10^{-5},\\
w_2 &= 2.1 \times 10^{-5}, \quad w_2' = 0.35 \times 10^{-5},\\
w_3 &= \frac{k_B T}{h} e^{-\frac{\Delta G_{32}}{RT}}, \quad w_3' = 0.8 \times 10^{-5},\\
w_4 &= 3.2 \times 10^{-5}, \quad w_4' = 0.55 \times 10^{-5}.
\end{aligned} \tag{14}
$$

Here $k_B = 1.38 \times 10^{-23}$ J K^{-1} is Boltzmann's constant, $h = 6.626 \times 10^{-34}$ J s is Planck's constant, $R = 1.9872$ cal K^{-1} mol^{-1} is a gas constant, and the empirical values ΔG_{ij} are (kcal mol^{-1})

$$
\begin{aligned}
\Delta G_{23} &= 25.6, \quad \Delta G_{32} = 30.3,\\
\Delta G_{41} &= 25.3, \quad \Delta G_{14} = 30.
\end{aligned} \tag{15}
$$

Since the processes $1 \to 2$ and $3 \to 4$ are photochemically induced conversions, the adjacent transition rates u_1, u_3, w_2, and w_4 and all the nonadjacent transition rates u_i' and w_i' for $i = 1$–4 scale linearly with the light intensity l. Illumination with $\lambda = 365$ nm corresponds to light intensity $l = 1$ (for details, see [16]).

The numerical results of the mean velocity and effective diffusion constant as functions of temperature T are plotted in Fig. 3. In the calculations, we assumed that the motor rotates $\pi/2$ in each of the four processes. The velocity $v = 1$ s^{-1} means the motor completes one quarter rotation per second, with a similar meaning for the effective diffusion constant D. Figures 3(a) and 3(b) are almost the same as the ones obtained in [16].[1] Note that in [16], different methods are used by Feringa and coworkers, which implies that our methods to get the mean velocity and effective diffusion constant of

[1]The values of velocity V versus temperature T [see Fig. 3(a)] are the same as the ones obtained in [16] [see Fig. 3(A) therein]. But for the effective diffusion constant D, the values obtained here [see Fig. 3(b)] are only half of the ones in [16] [see Fig. 3(B) therein]. The reason is that a factor of $1/2$ is included in our definition (7), but no factor of $1/2$ is used in the expression in [16] [see Eq. (9) therein or its related supporting information]. Consequently, the values of Pe [see Fig. 5(a)], defined as V/D, are two times bigger than the values obtained in [16] [see Fig. 3(C) therein].

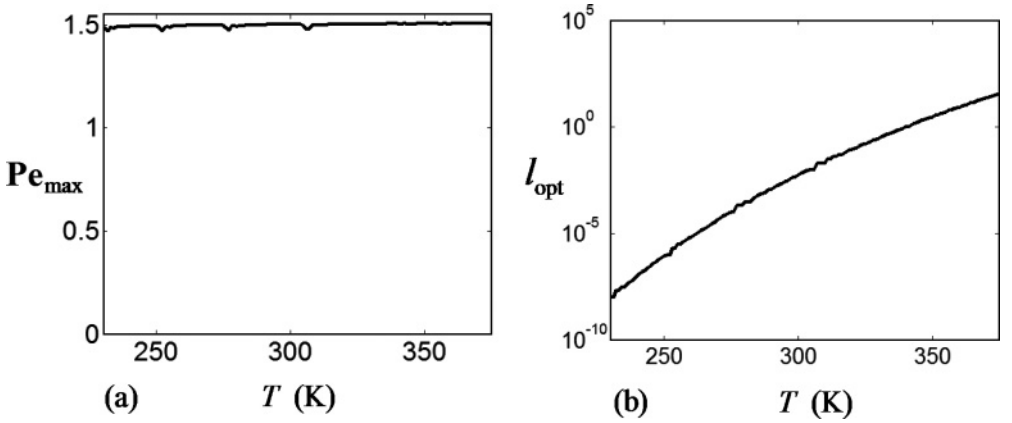

FIG. 6. (a) The maximum of the Péclet number as a function of temperature: $\mathrm{Pe}_{\max}(T) := \max_l \mathrm{Pe}(T,l)$. The numerical results indicate $\mathrm{Pe}_{\max}(T)$ is almost a constant. (b) Roughly speaking, the corresponding optimal value of light intensity l_{opt} at which $\mathrm{Pe}_{\max}(T)$ is reached increases exponentially with temperature T.

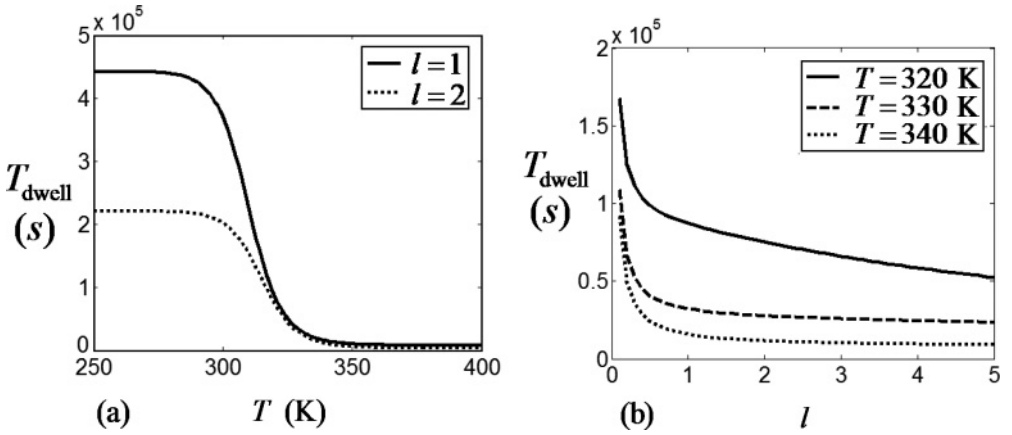

FIG. 7. The mean dwell time T_{dwell} of the synthetic rotatory molecular motor, as schematically depicted in Fig. 2, decreases with (a) temperature T and (b) light intensity l.

the modified one-dimensional hopping model are accurate to some extent.

To better understand the properties of this molecular motor, the numerical results of the mean velocity V and effective diffusion constant D as functions of light intensity l are plotted in Fig. 4. One can easily see that both V and D increase with T and l. But, as pointed out in [16], their ratio, i.e., the Péclet number $\text{Pe} = V/D$, has a maximum as a function of T. Further calculations show that the Péclet number Pe also has a maximum as a function of l (see Fig. 5). The numerical results also indicate that the maximum of Pe satisfies

$$\max_{T,l} \text{Pe}(T,l) \approx \max_{T} \text{Pe}(T,l) \approx \max_{l} \text{Pe}(T,l) \approx 1.5. \qquad (16)$$

So, for any given temperature T, we always can find an optimal value of light intensity $l_{\text{opt}}(T)$ at which the value of Pe is maximum. But, roughly speaking, $l_{\text{opt}}(T)$ increases exponentially with T (see Fig. 6).

From the results plotted in Fig. 7, one also finds that the mean dwell time T_{dwell} decreases with T and l; this is consistent with the corresponding results for the mean velocity [see Figs 3(a) and 4(a)]. Since there are no experimental data of mean dwell time T_{dwell} in [16], in the following we will verify our theoretical formulation of T_{dwell} through an indirect way. Instead, a dimensionless quantity, rotation excess, denoted by r_e, is defined.

In [23], it had been proved that, to some degree, a general N-state model can be approximated by a one-state model with *effective* forward and backward transition rates u_{eff} and w_{eff}. As a one-state model, the probability flux is $J = u_{\text{eff}} - w_{\text{eff}}$, while the mean dwell time in one single cycle is $T_{\text{dwell}} = 1/(u_{\text{eff}} + w_{\text{eff}})$ (see [8,9,26]). So the *effective* transition rates u_{eff} and w_{eff} satisfy

$$u_{\text{eff}} - w_{\text{eff}} = J, \qquad u_{\text{eff}} + w_{\text{eff}} = \frac{1}{T_{\text{dwell}}}; \qquad (17)$$

therefore,

$$u_{\text{eff}} = \frac{1}{2}\left(\frac{1}{T_{\text{dwell}}} + J\right), \; w_{\text{eff}} = \frac{1}{2}\left(\frac{1}{T_{\text{dwell}}} - J\right). \qquad (18)$$

Intuitively, for the rotary molecular motor devised in [16], u_{eff} and w_{eff} denote the numbers of full forward and backward rotations per unit time, which are denoted by Ω_+ and Ω_- in [16]. Therefore, the dimensionless *rotational excess* r_e defined by Feringa and coworkers, $r_e = (\Omega_+ - \Omega_-)/(\Omega_+ + \Omega_-)$, can be obtained here by

$$r_e = \frac{u_{\text{eff}} - w_{\text{eff}}}{u_{\text{eff}} + w_{\text{eff}}} = J T_{\text{dwell}}. \qquad (19)$$

The numerical results plotted in Fig. 8 are almost the same as the ones obtained by Feringa and coworkers in [16], where a completely different and special method is used. This implies

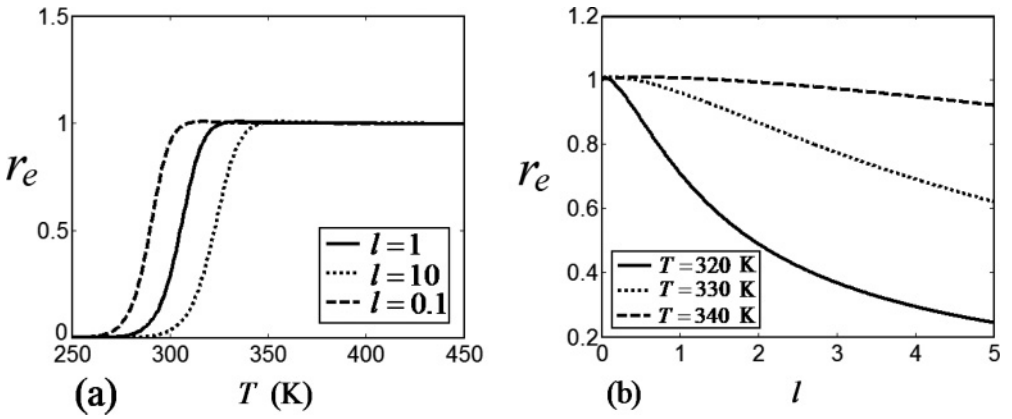

FIG. 8. The rotational excess $r_e = J T_{\text{dwell}}$ [see Eq. (19)] as a function of (a) temperature T and (b) light intensity l.

that the formulation (11) of the mean dwell times is accurate enough. In fact, the numerical results about $u_{\rm eff}$ and $w_{\rm eff}$ are also almost the same as the ones obtained in [16].

In conclusion, by applying our formulations of the mean velocity V, effective diffusion constant D, and mean dwell time $T_{\rm dwell}$ to the model of recently devised second generation rotary molecular motors, one can easily see that our results are accurate and valuable.

VI. SUMMARY

In this paper, one modified periodic one-dimensional hopping model is discussed. In this model, the particle in state i can not only jump forward to state $i+1$ or backward to state $i-1$ but is also allowed to jump to states $i+2$ or $i-2$ directly. One example of this modified hopping model is the one given by Feringa and coworkers for their second generation rotary molecular motors. In this paper, similar methods to those in [6,8] are used to get the mean velocity and effective diffusion constant, and the basic idea in [9] is employed to obtain the mean dwell time in one single mechanochemical cycle. The theoretical results are illustrated and verified by being applied to the model of Feringa. The method used in this paper is universal. It also can be employed for detailed studies of other types of one-dimensional hopping models and, consequently, to theoretical analysis of various kinds of biophysical and biochemical precesses, including the motion of many other synthetic molecular motors [17–21,27–32].

ACKNOWLEDGMENTS

This study is funded by the Natural Science Foundation of Shanghai (Grant No. 11ZR1403700). The author thanks Michael E. Fisher and Michael Hinczewski for their helpful suggestions in this study. The referees' critical reading is also very appreciated.

APPENDIX: THE CALCULATION OF p_i, a_i, AND b_i

From Eqs. (4), (5), and (6), one can easily verify that p_i, a_i, and b_i for $1 \leqslant i \leqslant N$ satisfy the following equations:

$$\begin{aligned} 0 = {} & (u'_{i-2}p_{i-2} + u_{i-1}p_{i-1} + w_{i+1}p_{i+1} \\ & + w'_{i+2}p_{i+2}) - (u_i + u'_i + w_i + w'_i)p_i, \end{aligned} \tag{A1}$$

$$\begin{aligned} 0 = {} & (u'_{i-2}a_{i-2} + u_{i-1}a_{i-1} + w_{i+1}a_{i+1} \\ & + w'_{i+2}a_{i+2}) - (u_i + u'_i + w_i + w'_i)a_i, \end{aligned} \tag{A2}$$

and

$$\begin{aligned} a_i = {} & (u'_{i-2}b_{i-2} + u_{i-1}b_{i-1} + w_{i+1}b_{i+1} + w'_{i+2}b_{i+2}) \\ & -(u_i + u'_i + w_i + w'_i)b_i + 2u'_{i-2}p_{i-2} \\ & + u_{i-1}p_{i-1} - w_{i+1}p_{i+1} - 2w'_{i+2}p_{i+2}. \end{aligned} \tag{A3}$$

If we define

$$\begin{aligned} J_{i-\frac{1}{2}} = {} & (u'_{i-2}p_{i-2} + u_{i-1}p_{i-1} + u'_{i-1}p_{i-1}) \\ & - (w_i p_i + w'_i p_i + w'_{i+1}p_{i+1}), \end{aligned} \tag{A4}$$

then Eq. (A1) implies $J_{i-\frac{1}{2}} =: J$ is constant. For the sake of convenience, we rewrite the linear algebraic equation (A1) as

$$A\vec{p} = 0, \tag{A5}$$

where $\vec{p} = (p_1, \ldots, p_N)^T$ is a column vector and A is the corresponding coefficient matrix.

Usually, the steady state probability p_i for $1 \leqslant i \leqslant N$ can be determined by Eq. (A1) or (A5), and the normalization condition $\sum_{i=1}^N p_i = 1$. In our discussion, we always assume this fact holds, which implies that the characteristic space of eigenvalue 0 of matrix A is one-dimensional. [It is easy to see that $\det(A) = 0$ since $\sum_{i=1}^N A_{ij} = 0$ for any $1 \leqslant j \leqslant N$. This implies 0 is one of the eigenvalues of matrix A.] So, from (A1) and (A2), one can see

$$a_i = \overline{V} p_i, \tag{A6}$$

where $\overline{V}$ is a constant. In view of the normalizing condition $\sum_{i=1}^N p_i = 1$, one easily finds

$$\begin{aligned} \overline{V} = \sum_{i=1}^N a_i = {} & \sum_{i=1}^N (2u'_{i-2}p_{i-2} + u_{i-1}p_{i-1} \\ & - w_{i+1}p_{i+1} - 2w'_{i+2}p_{i+2}) \\ = {} & \sum_{i=1}^N J_{i-\frac{1}{2}} = NJ = V. \end{aligned} \tag{A7}$$

To get $\vec{b} = (b_1, \ldots, b_N)^T$, we rewrite Eq. (A3) as follows:

$$A\vec{b} = \vec{a} - G\vec{p} = (VI - G)\vec{p}, \tag{A8}$$

where $I = \mathrm{diag}(1, \ldots, 1)$ is the unit matrix, $\vec{a} = (a_1, \ldots, a_N)^T$ is a column vector, and G is the matrix corresponding to vector $\vec{p}$ in Eq. (A3).

Since $\det(A) = 0$, $\vec{b}$ cannot be uniquely determined by Eq. (A8). However, the value of the effective diffusion constant D is uniquely determined by (10) and (A8). In fact, if $\vec{b}_1$ and $\vec{b}_2$ are two different solutions of (A8), then their difference $\vec{b}_0 = \vec{b}_1 - \vec{b}_2$ satisfies $A\vec{b}_0 = 0$. Since the characteristic space of eigenvalue 0 of A is one-dimensional, from (A5), one finds that $\vec{b}_0 = c\vec{p}$ with constant $c = (\sum_{k=1}^N b_{0k})$, i.e., $b_{0i} = (\sum_{k=1}^N b_{0k})p_i$. Thus,

$$\begin{aligned} & D(\vec{b}_1) - D(\vec{b}_2) \\ & = \sum_{i=1}^N (2u'_i + u_i - w_i - 2w'_i)b_{0i} - V\sum_{k=1}^N b_{0k} \\ & = \sum_{i=1}^N (2u'_i + u_i - w_i - 2w'_i)b_{0i} \\ & \quad - \left[\sum_{i=1}^N (2u'_i + u_i - w_i - 2w'_i)p_i\right]\left(\sum_{k=1}^N b_{0k}\right) \\ & = \sum_{i=1}^N \left\{(2u'_i + u_i - w_i - 2w'_i)\left[b_{0i} - p_i\left(\sum_{k=1}^N b_{0k}\right)\right]\right\} \\ & = 0. \end{aligned} \tag{A9}$$

Therefore, in the numerical calculations of the effective diffusion constant D [see Eq. (10)], we can use any one of the solutions of Eq. (A8). This random choice will not affect the final value of the effective diffusion constant D.

[1] S. Alexander, J. Bernasconi, W. R. Schneider, R. Biller, W. G. Clark, G. Grüner, R. Orbach, and A. Zettl, Phys. Rev. B **24**, 7474 (1981).

[2] S. Alexander, J. Bernasconi, W. R. Schneider, and R. Orbach, Rev. Mod. Phys. **53**, 175 (1981).

[3] R. D. Vale, Cell **112**, 467 (2003).

[4] A. B. Kolomeisky and M. E. Fisher, Annu. Rev. Phys. Chem. **58**, 675 (2007).

[5] Y. Zhang, Biophys. Chem. **136**, 19 (2008).

[6] M. E. Fisher and A. B. Kolomeisky, Proc. Natl. Acad. Sci. USA **98**, 7748 (2001).

[7] Y. Zhang, Phys. A **383**, 3465 (2009).

[8] B. Derrida, J. Stat. Phys. **31**, 433 (1983).

[9] P. A. Pury and M. O. Cáceres, J. Phys. A **36**, 2695 (2003).

[10] A. B. Kolomeisky, E. B. Stukalin, and A. A. Popov, Phys. Rev. E **71**, 031902 (2005).

[11] Y. Zhang, Chin. J. Chem. Phys. **23**, 65 (2010).

[12] A. B. Kolomeisky and M. E. Fisher, Phys. A **279**, 1 (2000).

[13] E. M. Geertsema, N. Koumura, M. K. J. t. Wiel, A. Meetsma, and B. L. Feringa, Chem. Commun. **24**, 2962 (2002).

[14] R. Eelkema, M. M. Pollard, J. Vicario, N. Katsonis, B. S. Ramon, C. W. M. Bastiaansen, D. J. Broer, and B. L. Feringa, Nature (London) **440**, 163 (2006).

[15] M. M. Pollard, M. Klok, D. Pijper, and B. L. Feringa, Adv. Funct. Mater. **17**, 718 (2007).

[16] E. M. Geertsema, S. J. van der Molen, M. Martens, and B. L. Feringa, Proc. Natl. Acad. Sci. USA **106**, 16919 (2009).

[17] R. Pei, S. K. Taylor, D. Stefanovic, S. Rudchenko, T. E. Mitchell, and M. N. Stojanovic, J. Am. Chem. Soc. **128**, 12693 (2006).

[18] P. Yin, H. M. T. Choi, C. R. Calvert, and N. A. Pierce, Nature (London) **451**, 318 (2008).

[19] T. Omabegho, R. Sha, and N. C. Seeman, Science **324**, 67 (2009).

[20] F. C. Simmel, Chem. Phys. Chem. **10**, 2593 (2009).

[21] L. Samii, H. Linke, M. J. Zuckermann, and N. R. Forde, Phys. Rev. E **81**, 021106 (2010).

[22] D. Tsygankov and M. E. Fisher, J. Chem. Phys. **128**, 015102 (2008).

[23] Y. Zhang, Sci. China, Ser. G **54**, 401 (2011).

[24] Y. Zhang, Phys. Lett. A **373**, 2629 (2009).

[25] S. Redner, *A Guide to First-Passage Processes* (Cambridge University Press, Cambridge, 2001).

[26] M. E. Fisher and A. B. Kolomeisky, Phys. A **274**, 241 (1999).

[27] D. A. Leigh, J. K. Y. Wong, F. Dehez, and F. Zerbetto, Nature (London) **424**, 174 (2003).

[28] J. V. Hernández, E. R. Kay, and D. A. Leigh, Science **306**, 1532 (2004).

[29] R. D. Astumian, Proc. Natl. Acad. Sci. USA **102**, 1843 (2005).

[30] R. D. Astumian, Proc. Natl. Acad. Sci. USA **104**, 19715 (2007).

[31] S. J. Green, J. Bath, and A. J. Turberfield, Phys. Rev. Lett. **101**, 238101 (2008).

[32] E. H. C. Bromley, HFSP J. **3**, 204 (2009).