RAPID COMMUNICATIONS

PHYSICAL REVIEW E VOLUME 56, NUMBER 3 SEPTEMBER 1997

Rectification by hopping motion through nonsymmetric potentials with strong bias

K. W. Kehr, K. Mussawisade, and T. Wichmann
Institut für Festkörperforschung, Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany

W. Dieterich
Fakultät für Physik, Universität Konstanz, D-78434 Konstanz, Germany

(Received 2 May 1997)

Hopping motion of particles on linear chains under the influence of bias is considered where the transition rates represent arbitrary potentials. An exact expression for the stationary current is given and verified by numerical simulations. It exhibits rectification effects for nonsymmetric potentials in the regime of strong bias. Applications to two- and three-dimensional systems are indicated. [S1063-651X(97)50909-3]

PACS number(s): 05.40.+j, 68.35.Fx, 87.22.Fy

The motion of particles in nonsymmetric potentials under the influence of stochastic forces is of great current interest for several reasons. One problem is to understand the conditions under which unidirectional motion of the particles can occur. Apparently, there is a connection of this problem to the second law of thermodynamics [1,2]. In addition, there are interesting relations between this model and, e.g., biological systems [2–8] or surface diffusion problems [9,10]. There is agreement that under the influence of thermal noise a particle in a static nonsymmetric potential without bias will not move uniformly in one direction. In the presence of fluctuating external forces with sufficiently long correlation times, however, unidirectional motion can arise; that is, such potentials exhibit rectification properties with respect to the slowly varying components of the external force. Most of the previous work in this area is based on continuous diffusion models that are defined in terms of Langevin-type equations of motion [2–5,7,8,11–13]. However, the apparent relevance of such models to problems of transport on microscopic scales suggest a treatment that at least in principle can account for a specific microscopic environment. The simplest microscopic models may be defined as hopping models for particles on linear chains with discrete binding sites and potential barriers in between.

From this point of view we investigate in this paper the hopping motion of particles in a one-dimensional discrete model without inversion symmetry under the influence of an arbitrary bias in one direction. We will give a quantitative description of rectification effects that can appear in the region of nonlinear response. Rectification effects from a rate equation model for carrier-mediated transport through channels of biological membranes have been discussed previously [14]. Here we treat hopping of particles in an arbitrary sequence of barriers and trapping sites under periodic boundary conditions. Hence, our calculations are also valid for periodic repetitions of potential structures without inversion symmetry. We emphasize that our derivations give a microscopic description of rectification effects of hopping motion through nonsymmetric potentials. Possible applications of our model are outlined at the end of this paper.

Consider a chain consisting of $N+1$ sites $l=0,\dots,N$ with site energies E_l, where we assume periodic boundary conditions, i.e., $E_0=E_N$. In the stationary situation, the current between site l and $l+1$ is given in terms of the site occupation probabilities P_l by

$$J_l=\Gamma_{l+1,l}e^{K/2}P_l-\Gamma_{l,l+1}e^{-K/2}P_{l+1}. \tag{1}$$

$\Gamma_{l+1,l}$ is the rate for a transition from site l to site $l+1$, which is multiplied by the bias factor $e^{K/2}$ and the rate for the reverse transition $\Gamma_{l,l+1}$ is multiplied by the inverse bias factor. It is assumed that an equilibrium state exists in the absence of bias, $K=0$. The condition of detailed balance holds for the equilibrium state,

$$\Gamma_{l+1,l}P_l^{eq}=\Gamma_{l,l+1}P_{l+1}^{eq}, \tag{2}$$

with the equilibrium site probabilities given by

$$P_l^{eq}=\exp(-\beta E_l)\left(\frac{1}{N}\sum_{n=0}^{N-1}\exp(-\beta E_n)\right)^{-1}. \tag{3}$$

In the stationary situation the current between any pair of neighbor sites must be the same, $J=J_l$ for all l. We express the current by the transition rates to the right, $\gamma_l\equiv\Gamma_{l+1,l}$, using the condition of detailed balance (2), and introduce the decomposition $P_l=P_l^{eq}h_l$. We have

$$J=\gamma_l P_l^{eq}(e^{K/2}h_l-e^{-K/2}h_{l+1}). \tag{4}$$

From this equation follows the recursion relation

$$h_{l+1}=e^K h_l-\frac{Je^{K/2}}{\gamma_l P_l^{eq}}. \tag{5}$$

This recursion relation can be brought into the form

$$h_l=e^{lK}h_0-e^{-K/2}Je^{lK}S_l, \tag{6}$$

with

$$S_l=\sum_{n=0}^{l-1}\frac{\exp(-nK)}{\gamma_n P_n^{eq}},\quad S_0=0. \tag{7}$$

We want to eliminate h_0 in favor of S_N. This can be achieved by writing Eq. (6) for $l=N$ and solving with respect to h_0. Note that $h_0\equiv h_N$ as a consequence of the periodic boundary conditions. Inserting the result into Eq. (6) and multiplying with P_l^{eq} we obtain

$$h_l P_l^{eq} = P_l^{eq} e^{(l-1/2)K} J\left(\frac{S_N}{1-\exp(-NK)} - S_l\right). \tag{8}$$

Equation (8) represents the probability of finding the particle on site l in the stationary situation; this quantity must be normalized to unity. From the normalization condition follows the expression for the current per particle

$$J = e^{K/2}\left(\frac{1}{1-\exp(-NK)} \sum_{l=0}^{N-1} f_l \sum_{n=0}^{N-1} \frac{1}{\gamma_n f_n} - \sum_{l=1}^{N-1} f_l \sum_{n=0}^{l-1} \frac{1}{\gamma_n f_n}\right)^{-1}. \tag{9}$$

The quantity f_l is defined by $f_l \equiv P_l^{eq} e^{lK}$. Expression (9) can be regarded as the discrete analogue of the stationary current derived by Ambegaokar and Halperin [15] in their classic work on a driven Smoluchowski system.

A more transparent form for J can be achieved by rearrangements of the sums,

$$J = (e^{NK}-1)\left(\sum_{l=0}^{N-1} \frac{Y_l}{\gamma_l \exp(-\beta E_l)}\right)^{-1}, \tag{10}$$

with

$$Y_l = e^{(N-1/2)K} \sum_{n=0}^{l} e^{(n-l)K - \beta E_n} + e^{-K/2} \sum_{n=l+1}^{N-1} e^{(n-l)K - \beta E_n}, \tag{11}$$

where the last term is taken to be zero if $l = N-1$. Equation (10) is the final result of the formal derivations. It is easily examined on simple examples that the arrangement of barrier and of site energies is relevant for the magnitude of the resulting current, for arbitrary bias.

The well-known result for the linear response regime [16,17] is easily deduced from Eqs. (10) and (11),

$$J = K\left(\sum_{l=0}^{N-1} (\gamma_l P_l^{eq})^{-1}\right)^{-1}. \tag{12}$$

By virtue of detailed balance, Eq. (2), the current Eq. (12) can also be expressed in terms of the left transition probabilities. Rectification effects are absent in the linear-response regime, i.e., the current Eq. (12) simply changes sign if the sign of K is reversed. Note also that the terms under the sum can be arbitrarily rearranged.

Next we consider nonlinear transport in the barrier model with constant site energies $E_l = 0$, where the transition rates are symmetric, $\Gamma_{l+1,l} = \Gamma_{l,l+1}$, cf. also Eq. (2). From Eqs. (10) and (11) we immediately obtain

$$J = 2\sinh\left(\frac{K}{2}\right)\left(\sum_{l=0}^{N-1} \gamma_l^{-1}\right)^{-1}, \tag{13}$$

which has been derived before in the context of transport in systems with quenched disorder [18]. Obviously, also in this expression the contributions of individual barriers can be rearranged, implying the absence of rectification effects. Hence, models that include varying site energies E_l are required for the occurrence of rectification effects.

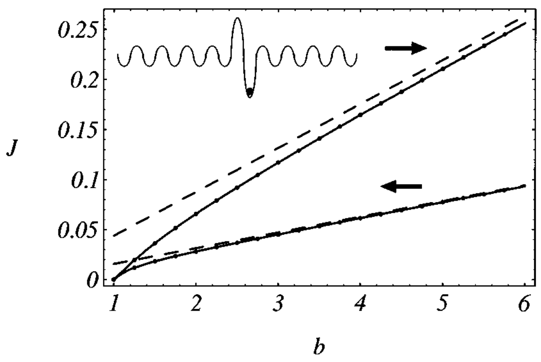

FIG. 1. Particle current J (arbitrary units) as a function of the dimensionless bias parameter b in a model with Schwoebel barriers with $N = 10$ sites (see inset). The relative hopping rates to the right are e^{-2} at the barrier and out of the trap, the rate out of the trap to the left is e^{-4}. Points: simulations, full lines: complete theory, dashed lines: asymptotic theory.

Before we discuss specific cases of rectification, a remark concerning hopping models for the current through membrane channels, which have been studied in biologically motivated works [19–21], is in order. The boundary conditions that are used there differ from those used here. In the works on transport across membranes the concentrations P_0 and P_N at both sites of the membrane are given quantities, corresponding to a chemical bias. In Ref. [21] saturation effects of the channels are included, with the restriction of at most one particle per channel, whereas in this work we normalized the current to exactly one particle per ''channel.'' Some formal similarities between the expression for the current given in [21] and our results can be recovered for large concentrations P_0 and P_N, such that each channel is occupied by a particle.

We now turn to a comparison of our general formula Eqs. (10) and (11) with numerical simulations. We first study the model of particle diffusion in a potential that represents Schwoebel barriers which are relevant for diffusion on surfaces [9,10]. The model is depicted in Fig. 1 (see inset) and it has no inversion symmetry. Results of the numerical simulations are shown in this figure as functions of the bias parameter $b \equiv \exp(|K|/2)$, together with the analytical result according to Eq. (10). One recognizes perfect agreement between theory and simulations.

It is seen that the currents are linear in the bias parameter b for $b \gg 1$. This behavior is easily deduced from the recursion relations Eq. (1). We assume the bias to be so strong that we can neglect the reverse hopping processes. Under this assumption the recursion relations simplify to $(K>0)$

$$J_l = \gamma_l \exp(K/2) P_l. \tag{14}$$

We take $J_l = J$ and solve the equation with respect to P_l. Normalization of the P_l yields

$$J = e^{K/2}\left(\sum_{l=0}^{N-1} \gamma_l^{-1}\right)^{-1}. \tag{15}$$

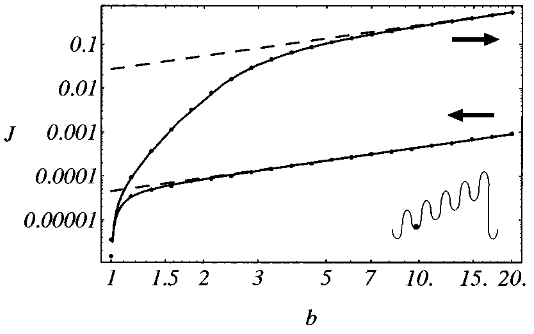

FIG. 2. Particle current J (arbitrary units) as a function of the bias parameter b in a staircase model with $N=5$ sites. The relative right hopping rates are e^{-2}, the relative left hopping rate over the barrier is e^{-10}. Points: simulations, full lines: complete theory, dashed lines: asymptotic theory.

The current in the region of large bias is determined by the inverse of the sum over the inverse transition rates in direction of the bias. If the direction is reversed, $K<0$, we have

$$J=e^{-K/2}\left(\sum_{l=0}^{N-1}(\gamma_l')^{-1}\right)^{-1}, \qquad (16)$$

where $\gamma_l'\equiv\Gamma_{l,l+1}$ are the left transition rates. Equations (15) and (16) are represented by dashed lines in Fig. 1; the complete theory and the numerical results approach the asymptotic behavior.

The result for strong bias immediately suggests that a large ratio between right and left current is achieved by the choice of a potential as depicted in the inset of Fig. 2. The right and left currents in such a model were determined by numerical simulations and they are presented in Fig. 2 together with the full and asymptotic results. There is agreement of the simulations with the complete result and one recognizes that the asymptotic behavior is reached for sufficiently strong bias.

The discrete nature of our model also enables us to provide a general estimate of the cross-over bias beyond which the induced current is driven to the limiting behavior implied by Eqs. (15) and (16). Note that in Figs. 1 and 2 the cross-over to the asymptotic behavior occurs for larger values of b for the bias direction with the larger current, and for smaller b values in the opposite direction. The correction to the asymptotic behavior of the current is governed by hopping processes along the chain with all except one transitions in bias direction. Taking all possibilities of zero and one transitions in the reverse bias direction into account, one obtains the current for $K>0$,

$$J=e^{K/2}\left(\sum_{l=0}^{N-1}\frac{1}{\gamma_l}+e^{-K}\sum_{l=0}^{N-1}\frac{\gamma_{l+1}'}{\gamma_l\gamma_{l+1}}\right)^{-1}, \qquad (17)$$

where the γ_l,γ_l' are defined as above. The corresponding formula for $K<0$ is easily given. Equation (17) predicts bias values where the current is 90% of the asymptotic current, of $b=3$ (right current), $b=1.48$ (left current) for the example of Fig. 1, and $b=7.31$ (right current), $b=1.53$ (left current) for the example of Fig. 2. The figures verify the predictions.

In summary, we have given a microscopic description of rectification effects of hopping motion through nonsymmetric potentials under the influence of strong bias. The connection of our derivations to particle transport through channels in membranes has already been mentioned. We point out that the applicability of this investigation is not restricted to linear systems such as channels with nonsymmetric potentials. The results can also be extended to two- or three-dimensional systems, for instance, to stepped surfaces or to layered systems. If the underlying structure has a nonsymmetric potential in x direction, and periodic, uniform potentials in y and z directions, the results can be applied directly. The reason is that hopping of particles in these directions is independent of the x direction. If a bias is applied in the x direction, the current will then show the nonlinear dependence discussed above, while there is only diffusion in the two other directions. The situation would be completely different if there were defects in the structures. Also the influence of many particles that compete for the sites is not known. See, however, in this context [22,23], where already motion of many particles in nonsymmetric potentials is considered.

We thank G. Schütz and G. Stark for discussions.

---

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

[2] M. O. Magnasco, Phys. Rev. Lett. **71**, 1477 (1993).

[3] R. D. Astumian and M. Bier, Phys. Rev. Lett. **72**, 1766 (1994).

[4] J. Prost, J. F. Chauwin, L. Peliti, and A. Ajdari, Phys. Rev. Lett. **72**, 2652 (1994).

[5] M. O. Magnasco, Phys. Rev. Lett. **72**, 2656 (1994).

[6] M. M. Millonas and D. R. Chialvo, Phys. Rev. Lett. **76**, 550 (1996).

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

[8] J. E. Hall, C. A. Mead, and G. Szabo, J. Membrane Biol. **11**, 75 (1973).

[9] G. Ehrlich, F. G. Hudda, J. Chem. Phys. **44**, 1039 (1966).

[10] R. L. Schwoebel and E. J. Shipsey, J. Appl. Phys. **37**, 3682 (1966); R. L. Schwoebel, J. Appl. Phys. **40**, 614 (1969).

[11] C. R. Doering, W. Horsthemke, and J. Riordan, Phys. Rev. Lett. **72**, 2984 (1994).

[12] I. Zapata, R. Bartussek, F. Sols, and P. Hänggi, Phys. Rev. Lett. **77**, 2292 (1996).

[13] G. A. Cecchi and M. O. Magnasco, Phys. Rev. Lett. **76**, 1968 (1996).

[14] G. Stark, Biochem. Biophys. Acta. **298**, 323 (1973).

[15] V. Ambegaokar and B. I. Halperin, Phys. Rev. Lett. **22**, 1364 (1969).

[16] R. Kutner, D. Knödler, P. Pendzig, R. Przenioslo, and W. Dieterich, in *Diffusion Processes: Experiment, Theory, Simulations*, edited by A. Pekalski, Lecture Notes in Physics Vol.

438 (Springer, Berlin, 1994), p. 197.
[17] R. Kutner, Physica A **224**, 558 (1996).
[18] R. Biller, Z. Phys. B **55**, 7 (1984).
[19] B. J. Zwolinski, H. Eyring, and C. E. Reese, J. Phys. Chem. **53**, 1426 (1949).
[20] F. H. Johnson, H. Eyring, and M. I. Polissar, *The Kinetic Basis of Molecular Biology* (Wiley, New York, 1954), Chap. 14.
[21] P. Läuger, Biochem. Biophys. Acta. **311**, 423 (1973).
[22] I. Derenyi and T. Vicsek, Phys. Rev. Lett. **75**, 374 (1995).
[23] F. Marchesoni, Phys. Rev. Lett. **77**, 2364 (1996).