Rectification Through Entropic Barriers

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Abstract. The dynamics of Brownian motion has widespread applications extending from transport in designed micro-channels up to its prominent role for inducing transport in molecular motors and Brownian motors. Here, Brownian transport is studied in micro-sized, two dimensional periodic channels, exhibiting periodically varying cross sections. The particles in addition are subjected to a constant external force acting alongside the direction of the longitudinal channel axis. For a fixed channel geometry, the dynamics of the two dimensional problem is characterized by a single dimensionless parameter which is proportional to the ratio of the applied force and the temperature of the environment. In such structures entropic effects may play a dominant role. Under certain conditions the two dimensional dynamics can be approximated by an effective one dimensional motion of the particle in the longitudinal direction. The Langevin equation describing this reduced, one dimensional process is of the type of the Fick-Jacobs equation. It contains an entropic potential determined by the varying extension of the eliminated transversal channel direction, and a correction to the diffusion constant that introduces a space dependent diffusion. We analyze the influence of broken channel symmetry and the validity of the Fick-Jacobs equation. For the nonlinear mobility we find a temperature dependence which is opposite to that known for particle transport in periodic energetic potentials. The influence of entropic effects is discussed for both, the nonlinear mobility, and the effective diffusion constant. In case of broken reflection symmetry rectification occurs and there is a favored direction for particle transport. The rectification effect could be maximized due to the optimal chosen absolute value of the applied bias.

1 Introduction

The phenomenon of entropic transport is ubiquitous in biological cells, ion channels, nano-porous materials, zeolites and microfluidic devices etched with grooves and chambers. Instead of diffusing freely in the host liquid phase the Brownian particles frequently undergo a constrained motion. The geometric restrictions to the system's dynamics results in entropic barriers and regulate the transport of particles yielding important effects exhibiting peculiar properties. The results have prominent implications in processes such as catalysis, osmosis and particle separation [1–12] and, as well, for the noise-induced transport in periodic potential landscapes that lack reflection symmetry (Brownian ratchet systems) [13–15] or Brownian motor transport occurring in arrays of periodically arranged asymmetric obstacles, termed "entropic" ratchet devices [16–20]. Motion in these systems can be induced by imposing different concentrations at the ends of the channel, or by the presence of external driving forces supplying the particles with the energy necessary to proceed. The study of the kinetics of the entropic transport, the properties of transport coefficients in far from equilibrium situations and the possibility for transport control mechanisms are pertinent objectives in the dynamical characterization of those systems.

Because the role of inertia for the motion of the particles through these structures can typically be neglected the Brownian dynamics can safely be analyzed by solving the Smoluchowski equation in the domain defined by the available free space upon imposing the appropriate boundary conditions. Whereas this method has been very successful when the boundaries of the system possess a rectangular shape, the challenge to solve the boundary value problem in the case of nontrivial, corrugated domains represents a difficult task. A way to circumvent this difficulty consists in coarsening the description by reducing the dimensionality of the system, keeping only the main direction of transport, but taking into account the physically available space by means of an entropic potential. The resulting kinetic equation for the probability distribution, the so called Fick-Jacobs (FJ) equation, is similar in form to the Smoluchowski equation, but now contains an entropic term. The entropic nature of this term leads to a genuine dynamics which is distinctly different from that observed when the potential is of energetic origin [21]. It has been shown that the FJ equation can provide a very accurate description of entropic transport in channels of varying cross-section [21–24]. However, the derivation of the FJ equation entails a tacit approximation: The particle distribution in the transversal direction is assumed to equilibrate much faster than in the main (unconstrained) direction of transport. This equilibration justifies the coarsening of the description leading in turn to a simplification of the dynamics, but raises the question about its validity when an *external force is applied*. To establish the validity criterion of a FJ description for such biased diffusion in confined media is, due to the ubiquity of this situation, a subject of primary importance.

Our objective with this work is to investigate in greater detail the FJ-approximation for biased diffusion and to study rectification due to the asymmetry of a geometrical confinement. We will analyze the biased movement of Brownian particles in 2D periodic, but asymmetric channels of varying cross-section. On the basis of our numerical and analytical results we recapitulate the striking and sometimes counterintuitive features [21], which arises from entropic transport and which are different from those observed in the more familiar case with energetic, metastable landscapes [25].

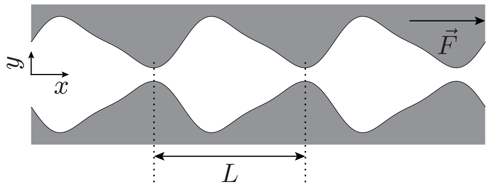

Fig. 1. Schematic diagram of a channel confining the motion of forced Brownian particles. The half-width ω is a periodic function of x with periodicity L.

2 Diffusion in confined systems

Transport through pores or channels (like the one depicted in Fig. 1) may be caused by different particle concentrations maintained at the ends of the channel, or by the application of external forces acting on the particles. Here we will exclusively consider the case of force driven transport. The external driving force is denoted by $\boldsymbol{F} = F\boldsymbol{e}_x$. It points into the direction of the channel axis. In general, the dynamics of a suspended Brownian particles is overdamped [26] and well described by the Langevin equation in dimensionless variables [23],

$$\frac{\mathrm{d}\boldsymbol{r}}{\mathrm{d}t} = \boldsymbol{f} + \boldsymbol{\xi}(t)\,, \tag{1}$$

where t is dimensionless time, $\boldsymbol{r}$ corresponds to the position vector of the particle (given in units of the period length L), $\boldsymbol{\xi}$ to Gaussian white noise with $\langle \boldsymbol{\xi}(t) \rangle = 0$ and $\langle \boldsymbol{\xi}(t)\boldsymbol{\xi}(t') \rangle = 2\delta_{i,j}\delta(t - t')$ for $i, j = x, y, z$ and with the dimensionless force

$$\boldsymbol{f} = f\boldsymbol{e}_x \ \text{ and } f = \frac{LF}{k_\mathrm{B}T}\,. \tag{2}$$

The dimensionless parameter f characterizes the force as the ratio of the work which it performs on the particle along a distance of the length of the period L and the thermal energy $k_\mathrm{B}T$.

The boundary of the 2D periodic channel which is mirror symmetric about its axis is given by the periodic functions $y = \pm\omega(x)$, i.e. $\omega(x + 1) = \omega(x)$ for all x, where x and y are the Cartesian components of $\boldsymbol{r}$. Except for a straight channel with $\omega = const$ there are no periodic channel shapes for which an exact analytical solution of Eq. (1) and the corresponding Fokker-Planck equation with boundary conditions is known [27, 28]. Approximate solutions though can be obtained on the basis of an one dimensional diffusion problem in an effective potential. Narrow channel openings, which act as geometric hindrances in the full model, show up as entropic barriers in this one dimensional approximation [21–23, 29–32]. This approach is valid under conditions that will be discussed below in some detail.

3 Transport in periodic channels with broken symmetry

3.1 The Fick-Jacobs approximation

In the absence of an external force, i.e. for $\boldsymbol{f} = 0$, it was shown [29–32] that the dynamics of particles in confined structures (such as that of Fig. 1) can be described approximatively for $|\omega'(x)| \ll 1$ by the FJ equation, with a spatial dependent diffusion coefficient:

$$\frac{\partial P(x,t)}{\partial t} = \frac{\partial}{\partial x} D(x) \left(\frac{\partial P(x,t)}{\partial x} - \frac{\omega'(x)}{\omega(x)} P(x,t) \right) , \tag{3}$$

obtained from the full 2D Smoluchowski equation upon the elimination of the transversal y coordinate assuming fast equilibration in that direction. Here $P(x,t) = \int_{-\omega(x)}^{\omega(x)} \mathrm{d}y \, P(x,y,t)$ denotes the marginal probability density along the axis of the channel. We note that for three dimensional channels an analogue approximate Fokker-Planck equation holds in which the function $\omega(x)$ is to be replaced by $\pi\omega^2(x)$ (area of cross-section). The prime refers to the derivative of the function with respect to its argument, i.e. $\omega'(x) = \mathrm{d}\omega/\mathrm{d}x$. In the original work by Jacobs [29] the 1D diffusion coefficient $D(x)$ is constant and equals the bare diffusion constant which is unity in the present dimensionless variables. Later, Zwanzig [30] and Reguera and Rubí [31] proposed different spatially dependent forms of the 1D diffusion coefficient which allows for an extended regime of validity of the FJ-description.

Reguera and Rubí [31] put forward this form of the 1D diffusion coefficient:

$$D(x) = \frac{1}{(1 + \omega'(x)^2)^\gamma} , \tag{4}$$

where $\gamma = 1/3$ for 2D structures and $\gamma = 1/2$ for 3D systems. The right hand side of Eq. (4) can be considered as a resummation of Zwanzig's original perturbational result [30].

In the presence of a constant force F along the direction of the channel the FJ equation (3) can be recast into the form [21–23, 31]:

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial x} D(x) \left(\frac{\partial P}{\partial x} + \frac{\mathrm{d}A(x)}{\mathrm{d}x} P \right) \tag{5}$$

with the dimensionless free energy $A(x) := E - S = -f\, x - \ln \omega(x)$. In physical dimensions the energy is $\tilde{E} \equiv k_\mathrm{B} T E = -F\tilde{x}$ ($\tilde{x} = xL$) and the dimensional entropic contribution is $\tilde{S} \equiv k_\mathrm{B} T S = k_\mathrm{B} T \, \ln \omega$. For a periodic channel with broken reflection symmetry this free energy assumes the form of a tilted periodic, ratchet-like potential. In the absence of a force the free energy is purely entropic and Eq. (5) reduces to the FJ equation (3). On the other hand, for a straight channel the entropic contribution vanishes and the particle is solely driven by the external force.

3.2 Transport characteristics

Key quantities of particle transport through periodic channels are the average *particle current*, or equivalently the nonlinear *mobility*, and the *effective diffusion coefficient*. For a particle moving in a one dimensional tilted *energetic* periodic potential the heights ΔE of the barriers separating the potential wells provide an additional energy scale apart from the work of the force FL and the thermal energy $k_B T$. Hence, at least two dimensionless parameters, say $\Delta E/(k_B T)$ and $FL/(k_B T)$ govern the transport properties of these systems. In contrast, as already noted in the context of the full 2D model the transport through channels is governed by the single dimensionless parameter $f = FL/(k_B T)$ [21–23]. This, of course, remains to hold true in the one dimensional approximation which models the transversal spatial variation in terms of an entropic potential.

For any non negative force the average particle current in periodic structures can be obtained from mean-first-passage time analysis [21–23, 34, 35], i.e. the average particle current $\langle \dot{x} \rangle$ is given as ratio of period length L and the mean-first-passage time $\langle T \rangle$ for a particle to overcome one period length, i.e. in dimensionless units: $\langle \dot{x} \rangle = 1/\langle T \rangle$.

The nonlinear mobility $\mu(f)$ is defined by

$$\mu(f) = \frac{\langle \dot{x} \rangle}{f} \,. \tag{6}$$

Consequently, one can obtain the following Stratonovich formula for the nonlinear mobility [21–23]

$$\mu(f) = \frac{1 - \exp(-f)}{f \displaystyle\int_0^1 \mathrm{d}z \, I(z, f)} \,, \tag{7}$$

where

$$I(z, f) := \frac{h^{-1}(z)}{D(z)} \exp(-f\, z) \int_{z-1}^{z} \mathrm{d}\tilde{z} \, h(\tilde{z}) \exp(f\, \tilde{z}) \,, \tag{8}$$

depends on the dimensionless position z, the force f and the shape of the tube given in terms of the half width $\omega(x)$ and its first derivative.

The effective diffusion coefficient of the movement alongside the channel axis is defined as the asymptotic behavior of the variance of the position

$$D_{\text{eff}} = \lim_{t \to \infty} \frac{\langle x^2(t) \rangle - \langle x(t) \rangle^2}{2t} \,. \tag{9}$$

It is related to the first two moments of the first passage time $\langle T \rangle$ and $\langle T^2 \rangle$ by the expression [34–36]:

$$D_{\text{eff}} = \frac{\langle T^2 \rangle - \langle T \rangle^2}{2 \, \langle T \rangle^3} \,. \tag{10}$$

After some algebra it can be transformed to read

$$D_{\mathrm{eff}} = \frac{\displaystyle\int_0^1 \mathrm{d}z \int_{z-1}^{z} \mathrm{d}\tilde{z}\, \mathcal{N}(z,\tilde{z},f)}{\left[\displaystyle\int_0^1 \mathrm{d}z\, I(z,f)\right]^3}\,, \tag{11}$$

where

$$\mathcal{N}(z,\tilde{z},f) := \frac{D(\tilde{z})}{h(z)}\,\frac{h(\tilde{z})}{D(z)}\,[I(\tilde{z},f)]^2 \exp(-f\,z + f\,\tilde{z})\,. \tag{12}$$

The predicted dependence of the average particle current and the effective diffusion coefficient was compared with 2D Brownian dynamic simulations performed by a numerical integration of the Langevin equation (1), within the stochastic Euler-algorithm. The shape of the exemplarily taken 2D channel is described (in dimensionless units) by

$$\omega(x) := \sin(2\pi x) + 0.25\,\sin(4\pi x) + 1.12\,. \tag{13}$$

For the considered channel configuration, the widest opening of the channel is by a factor of 116 larger than the width at the narrowest openings, i.e. at the bottlenecks. One may therefore expect strong entropic effects for these channels. The particle current and effective diffusion coefficient were derived from an ensemble-average of about $3 \cdot 10^4$ trajectories:

$$\langle \dot{x} \rangle = \lim_{t\to\infty} \frac{\langle x(t) \rangle}{t}\,, \tag{14}$$

and Eq. (9), respectively.

Transport in one dimensional periodic *energetic* potentials behaves very differently from one dimensional periodic systems with *entropic* barriers [21]. The fundamental difference lies in the temperature dependence of these models. Decreasing temperature in an energetic periodic potential decreases the transition rates from one period to the neighboring by decreasing the Arrhenius factor $\exp\{-\Delta V/(k_{\mathrm{B}}T)\}$ where ΔV denotes the activation energy necessary to proceed by a period [25]. Hence decreasing temperature leads to a decrease in mobility. For a one dimensional periodic system with an entropic potential, a decrease of temperature leads to an increase of the dimensionless force parameter f and consequently to an increase of the mobility, cf. Fig. 2.

The reduction of dimensionality leading to the FJ equation relies on the assumption of equilibration in the transversal direction which results in an almost uniform distribution of the transversal positions y at fixed values of the longitudinal coordinate x. One can formulate criteria determining whether the FJ equation approximatively describes the stationary state of the considered problem [23]. For channels with varying width the narrow positions confine the positions of the particles. From there they are dragged by the force and

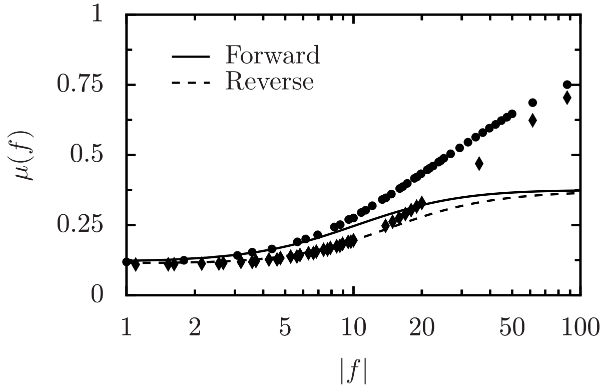

Fig. 2. The numerically simulated (*symbols*) and analytically calculated (cf. Eq. (7) – lines) dependence of the absolute value of the nonlinear mobility $\mu(f)$ *vs.* the dimensionless force $f = FL/k_{\mathrm{B}}T$ is depicted for a 2D channel with the scaled half-width given by $\omega(x) = \sin(2\pi x) + 0.25\sin(4\pi x) + 1.12$; for transport in positive x-direction, i.e. positive f: *circles and solid line*, for negative f-values: diamonds and *dashed line*. For the linear response regime, i.e. small $|f|$, the nonlinear mobility for forward and backward transport converge to each other.

– at the same time – they perform a diffusive motion until the channel narrows again. The required uniform distribution in the transversal direction can only be achieved if the diffusional motion is fast enough in comparison to the deterministic drift under the influence of f. Therefore the time scale of equilibration in transversal direction must be short compared to the time it takes to drag a particle from the narrow position to the position with largest channel width. The latter requirement leads to an estimate of the minimal forcing above which the FJ description is expected to fail in providing an accurate description of the transport properties in the long time limit [22, 23]. Detailed analysis demonstrates that, for the considered asymmetric channel, cf. Fig. 1, the FJ description holds for larger force value when forcing towards the negative x-direction than for forcing in the positive x-direction, cf. Fig. 2. The dependence of the nonlinear mobility on the direction of the forcing which arises due to the asymmetry of the shape of the channel walls is addressed in Sect. 3.3.

Another interesting effect can be observed for the effective diffusion if looked as a function of the force f. Already the expression for the effective diffusion (11) which follows rigorously from the FJ equation displays a maximum as a function of f which may even exceed the value 1 of the bare diffusion, cf. Fig. 3. For $f \to \infty$ the effective diffusion approaches the bare value 1. If one decreases the force to finite but still large values then the stationary distribution acquires a finite width in the transversal direction with a "crowded" region in front of the narrowest place of the channel Refs. [22, 23]. The transport becomes more noisy and consequently the effective diffusion exceeds the

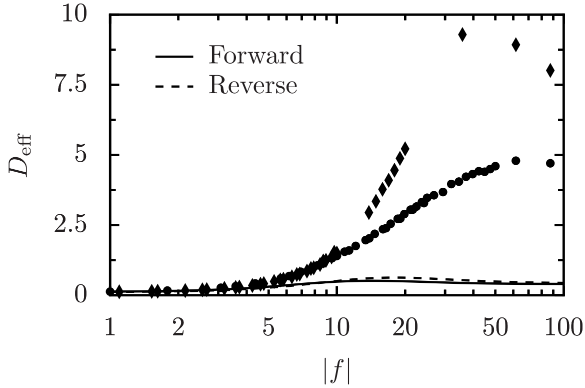

Fig. 3. The numerically simulated (*symbols*) and analytically calculated (cf. Eq. (11) – lines) dependence of the effective diffusion coefficient D_{eff} is depicted *vs.* the dimensionless force $f = FL/k_BT$ for two channels in 2D. For both channels the scaled half-width is given by $\omega(x) = \sin(2\pi x) + 0.25\sin(4\pi x) + 1.12$; transport in positive x direction ($f > 0$): *circles and solid line*; transport in negative x direction ($f < 0$): *diamonds and dashed line.*

bare value 1. On the other hand if one starts at $f = 0$ the entropic barriers diminish the diffusion such that the effective diffusion is less than bare diffusion. Consequently, somewhere in between there must be a value of f with maximal effective diffusion [21–23]. For the considered 2D channel defined by Eq. (13) the value of the force at the maximal effective diffusion is outside the regime of validity of the FJ equation for both forcing directions. The numerical simulations give a much more pronounced peak of the effective diffusion. These observations lead us to the conclusion that entropic effects increase the randomness of transport through a channel and in this way decrease the mobility and increase the effective diffusion. A similar enhancement of effective diffusion was found in titled periodic *energetic* potentials [33–36].

3.3 Rectification in asymmetric channels

Due to different focussing towards the narrowest width of the channel, i.e. the bottlenecks, which is a consequence of the broken reflection symmetry of the channel, the nonlinear mobility depends on the direction of the constant bias and not only on its absolute value, cf. Fig. 2. Moreover, within the FJ description an asymmetric shape of the channel walls leads to a ratchet-like free energy landscape facilitating the transport in one direction. The rectification thereby not only depends on the channel geometry but also on f. To quantify the rectification effect, we define the quantity

$$\alpha = \frac{|\mu(f) - \mu(-f)|}{\mu(f) + \mu(-f)}\,, \tag{15}$$

as a rectification-measure.

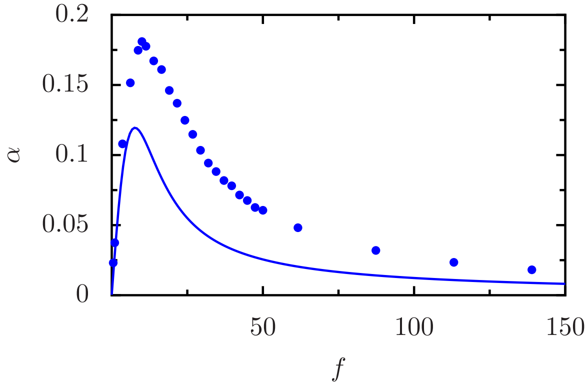

Fig. 4. The numerical simulated dependence of the rectification measure α, cf. Eq. (15), giving the relative discrepancy of the nonlinear mobilities in positive and negative x-direction on the force value f is depicted by the *symbols*. There is an optimal forcing value for which the rectification for the given 2D channel, cf. Eq. (13), is maximal. Within the FJ description (*solid line*) the same qualitative behavior could be observed.

In Fig. 4 the dependence of the rectification measure α on the force f is depicted. Interestingly, there is an optimal value for f where the rectification is maximum. Due to the favoring of transport in one direction for finite f-values there is rectification, whereas for $f \to 0$ (linear response) and $f \to \infty$ (corresponding to a flat channel geometry [22, 23]) the nonlinear mobilities for forward and backward propagation equal each other, i.e. $\mu(f) = \mu(-f)$. Consistently, within the FJ approximation the qualitative behavior could be observed.

4 Conclusions

In summary, we demonstrated that transport phenomena in periodic channels with varying width exhibit some features that are radically different from conventional transport occurring in energetic periodic potential landscapes. The most striking difference between these two physical situations lies in the fact that for a fixed channel geometry the dynamics is characterized by a single parameter $f = FL/(k_{\mathrm{B}}T)$ which combines the external force F causing a drift, the period length L of the channel, and the thermal energy $k_{\mathrm{B}}T$, which is a measure of the strength of the acting fluctuating forces. Transport in periodic energetic potentials depends, at least, on one further parameter which is the activation energy of the highest barrier separating neighboring periods. This leads to an opposite temperature dependence of the mobility. While the mobility of a particle in an energetic potential increases with increasing temperature the mobility of a particle in a channel of periodically varying width decreases. The incorporation of the spatial variation of the channel width as an entropic potential in the FJ equation allows a qualitative understanding of the dependence of the transport properties on the channel geometry. In channels without a mirror symmetry about a vertical axis rectification favoring transport in one channel direction occurs. An optimal forcing regime could be found for which the rectification effect is maximal.

The effective diffusion exhibits a non monotonic dependence versus the dimensionless force f. It starts out at small f with a value that is less than the bare diffusion constant, reaches a maximum with increasing f and finally approaches the value of the bare diffusion from above.

Under certain conditions, the two dimensional Fokker-Planck equation governing the time dependence of the probability density of a particle in the channel can be approximated by one dimensional Fokker-Planck equation: the approximated equation is termed the Fick-Jacobs equation; it contains an entropic potential and a position dependent diffusion coefficient. In principle the FJ equation describes both the transient behavior of a particle and also the stationary behavior of the particle dynamics which is approached in the limit of large times. In this paper we demonstrated the suitability of the FJ approximation on describing the biased Brownian motion in periodic channels with broken symmetry where rectification takes place. Although we restricted our discussion to two dimensional channels, a generalization of the presented methods to three dimensional pores with varying cross section is straight forward.

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