PHYSICAL REVIEW E **68**, 021906 (2003)

Rectification efficiency of a Brownian motor

Daisuke Suzuki* and Toyonori Munakata†
Graduate School of Informatics, Kyoto University, Kyoto 606-8501, Japan

(Received 10 August 2002; revised manuscript received 22 November 2002; published 15 August 2003)

The energy balance of a Brownian motor is discussed based on a Langevin equation without the overdamped approximation. Energetics of the system suggests that the frictional dissipation energy associated with the unidirectional movement should be counted as a part of the useful energy for the rectification process of a Brownian motor. This leads to a new definition of the efficiency, which is applicable, contrary to the conventional one, even if the external load is absent. For the so-called flashing ratchet model, we numerically solve the Langevin equation for various situations and discuss both the temperature and the friction strength dependence of the rectification efficiency and the role of the *duty ratio*.

DOI: 10.1103/PhysRevE.68.021906 PACS number(s): 87.10.+e, 05.40.−a

I. INTRODUCTION

Recently there has been an increasing interest in the so-called "ratchets" or Brownian motors. These systems consist of Brownian particles moving in asymmetric potentials and are subject to a source of nonequilibrium such as external fluctuations or temperature gradients. As a consequence of these two ingredients—asymmetric potentials and nonequilibrium— a flow of particles can be induced.

Many kinds of models for "Brownian motors," which do not have direct contact with the chemical reactions, are derived from Feynman's ratchet and pawl system [1], which consists of a so-called ratchet, reminiscent of a circular saw with asymmetric saw teeth, and a pawl which admits the saw teeth to proceed without much effort into one direction (henceforth called "forward") but practically exclude the rotation in the opposite ("backward") direction. The ratchet is connected by an axle with a windmill whose vanes are surrounded by a gas at a finite temperature T_1. The ratchet and pawl are kept at a different temperature $T_2(<T_1)$. The random collisions of the surrounding gas molecules with the vanes will cause the ratchet to rotate in the forward direction. Such rectification of thermal noise could be utilized to perform work such as lifting a load.

Many variations of the Brownian motors have been developed [2–6], one of which is the "flashing ratchet model," which is mainly treated in this paper. In the flashing ratchet model, a Brownian particle moves in the potential which is asymmetric and periodic in space. The potential is switched on and off in time and this is the origin of the name "flashing." The time variation of the potential enables the system to absorb the energy from the external system and the particle can move in one direction using this energy.

The Brownian motor, which includes the flashing ratchet, aims at the unidirectional movement of the particle in situations where the scale of the system is so small that the thermal noise cannot be ignored. Because of such an interesting aim, the models are paid attention to not only by researchers of thermodynamics and statistical mechanics but also by those who try to explain the principle of the movement of real motor proteins [7,8] and those who try to invent the nanomachine [9,10].

When one evaluates how effectively a system works, efficiency is an important measure. For example, in a heat engine, efficiency is generally defined as the ratio of the work done by the engine to the input energy. In the case of Brownian motors, how is efficiency to be defined? So far, efficiency is generally defined as the ratio of the work done by the particle against the load to the input energy [5–8]. In other words, the efficiency above is the one of converting the input energy to the potential energy associated with the load. In this way the load is inevitably included in the model to define the efficiency. In the flashing ratchet model, the time-independent part of the gradient of the potential, which represents the load, has been incorporated to the system and the particle performs work such as ascending the potential gradient. These considerations lead to the conclusion that the efficiency is zero when the load is zero.

We have some question on the assertion that the system without the load does not utilize the input energy at all. Of course, the Brownian motor may be the system which can store part of the input energy as the potential. But originally, the aim of the Brownian motor is to move the particle in one direction. Assume that we have two kinds of Brownian particles, "*A*" and "*B*" and both consume (or dissipate) the same amount of input energy. The particle *A* kicks around but on an average moves in one direction and reaches the destination within a given time. The particle *B* also kicks around but does not achieve one directional movement and therefore cannot reach the destination within a given time. It is obvious that the particle *A* more efficiently uses the input energy than the particle *B*. But the conventional efficiency is zero for both particles.

Motivated by this point, Derenyi *et al.* [11] proposed a new efficiency which does not recourse to the load. That is, a task is first specified, i.e., to translocate the motor over a distance L during a given time τ, and the efficiency is defined as the ratio of the minimum energy necessary for the task to the input energy. Although this approach is general and applicable to chemical as well as heat engines, we take another approach, i.e., energetics of the Langevin dynamics [12], from which we are rather naturally led to a new defi-

*Electronic address: sdaisuke@amp.i.kyoto-u.ac.jp
†Electronic address: munakata@amp.i.kyoto-u.ac.jp

nition of the efficiency, "rectification efficiency," which has some similarity to the one proposed in Ref. [11].

In Sec. II, the Langevin equation, with the inertia effect taken into account, for the Brownian motor is introduced. Employing a time average method, we formulate the energy balance in the steady state. In Sec. III, a new "rectification efficiency" is introduced. In Sec. IV, the new efficiency is used to discuss the effects of temperature, friction, and the duty ratio on the directional movement of a ratchet. Here the duty ratio [13] denotes the ratio of the time t_{on} during which the asymmetric potential is on to the one cycle of the time variation of the potential $t_{on}+t_{off}$. Finally in Sec. V, we give some comments on the inertia effect and on the significance of the duty ratio in our model.

II. ENERGY BALANCE

We treat a Brownian particle in the heat bath. The system is described by the Langevin equations

$$\frac{dp}{dt}=-\xi p+R(t)-\partial_x U(x,t), \tag{1}$$

$$\frac{dx}{dt}=\frac{p}{m}, \tag{2}$$

where x denotes the position of the particle, p is the momentum, m is the mass, and $U(x,t)$ is an explicitly time-dependent potential along which the particle moves. We assume that the potential is expanded as

$$U(x,t)=U_0(x,t)+Lx, \tag{3}$$

where L is the load and $U_0(x,t)$ is periodic in x and t. The forces on the particle from the heat bath are the friction force $-\xi p$ and the random force which satisfies

$$\langle R(t)\rangle=0,$$

$$\langle R(s)R(t)\rangle=2m\xi k_b T\delta(s-t). \tag{4}$$

Instead of using the δ-correlated "white noise" $R(t)$, we can express Eqs. (1) and (2) as

$$dp=-\xi p\,dt+dW(t)-\partial_x U(x,t)dt, \tag{5}$$

$$dx=\frac{p}{m}dt, \tag{6}$$

where $dW(t)\equiv\int_t^{t+dt}R(t)dt$ denotes the increment of the Wiener process $W(t)$ in time dt, which satisfies

$$\langle dW(t)\rangle=0,$$

$$\langle dW(t)^2\rangle=2m\xi k_b T dt. \tag{7}$$

Now we discuss the energy balance based on Eqs. (5) and (6). For this purpose we first multiply Eq. (5) by p/m and interpret the resulting Eq. (8) in the Stratonovich sense [14],

$$\frac{p}{m}dp=-\xi\frac{p^2}{m}dt+\frac{p}{m}dW(t)-\partial_x U(x,t)\frac{p}{m}dt. \tag{8}$$

Since we consider the energy balance in the steady state and are not interested, in this paper, in the fluctuation from the average behavior, we take the long-time(τ) average of Eq. (8) as

$$\frac{1}{\tau}\int_0^\tau\frac{p(t)}{m}dp(t)=-\frac{1}{\tau}\int_0^\tau\xi\frac{p^2}{m}dt+\frac{1}{\tau}\int_0^\tau\frac{p}{m}dW(t)-\frac{1}{\tau}\int_0^\tau\partial_x U(x,t)\frac{p}{m}dt. \tag{9}$$

For infinitesimal time interval dt, it holds that

$$\frac{1}{m}\int_t^{t+dt}p(t)dp(t)\equiv\frac{1}{m}\left[\frac{p(t)+p(t+dt)}{2}\right][p(t+dt)-p(t)]$$

$$=\frac{1}{2m}[p(t+dt)^2-p(t)^2]$$

$$=\int_t^{t+dt}d\left(\frac{p^2}{2m}\right). \tag{10}$$

Thus the left-hand side of Eq. (9) vanishes as

$$\frac{1}{\tau}\int_0^\tau\frac{p(t)}{m}dp(t)=\frac{1}{\tau}\int_0^\tau d\left(\frac{p^2}{2m}\right)$$

$$=\frac{1}{\tau}\frac{[p^2(\tau)-p^2(0)]}{2m}\to 0\qquad(\tau\to\infty), \tag{11}$$

where we use the fact that $p(t)^2$ is bounded.

The first term on the right-hand side of Eq. (9) expresses the power of the friction force. Putting $\delta p(t)\equiv p(t)-\overline{p}$ with $\overline{p}\equiv(1/\tau)\int_0^\tau p(t)dt$, we have

$$\frac{1}{t}\int_0^t\xi\frac{p^2}{m}dt=\frac{\xi}{m}\overline{p^2}=\frac{\xi}{m}\overline{(\overline{p}+\delta p)^2}=\frac{\xi}{m}\overline{p}^2+\frac{\xi}{m}\overline{\delta p^2}. \tag{12}$$

The second term on the right-hand side of Eq. (9), related to the work done by the random force, can be dealt with similarly as Eq. (10). That is, for the infinitesimal time interval dt,

$$\int_{t_i}^{t_i+dt}p(t)dW(t)\equiv\left[\frac{p(t_i)+p(t_i+dt)}{2}\right]dW(t_i)$$

$$=\left[p(t_i)+\frac{dp(t_i)}{2}\right]dW(t_i)$$

$$\simeq p(t_i)dW(t_i)+\frac{1}{2}dW(t_i)^2, \tag{13}$$

where $dW(t)\equiv W(t+dt)-W(t), dp(t)\equiv p(t+dt)-p(t)$, and Eq. (5) is used. Therefore, we immediately see that

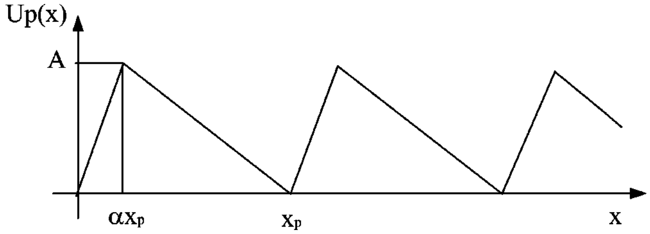

FIG. 1. Profile of potential $U_p(x)$. $U_p(x)$ is periodic, $U_p(x+x_p)=U_p(x)$, and asymmetric.

$$\frac{1}{\tau}\int_0^\tau \frac{1}{m} p(t)dW(t)=\xi k_b T. \tag{14}$$

The third term on the right-hand side of Eq. (9) is the rate of work done by the external field. It is seen from Eqs. (3) and (6) that

$$\frac{1}{\tau}\int_0^\tau \partial_x U(x,t)\frac{p}{m}dt=\frac{1}{\tau}\int_0^\tau \partial_x U_0(x,t)dx+\frac{1}{\tau}\int_0^\tau L\frac{p}{m}dt$$
$$=\frac{1}{\tau}\int_0^\tau \partial_x U_0(x,t)dx+L\overline{\frac{p}{m}}. \tag{15}$$

Substituting Eqs. (11), (12), (14), and (15) into Eq. (9), we have the following equation of the energy balance in the steady state:

$$-\frac{1}{\tau}\int_0^\tau \partial_x U_0 dx(t)=\frac{L}{m}\overline{p}+\frac{\xi}{m}\overline{p}^2+\xi\left[\frac{\overline{\delta p^2}}{m}-k_b T\right]. \tag{16}$$

This equation is general and valid regardless of the details of the model. To reveal the meaning of the energy balance equation (16), we study below numerically a typical model for the Brownian motor and obtain a concrete picture of the energy balance.

III. DERIVATION OF THE RECTIFICATION EFFICIENCY

In order to examine and interpret Eq. (16), we take a popular model for Brownian motor, that is, the so-called "Flashing Ratchet" model. In this model the potential $U_0(x,t)$ in Eq. (3) takes the form $U_0(x,t)=\Theta(t)U_p(x)$ with

$$\Theta(t)=\begin{cases}1, & t\in[0,t_{on})\\ 0, & t\in[t_{on},t_{on}+t_{off}),\end{cases}$$

$$U_p(x)=\begin{cases}\frac{A}{\alpha x_p}x, & x\in[0,\alpha x_p)\\ -\frac{A}{(1-\alpha)x_p}(x-x_p), & x\in[\alpha x_p,x_p),\end{cases}$$

where $\Theta(t)$ and $U_p(x)$ are periodic with the period $t_{on}+t_{off}$ and x_p, respectively. In Fig. 1, the asymmetric potential $U_p(x)$ is depicted, which flashes for t_{on} in one cycle. In order to grasp the general features of the dynamical behavior of the system, we performed numerical calculations for various values of the parameters which appear in the potential $U_0(x,t)$. We show the results for one typical case below for which $\alpha=0.1, A=6\pi, x_p=2\pi, L=1$, and $t_{on}=t_{off}=\pi/2$. We note that in all our simulations below, $\alpha, t_{on}+t_{off}, A$, and x_p are set as above.

The trajectories $x(t)$ and $p(t)$ are calculated on the basis of the Langevin equation [Eqs. (1) and (2)] for which we choose $\xi=T=1.0$. m and k_b are set to 1.0 throughout this paper. Each trajectory is obtained for time $t=20\,000$ and this is repeated 100 times. This trajectory information enables us to take average of each term of Eq. (16). Numerically, we obtained

$$-\frac{1}{\tau}\int_0^\tau \partial_x U_0 dx(t)=0.95,$$

$$\frac{L}{m}\overline{p}=0.10,$$

$$\frac{\xi}{m}\overline{p}^2=0.01,$$

$$\xi\left[\frac{\overline{\delta p^2}}{m}-k_b T\right]=(1.84-1.00).$$

The left-hand side of Eq. (16), which we denote as E_{influx}, represents the influx of energy to the system (the Brownian motor) due to the external field, which is consumed partly as the work against the load (the first term on the right-hand side), partly for the directional movement against the frictional force (the second term on the right-hand side), and partly as the fluctuation dissipation (the third term on the right-hand side). At this point, we comment on E_{influx} for latter convenience. During the long-time period τ, the external potential is on N times with $N\simeq\tau/[t_{on}+t_{off}]$. Numbering these intervals by i and denoting the position and time of both ends of the ith interval by $(x_{i,-},t_{i,-})$ and $(x_{i,+},t_{i,+})$, we express E_{influx} as $E_{influx}=(1/\tau)\Sigma_i[U_0(x_{i,-},t_{i,-})-U_0(x_{i,+},t_{i,+})]$. What is found from our numerical experiments is that normally the Brownian motor goes down the slope of the potential energy $U_0(x,t)$ and gets energy supply from this. If one wants to lay more emphasis on the chemical aspects of the flashing ratchet, we use the expression $dU_0(x,t)=\partial U_0/\partial x dx+\partial U_0/\partial t dt$ and from $(1/\tau)\int_0^\tau dU_0(x,t)=0$ reinterpret the expression for E_{influx} above, resulting from the jump or discontinuous (in time) events where the motor on average gets (chemical) energy. The second and the third terms on the right-hand side of Eq. (16) may be collectively treated as the dissipation energy given to the reservoir. Actually if one starts from the overdamped model [12,15] instead of the model described by Eqs. (1) and (2), the stochastic process (dx/dt) or $p(t)$ would not be well defined and we could not have divided the dissipation into the two components as was done in Eq. (16). This is one of the merits of our Langevin approach, which takes into account inertial effects and enables us to extract the dissipation energy associated with the directional movement even if $L=0$.

Based on the arguments above, we are naturally led to a new definition of (rectification) efficiency as

$$\eta_r \equiv \left[\frac{L}{m}\overline{p} + \frac{\xi}{m}\overline{p^2}\right] \Big/ \left|\frac{1}{\tau}\int_0^\tau \partial_x U_0 dx(t)\right|, \tag{17}$$

where both work against load L and dissipation energy associated with the directional movement against the frictional force are regarded as meaningful from a ''rectification'' viewpoint. This efficiency lays more emphasis on how the input energy is utilized for the directional movement than on how much energy is stored inside the total system. Somewhat similar efficiency to η_r was recently proposed by Wang and Oster, who introduced the so-called *Stokes* efficiency [16,17] by

$$\eta_{Stokes} = \frac{\xi}{m}\overline{p^2} / [\Delta F + \Delta W]. \tag{18}$$

Here ΔF represents the chemical energy supplied to the Brownian motor, which is given in their theoretical framework externally as the boundary condition on the (chemical) energy profile. If we consider the flashing of the potential in our model as the transition between the chemical states, we may regard ΔF as E_{influx}. ΔW is the work done by the external field on the Brownian motor. In this paper, we consider only the case in which the external field exerts (constant in time) force against the movement of the motor, i.e., $(L/m)\overline{p} > 0$. If $(L/m)\overline{p} < 0$ however, we would put $\Delta W = -(L/m)\overline{p}$ and employ the same expression as *Stokes* since L is not the load anymore. The difference between η_r and η_{Stokes} thus consists in the treatment of the load. We consider, as many others, that the work done against the external force and stored in the system should be in the numerator of the efficiency. With these preparations, we now investigate the rectification efficiency of the Brownian motor for the flashing ratchet model in detail.

IV. APPLICATION OF THE RECTIFICATION EFFICIENCY

A. Efficiency versus temperature and friction

We now calculate the rectification efficiency in various cases. First, we consider the rectification efficiency and the average momentum or velocity as a function of temperature T and the friction constant ξ. In Fig. 2, we show the T dependence of the rectification efficiency and the average momentum. We observe the peaks of the efficiency and the average momentum around $T \approx 1.0$. This is due to the fact that thermal activation is impossible when the coupling with the reservoir becomes too weak (low temperature), and the systematic or directional motion is prohibited when the coupling with the reservoir becomes too strong (high temperature). Friction constant dependence of the efficiency and the average momentum are also studied for a constant temperature. The result, which is not shown here, represents a typical Kramers' behavior, showing peaks as in Fig. 2. When ξ is small we have a weak contact with the reservoir and thermal activation is not expected. On the other hand, too large ξ makes the Brownian particle immobile. For $T=1$, we observed the optimum friction, $\xi \approx 1.0$ ($L=0$).

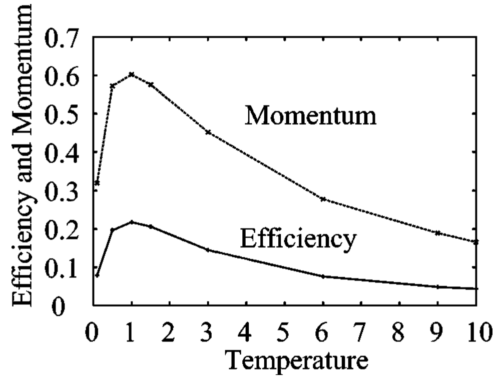

FIG. 2. The rectification efficiency and the average momentum as a function of temperature. ξ is set to 1.0 ($L=0$).

B. Efficiency versus duty ratio

The duty ratio [13] is originally defined as the fraction of the time that a motor spends attached to its filament, and it turned out to characterize the structural and functional properties of the motor proteins. For the flashing ratchet model, it may be defined as $t_{on}/[t_{on}+t_{off}]$ and we studied the duty ratio dependence of the efficiency in view of the crucial roles played by the duty ratio.

We investigated the rectification efficiency for the duty ratio. In Fig. 3 are plotted the duty ratio dependence of the energy influx $-(1/\tau)\int_0^\tau \partial_x U_0 dx(t)$, the dissipation related to the unidirectional movement $(\xi/m)\overline{p}^2$, and the rectification efficiency η_r, with the load L set to zero and $\xi=1.0$ and $T=1.0$. From Fig. 3, we note that the duty ratio (0.3) corresponding to the maximum η_r is different from the one (0.5) corresponding to the maximum $(\xi/m)\overline{p}^2$. That is, if the main purpose of this motor is a fast directional movement regardless of its energy consumption, it is optimum for the ratio to be set to 0.5. In such a mode of the motion, the dissipation due to fluctuations, the last term on the right-hand side of Eq. (16), becomes also large and the efficiency is not largest there. On the other hand, if we want the motor to utilize the input energy efficiently for the directional movement, the duty ratio should be set to 0.3. In this case, the motor absorbs less energy than in the case where the duty ratio is 0.5, and

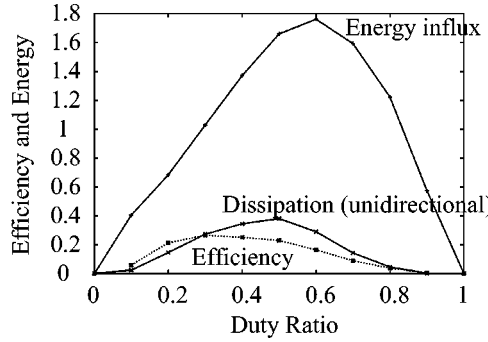

FIG. 3. The rectification efficiency as a function of the duty ratio. $\xi=T=1.0$.

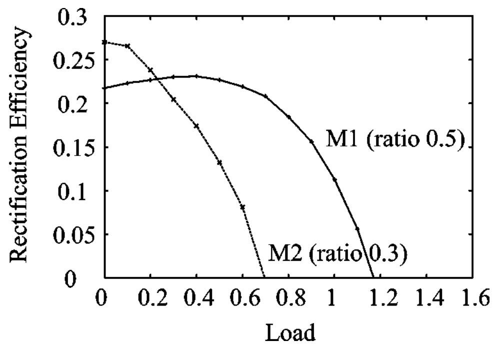

FIG. 4. The rectification efficiency as a function of load. The solid line is the motor of type $M1$ whose duty ratio is 0.5, and the other line is the motor $M2$ whose duty ratio is 0.3.

we can trace the origin of the high efficiency to the inertia effects by which the Brownian particle can keep on moving even if the potential is switched off.

This observation can be slightly generalized as follows. Figure 3 tells us that we have generally two duty ratios corresponding to a given average velocity, one with a larger duty ratio generally consumes more energy than the other with less duty ratio. This may be interpreted that the mode with the larger duty ratio is supplied with more energy than is necessary to achieve a target velocity. However, if the expected role for the motor is the work against the load, the scenario is different as is discussed below.

C. Efficiency versus load

In Fig. 4, we plot the rectification efficiency as a function of the load for two kinds of motors, one with large duty ratio of 0.5 (type $M1$) and the other with small duty ratio of 0.3 (type $M2$) ($\xi = T = 1.0$). The optimum load of the motor $M1$ is 0.4 with the efficiency 0.23, but the motor $M2$ has no optimum load and the maximum efficiency of 0.27 is accomplished when $L=0$. In other words, the motor $M2$ is more efficient than the motor $M1$ in the low load region and vice versa in the high load region. The above result illustrates that the difference of the duty ratio [13] makes the functional difference of motors, that is, load-pulling type ($M1$) and swift motion type ($M2$).

V. CONCLUDING REMARKS

In this paper, we studied the dynamical properties of a Brownian motor based on a Langevin equation for a flashing ratchet model. Inclusion of the inertia effects, which are usually neglected from the hydrodynamic considerations, makes momentum $p(t)$ of a Brownian particle a well-defined process, and this has profound effects in discussing the energy balance. If we had started from an overdamped Langevin equation $m\xi(dx/dt) = R(t) - \partial_x U(x,t)$ and multiply dx, which is no longer of order dt, on both sides, we immediately notice that both $\int dx R(t)$ and $m\xi \int dx dx(t)/dt$ diverge [but the diverging part cancels to give finite value to $\int dx \partial_x U(x,t)$]. In other words, in order to extract the work against the frictional force $(\xi/m)\overline{p^2}$, appearing on the right-hand side of Eq. (16), it is necessary to start from the full Langevin equation (1). Of course, if the overdamping condition is applicable for a given problem, one can employ the overdamped Langevin equation but still Eq. (17) should be used for the efficiency. This is because $(\xi/m)\overline{p^2}$ represents inevitable energy consumption for the particle to move in one direction against the frictional force, and this consumption is, so to speak, a meaningful dissipation. Similar result is obtained by Derenyi *et al.* [11] in a slightly different context. The difference between the efficiency in Ref. [11] and ours consists in the treatment of the input energy (the denominator of the efficiency). In Ref. [11], for application to chemical engines, adenosine triphosphate hydrolysis energy is adopted as the input. In this paper, on the other hand, from a standpoint of the thermodynamics, the net input energy supplied to the system by the external field is adopted as the input.

It is remarked that Eq. (17) may give an important information on the motor proteins from an energetic viewpoint. Actually, in Sec. IV, we discussed two types of motors, i.e., swift motion type ($M2$) and load-pulling type ($M1$) based on the duty ratio [13]. It was shown there (see Fig. 4) that a motor with small duty ratio (0.3) can moves swiftly with low energy consumption for small L, while a motor with large duty ratio (0.5) can work optimally under the high load condition. Although our Langevin model represents an oversimplified picture for real motor proteins, it confirms the importance of the duty ratio as the possible mechanism for the functional difference of motors.

Finally, we note that our energetic approach to the Langevin equation is general and it can be employed to various ratchet models, including the random-flashing ratchet and the rocking ratchet [5]. The Feynman ratchet [1] especially presents an interesting problem from the viewpoint of efficiency and we are currently engaged in this problem.

[1] R.P. Feynman, R.B. Leighton, and M. Sands, *The Feynman Lectures on Physics* (Addison-Wesley, Reading, MA, 1966), Vol. 1, Chap. 46.

[2] R.D. Astumian, Science **276**, 917 (1997).

[3] P. Reimann, Phys. Rep. **361**, 57 (2002).

[4] P. Reimann, R. Bartussek, R. Haubler, and P. Hanggi, Phys. Lett. A **215**, 26 (1996).

[5] H. Kamegawa, T. Hondou, and F. Takagi, Phys. Rev. Lett. **80**, 5251 (1998).

[6] J.M.R. Parrondo, J.M. Blanco, F.J. Cao, and R. Brito, Europhys. Lett. **43**, 248 (1998).

[7] A. Parmeggiani, F. Julicher, A. Ajdari, and J. Prost, Phys. Rev. E **60**, 2127 (1999).

[8] H.X. Zhou and Y.D. Chen, Phys. Rev. Lett. **77**, 194 (1996).

[9] M. Bier and R.D. Astumian, Phys. Rev. Lett. **76**, 4277 (1996).

[10] M. Schreier, P. Reimann, P. Hanggi, and E. Pollak, Europhys.

Lett. **44**, 416 (1998).
[11] I. Derenyi, M. Bier, and R.D. Astumian, Phys. Rev. Lett. **83**, 903 (1999).
[12] K. Sekimoto, J. Phys. Soc. Jpn. **66**, 1234 (1997).
[13] J. Howard, Nature (London) **389**, 561 (1997).
[14] C.W. Gardiner, *Handbook of Stochastic Methods* (Springer, Berlin, 1983).
[15] T. Munakata, A. Igarashi, and T. Shiotani, Phys. Rev. E **57**, 1403 (1998).
[16] H. Wang and G. Oster, Europhys. Lett. **57**, 134 (2002).
[17] H. Wang and G. Oster, Appl. Phys. A: Mater. Sci. Process. **75**, 315 (2002).