Asymmetry and direction reversal in fluctuation-induced biased Brownian motion

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(Received 2 March 1999)

The biased movement of a Brownian particle in a periodic potential fluctuating between a flat and a kinked ratchet state, as first studied by Chauwin, Ajdari, and Prost [3], is examined. The purpose is to study the physical origin of the frequency-dependent direction reversal of the biased Brownian motion in this system. We show that the existence of the directional reversal depends not only on the lengths of the projections of the two ratchet arms on the potential axis (the arm-projection asymmetry), but also the overall spatial geometry of the potential in a period. In particular, we show that the direction reversal can be obtained in this kinked ratchet model even when the two arm projections are equal. Since this two-state model is the simplest to generate direction reversal and particles can be separated more efficiently in a fluctuating potential if direction reversal exists, the results obtained in this study should be useful for future application in particle separation. [S1063-651X(99)08810-8]

PACS number(s): 05.40.Jc, 87.19.Rr

It has been shown theoretically that a Brownian particle can be made to undergo unidirectional movement in a one-dimensional asymmetric periodic potential if the ''interaction'' between the particle and the potential field is made to fluctuate randomly or regularly among a number of states [1–6]. The fluctuation of the interaction can be produced either by externally switching the potential among a number of different potential states [1–4] or by changing the interaction parameter (such as the charge, if the potential is electric) on the Brownian particle through a nonequilibrium chemical cycle [5,6]. It has been argued that the motility of biological motors might be governed by the same mechanism [7–9]. Recently, this biased Brownian motion has been proved experimentally [10]. The same device could be applied to the separation of proteins or particles based on their sizes, electric charges, etc.

To generate biased Brownian motion in a fluctuating periodic potential, the potential in each period must possess some sort of asymmetry [1–6]. A simple and widely used asymmetric potential is the ordinary *ratchet* potential, in which the potential in each spatial period is shaped like a saw-tooth with two *unequal straight* arms. The potential is referred to as having an ''arm-projection asymmetry'' since the projections of the two sides of a ratchet on the potential axis are not equal [see Fig. 1(a)]. It is well known that the net movement of a Brownian particle in a periodic ratchet potential fluctuating randomly between a flat and a nonflat state (the two-state on-off model) is always biased in one direction, independent of the frequency of the fluctuation [1,2].

Recently, the study of direction reversal (DR) in fluctuation-induced biased Brownian motion on asymmetric periodic potentials has attracted considerable attention [3,4,11]. Millonas and Dykman discussed the generation of DR in a stationary periodic potential induced by a Gaussian force noise with a nonwhite power spectrum [11]. Chauwin, Ajdari, and Prost have suggested that DR can be obtained in the two-state ratchet model shown in Fig. 1(a) if the *long* arm of the ratchet is *kinked*, as shown in Fig. 1(b) [3]. Bier and Astumian also have found DR in a fluctuating three-state model [4]. These models are potentially very useful because DR could lead to more efficient fluctuation-induced separation of particles [4]. In these models, the arm-projection asymmetry was assumed to be necessary for the generation of DR. Recently, we have shown that biased Brownian motion can be obtained for some models without the arm-projection asymmetry [6]. It is therefore interesting to see whether DR also can be obtained in models without this asymmetry. In this paper, we show that DR can be produced in the *kinked* two-state model of Chauwin, Ajdari, and Prost no matter whether the value of *a* in Fig. 1(b) is larger than, equal to, or smaller than *b*. That is, the arm-projection asymmetry is not necessarily required for this two-state model to

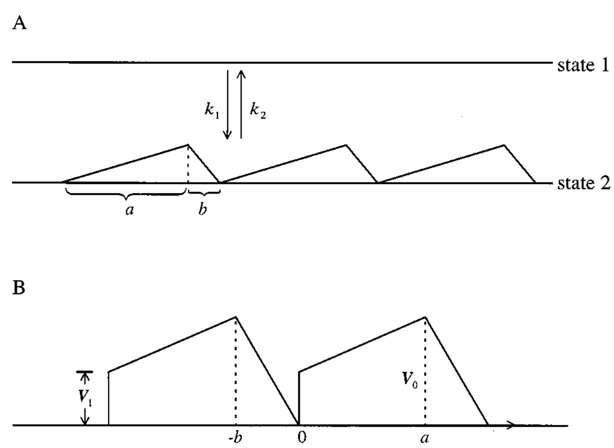

FIG. 1. (a) Two-state on-off model of a fluctuating periodic potential. The potential is said to have an ''arm-projection asymmetry'' when the projections of the two arms of a ratchet on the *x* axis, *a* and *b*, are not equal. (b) The kinked ratchet model of Chauwin, Ajdari, and Prost [3].

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generate the DR phenomenon; the asymmetry of the potential caused by the kink at the potential well (at $x=0$) also plays an important role. First we present the calculations demonstrating DR in this kinked two-state ratchet model and then discuss the physical mechanism underlying the generation of DR.

The mathematical formalism for calculating the transport flux (or the average velocity) of Brownian particles in a two-state on-off model is standard [1,5,6]. Briefly, the steady-state probabilities $[p_i(x),\ i=1,2]$ of finding a particle located at x when the fluctuating potential, $V(x)$, is in state i are obtained by solving two coupled linear second-order ordinary differential equations,

$$P_1''(x)-k_1p_1(x)+k_2p_2(x)=0, \tag{1}$$

$$P_2''(x)+V'p_2'(x)+V''p_2(x)+k_1p_1(x)-k_2p_2(x)=0, \tag{2}$$

using the normalization condition that the total probability of particles in one period is unity:

$$\int_0^1 (p_1(x)+p_2(x))dx=1, \tag{3}$$

and periodic boundary conditions for the probability and the flux at the two ends of a period. The transport flux u then is given by

$$u=-p_1'-p_2'-V'p_2. \tag{4}$$

Since Eqs. (1) and (2) imply $u'=0$, the transport flux will be a constant independent of x. For an arbitrary $V(x)$, Eqs. (1) and (2) can be solved approximately using a ''finite difference'' method [5,6]. This method requires large computer memory and computing time. On the other hand, for piecewise linear periodic potentials, Eqs. (1) and (2) can be solved using the eigenvalue-eigenfunction method [1,2]. This method can generate more accurate numerical results with less computing time and is used in this study [12]. Before presenting the calculation results, we emphasize that the parameters $V,x,u,$ and the k_i have been made dimensionless and are related to their corresponding physical quantities (signified by a bar) by $V=\overline{V}/k_BT$; $x=\overline{x}/L$; $u=\overline{u}L/D$; $k_i=\overline{k}_iL^2/D$ where L is the length of the period of the potential, D is the diffusion coefficient of the particle, and k_BT is the product of the Boltzmann constant and the temperature. Thus, it is obvious that $a+b=1$ [this is the reason why the integration in Eq. (3) is from 0 to 1]. Furthermore, the dimensionless rate constants k_1 and k_2 are not the same for different particles if they have different diffusion coefficients. This is exactly the reason why particles with different diffusion coefficients can be separated in this system [4].

There are many ways to modulate the average frequency of the fluctuation of the potential in a two-state model. For example, one could keep one rate constant in Fig. 1(a) fixed and vary the other or vary both rate constants simultaneously. From preliminary calculations we found that the existence of DR is more related to the value of k_2 than k_1. This may be due to the fact that particles can execute net biased movement only when the potential is in state 2. Thus, only k_2-dependent transport fluxes are considered in this study. That is, with any given potential $V(x)$ (characterized by the values of a, V_1, and V_0), the transport flux (or the net velocity of the Brownian particle) of the system is calculated and expressed as a function of k_2 at fixed k_1. In all calculations, V_0 was set to 10 and three arm-projection asymmetry parameters were considered: $a=0.6$, 0.5, and 0.48. The calculated $u(k_2)$ curves for a number of V_1 values at $k_1=0.01$ are shown in Figs. 2(a) and 2(b) for $a=0.5$ and 0.48, respectively. The curves for $a=0.6$ are qualitatively very similar to those in Fig. 2(a) and therefore are not shown. The k_2 at which the transport flux changes sign (k_2^*) can be obtained from the curves in Figs. 2(a) and 2(b) and are plotted in Fig. 2(c) as a function of V_1. The same set of calculations

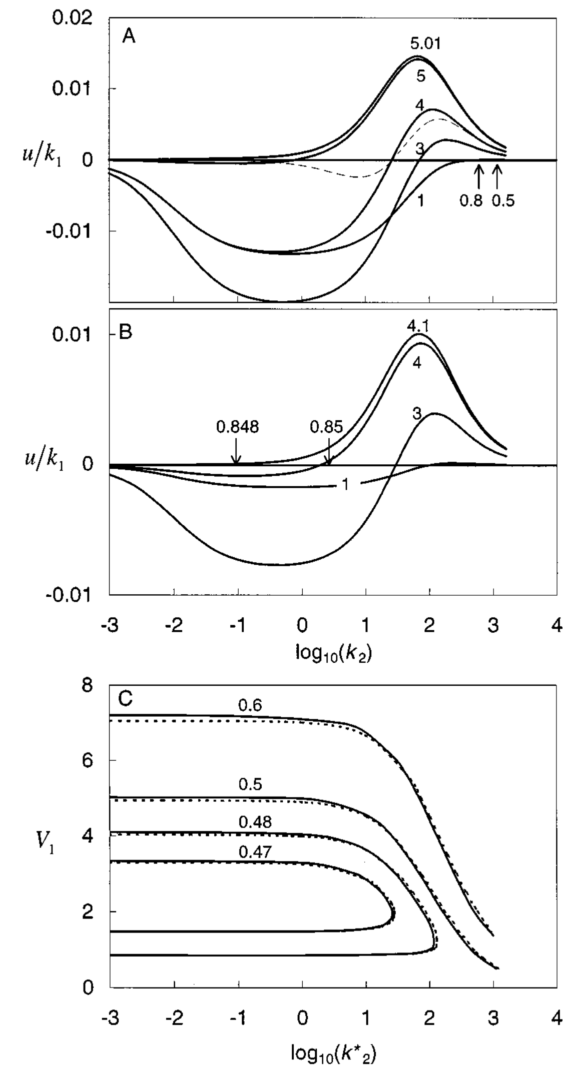

FIG. 2. (a). Transport fluxes calculated directly from Eqs. (1) and (2) at different kink height (V_1) for $a=0.5$ and $k_1=0.01$. The value of V_1 is indicated by each curve. The curves at $V_1=0.8$ and 0.5 are not shown in the figure; only the positions of the direction reversal (the intercepts) are indicated. The dotted curve is for $V_1=4$ and $k_1=10$. As can be seen, the reversal k_2 is not very sensitive to the value of k_1. (b) The calculated u for $a=0.48$ at different V_1. For $V_1=0.85$ and 0.845, only the intercept is shown in the figure. (c) The direction reversal frequency at which the transport flux changes sign (the reversal frequency) as a function of V_1 at different a values. k_1 is 0.01 for the solid curves and 10 for the dotted.

also were done for $k_1=10$ (a thousandfold increase). The results are shown as dotted curves in Figs. 2(a) and 2(c). Several conclusions can be drawn from these calculations: (i) Direction reversal (DR) can be obtained in this two-state kinked ratchet model even when the arm-projection asymmetry is absent [see Fig. 2(a)], in contrast to what was suggested by the theory of Chauwin, Ajdari, and Prost [3]; (ii) For a large kink height (large V_1), the flux is mostly positive and k_2^* decreases rapidly as V_1 increases (i.e., k_2^* is very sensitive to V_1), irrespective of the value of a; (iii) At small V_1, the flux is mostly negative and, as V_1 decreases, the value of k_2^* increases as a function of V_1 if $a\geqslant 0.5$ and decreases if $a<0.5$ [see the arrows in Figs. 2(a) and 2(b)]. That is, as shown in Fig. 2(c), the V_1-k_2^* curve at small V_1 for $a\geqslant 0.5$ is quite different from that for $a<0.5$. In fact, direction reversal does not exist for the $a<0.5$ case if V_1 is smaller than a limiting value; (iv) There exists a range of V_1 in which $u(k_2)$ contains both positive and negative amplitudes and the value of k_2^* changes slowly as V_1 is varied. This range of V_1 is therefore most suitable for particle separation; (v) As shown in Fig. 2(c), almost identical V_1-k_2^* curves are obtained when the value of k_1 is increased 1000 times from 0.01 to 10, showing that the generation of DR is mostly controlled by k_2, not k_1.

In the following, we show that the existence of DR at small k_1 as found in the calculations can be predicted using an approximate ‘‘deterministic’’ physical theory. For the two-state kinetic system in Fig. 1(a), the *average* time for the potential to stay in states 1 and 2 are constant and equal to $t_1=1/k_1$ and $t_2=1/k_2$, respectively. Thus, in this theory the potential is treated approximately as if it were oscillating regularly between states 1 and 2 with constant times of duration of t_1 and t_2. The distribution of particles on the x axis of the potential within each spatial period will be time dependent due to the translocation (or displacement) of the particles. At steady state (after the system has undergone a large number of oscillations), the distribution functions in both states 1 and 2 become invariant from one oscillation to the other, but are still time dependent. The net translocation of all particles in one spatial period in t_2 when the potential is in state 2 determines the steady-state transport flux of the system [3]. Thus, to see whether the k_2-dependent DR occurs in the system, one needs only to compare the signs of the net steady-state translocation of particles in one potential period at large and small t_2: DR is guaranteed to occur if the signs at these two extremes are different.

If the movement of an overly damped particle in a potential field is treated classically without considering the thermal fluctuation effect, the *velocity* of the particle is equal to $-\rho(x)/\gamma$ where $\rho(x)$ is the slope of the potential $[\rho(x)=dV(x)/dx]$ at x and γ is the friction coefficient of the particle. Let us consider the spatial period between $x=-b$ and $x=a$ as shown in Fig. 1(b). The slopes on the a and b sides of the potential are constant and can be expressed as

$$\rho_a=(V_0-V_1)/a, \quad \rho_b=-V_0/b. \tag{5}$$

Let t_a and t_b denote the average time for a particle to diffuse to the potential minimum at $x=0$ from the potential peaks at $x=a$ and $x=-b$, respectively. Then, we have

$$t_a=-a/(-\rho_a)=a^2/(V_0-V_1), \quad t_b=b/(-\rho_b)=b^2/V_0, \tag{6}$$

where the value of γ has been set to unity for simplicity. Let us assume that the time to stay in state 1 is very long (k_1 is very small) so that the particles on the x axis can be considered as uniformly distributed at $t=0$ when the potential is switched from state 1 to state 2. Then, when $t(=t_2)$ is smaller than either t_a or t_b, the total translocation (or the distance of movement) of particles on the two sides of the potential well can be expressed separately as

$$S_a=-(\rho_a t)^2/2-(a-\rho_a t)\rho_a t=-a\rho_a t+(\rho_a t)^2/2, \tag{7}$$

$$S_b=(\rho_b t)^2/2-(b+\rho_b t)\rho_b t=-b\rho_b t-(\rho_b t)^2/2, \tag{8}$$

where positive means translocation to the right and negative to the left. Note that the first term of the expression on the right-hand side of the first equal sign in Eqs. (7) or (8) represents the average translocation of particles that have moved to the potential minimum (at $x=0$) in time t, while the second term represents the translocation of the particles still on the slope. For sufficiently small $t(=t_2)$, the net translocation of particles in one spatial period then can be evaluated as

$$S_0=S_a+S_b=V_1 t+O(t^2) \quad \text{as} \quad t\to 0, \tag{9}$$

where Eq. (5) has been used to eliminate ρ_a and ρ_b. Thus, for any nonzero V_1, the net translocation of particles at small t_2 is always positive [toward the right in Fig. 1(a)] no matter whether a is larger than, equal to, or smaller than b. Then, if the net translocation of particles can be proved to be negative at large t_2 (or small k_2), the occurrence of DR in this model is guaranteed.

When t_2 is infinitely large ($k_2=0$), all the particles will reach an equilibrium distribution determined by the potential. In this case, the net translocation of the particles is equal to the difference of the ‘‘center of mass’’ of the particles in a spatial period between the final ($t=\infty$) and the initial ($t=0$) distributions. It is easy to see that for a uniform distribution the center of mass at $t=0$ is located at the midpoint between $-b$ and a, $m_0=(a-b)/2$. Let $f(x)\equiv C\exp[-V(x)]$ denote the equilibrium distribution of particles in one spatial period at large t where $C=1/\int_{-b}^{a}\exp[-V(x)]dx$ is the normalization constant. Then the center of mass at large t is located at

$$m_\infty=\int_{-b}^{a} x f(x)dx \tag{10}$$

and the net translocation of particles in the period between $x=-b$ and $x=a$ can be expressed as

$$S_\infty=m_\infty-m_0=\int_{-b}^{a} x f(x)dx-(a-b)/2. \tag{11}$$

Thus, DR occurs if S_∞ is negative. With given values of V_1, V_0 and a for the kinked linear ratchet potential in Fig. 1(b), the evaluation of S_∞ in Eq. (11) is straightforward. The calculated S_∞ at different a values are plotted as a function of V_1 in Fig. 3. As can be seen from the figure, S_∞ is always negative, independent of V_1, when $a\geqslant 0.5$. In this case, DR

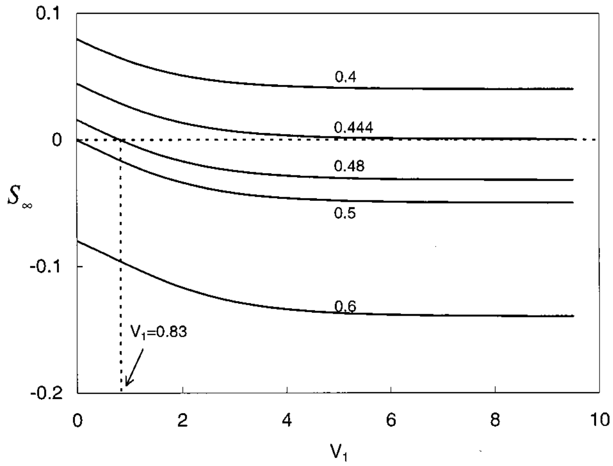

FIG. 3. S_∞ evaluated from Eq. (11) for different a values as a function of V_1. For the model to generate DR, the sign of S_∞ has to be negative.

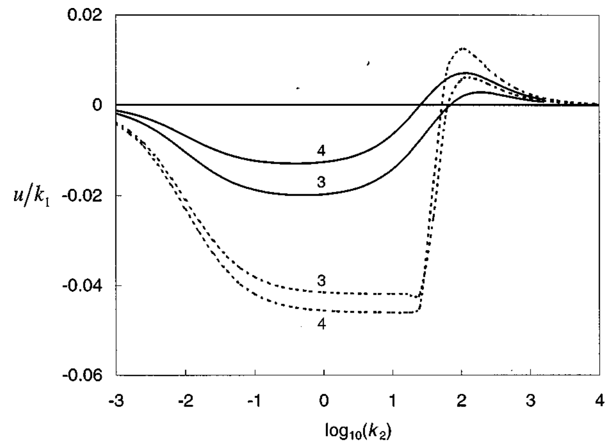

FIG. 4. $u(k_2)$ evaluated at $V_1=3$ and 4 using the approximate theory (the dotted lines) are compared with those evaluated exactly in Fig. 2(a). The values of a and k_1 are 0.5 and 0.01, respectively.

is always present in the system. One must note that this conclusion is not affected by the value of V_0. When $a<0.5$, S_∞ may be negative or positive depending on the values of a and V_1. As shown in Fig. 3, when $a \leqslant 0.444$, S_∞ is always positive, independent of V_1. In this case, no DR is expected. On the other hand, when $0.444<a<0.5$, DR is possible if V_1 is large enough. For example, DR can be obtained for the $a=0.48$ case if V_1 is larger than 0.83. This explains the limiting V_1 in the reversal frequency shown in Fig. 2(c) for $a=0.47$ and 0.48, as discussed above.

Thus, based on a simple physical argument, a rule is obtained for the prediction of the existence of DR for the fluctuation-induced movement of Brownian particles in a two-state periodic potential with kinked ratchets. With any given kinked ratchet [characterized by the values of a (and therefore b), V_1 and V_0], one simply calculates the S_∞ in Eq. (11) and examines its sign. If the sign of S_∞ is positive, DR may or may not be present. But, if the sign of S_∞ is negative, DR is definitely present.

The finding that DR can be obtained in this model even when $a=b$ implies that the ‘‘arm-projection’’ asymmetry is not a necessary condition for the kinked ratchet model of Chauwin *et al.* to generate DR. Chauwin, Ajdari, and Prost did not reach the same conclusion, because they failed to take into account the net translocation caused by the distribution of particles after they reach the bottom of the potential well. As will be shown elsewhere, DR also can be generated for this two-state model even when the kink in Fig. 1(b) is not vertical, but tilted.

The above physical theory can also be used to explain why the particle movement is unidirectional (no direction reversal) in the ordinary (nonkinked) ratchet potential in Fig. 1(a) as found before in [1,2]. Since $V_1=0$ in that case, the net translocation at small $t(=t_2)$ can be obtained from Eqs. (7) and (8) as $S_0=\frac{1}{2}(V_0 t/ab)^2\,(b-a)$. Thus, when $b>a$ (or $a<0.5$), S_0 is positive. From Fig. 3, S_∞ at $V_1=0$ is also positive in this case. As a result, there is no DR and the biased movement is always in the positive x direction. Similarly, when $a>b$ ($a>0.5$), both S_0 and S_∞ are negative, implying a unidirectional movement of the particle in the negative x direction.

The approximate deterministic theory can be also applied to simulate semiquantitatively the entire transport curve. When the value of k_1 is constant, the transport flux of the system, $u(k_2)$, can be evaluated approximately as $u(k_2)=S(t)/(t+1/k_1)$, where $k_2\equiv 1/t$ and $S(t)$ is the net translocation of particles in a spatial period at time t after the potential is switched (from state 1) to state 2. To evaluate $S(t)$, one has to divide the time t into three domains characterized by the t_a and t_b in Eq. (6): $t<t_b<t_a$, $t_b<t<t_a$, and $t>t_a>t_b$ (see [3]). The transport flux calculated with $k_1=0.01$ and $a=b=0.5$ are shown in Fig. 4 for $V_1=3$ and 4. As expected, the theory does produce DR correctly. But, the theoretical and the exact flux curves do not agree quantitatively. The deviations may come from four factors. First, the value of k_1 is not equal to zero as assumed in the theory. It is possible that better agreement could be obtained if k_1 is reduced. Second, the translocation of particles in state 2 is treated as a ‘‘deterministic’’ dynamic process in the theory. That is, only the downhill translocation of particles is considered. As a result, particles are not allowed to jump from one spatial period to the other. In a ‘‘stochastic’’ treatment (the exact solution), up-hill translocations and jumping over hills are allowed. How this factor contributes to the deviation is not known. Third, the particles that have translocated to the bottom of the potential well may not reach an equilibrium distribution as assumed in the theory. In general, an equilibrium distribution is reached only at long time (or at small k_2). This may be the reason why deviations seem to be smaller at small k_2, as shown in Fig. 4. Fourth, substituting a randomly fluctuating potential with a regularly oscillating one may also contribute to the deviation.

In conclusion, we have shown that the direction of the biased movement of Brownian particles in the two-state model of Chauwin, Ajdari, and Prost with kinked ratchet potentials can be easily changed by changing the frequency of the fluctuation, irrespective of the existence of the arm-projection asymmetry in the potential. The existence of a kink at the trough of the potential (at $x=0$) is necessarily required for this frequency-dependent direction reversal. The *existence* of direction reversal in this model can be predicted by a simple ''deterministic'' physical theory, although the theory may not be able to reproduce quantitatively the entire flux curve. The results obtained in this study should be useful in designing devices for particle separation based on potential fluctuation.

We are grateful to Dr. Greg Smith for many valuable discussions.

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[12] The eigenvalue-eigenfunction method discussed in [1,2] cannot be applied directly to the kinked ratchet potential in Fig. 1(b), because of the discontinuity of V at $x=0$. In our calculations, the vertical jump at $x=0$ is replaced by a slight tilt to the right and the final result is obtained by extrapolation. The details of the mathematical analysis will be presented elsewhere.