Directed molecular transport in an oscillating channel with randomness

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(Received 13 November 2007; published 14 February 2008)

Stability of directed transport and molecular separation in a *symmetric* channel is analyzed. The original mechanism is based on *harmonic* spatial oscillations of the channel, under which the system exhibits multiple regimes of a directed transport. The particles may be forced to move with different velocities and directions as the amplitude and/or frequency of the oscillations are adjusted to a proper *resonance*. The advantage of this mechanism in contrast to the ratchet systems is that the average particle velocity is larger than the velocity of the growing of the width of the particle spatial distribution. We have studied the stability of the directed transport with regard to random impacts to the channel parameters and oscillation frequency. Here we present the results of the simulations which show that the ability of the combined longitudinally and transversally vibrating randomized dynamic channel to perform directed molecular transport remains resilient to quite intensive random channel structure fluctuations (50–60 %) and relatively strong random impacts to its oscillations (15–20 %).

DOI: 10.1103/PhysRevE.77.021114 PACS number(s): 05.60.Cd, 05.45.−a, 87.16.Uv, 81.40.Pq

I. INTRODUCTION

Biological movements display highly complex phenomena involving biochemical and biophysical processes as well as strongly nonlinear multibody dynamics with noise, dissipation, self-organization, and self-regulation. An interdisciplinary research field has thus developed which attracts great interest of many specialists involved in very different areas, from biology to nanotribology. In recent years, there has been an increasing interest in the studies of the pumping of ions, molecules, and colloids through micro- and nanoscale channels. These investigations have been motivated by a desire to understand how biological molecular pumps operate and to develop new strategies for the fabrication of synthetic pumps [1–12].

Traditionally rigid channels without fluctuations of their structure were considered, in which particles were moving in spatially asymmetric ratchet potentials. Under nonequilibrium conditions these potentials can induce a directed drift, additional to the ordinary diffusion [13–15]. However, the dynamics of the channel proteins' internal degrees of freedom are found to be important in many ways for an appearance of net directional motion [16,17]. In paper [7] a pumping mechanism driven by spatial fluctuations of a symmetric channel was proposed. It has been shown that the pumping and separation of particles can be achieved by correlated oscillations of the channel walls in the lateral and normal directions. The space oscillations modulate the particle-wall interactions producing temporally asymmetric particle-driving forces, so the total temporally spatial symmetry of the system is broken to get directed transport.

The advantage of a pump based on such a mechanism is that it dynamically determines the direction of motion and does not require any spatial asymmetry of the channel. The transport velocity can also be varied within a wide range and particles may separate according to their masses and/or interaction with the channel walls. This system is closely related to other recently proposed molecular engines possessing the general concept of dynamic control of motion for which no static asymmetry is required [18–22,26,27]. Similar mechanism has also been published recently in [28,29], where the wall effects in directed transport were studied in the Fokker-Plank approach.

The characteristic property of the mechanism is that the average particle velocity is larger than the velocity of the growing of the width of the particle spatial distribution. This allows an effective manipulation by the ensemble of particles that is impossible in most ratchet systems where both velocities are of the same order. The model can be applied to real biological systems with some limitations. In particular, the motion of particles in the biological systems is strongly damped. It was shown in [7] that the proposed mechanism of pumping allows to produce directed motion also under overdamped conditions. It was found that the maximal value of current in the overdamped case is smaller than that for the underdamped conditions, but the parametric resonances are even more pronounced. The particles cannot be separated according their masses however the separation is still possible, e.g., according to their friction coefficient and interaction with the channel. The direction of the motion is dictated by an asymmetry of the drive and does not depend of the frequency and the amplitude of modulation.

The focus on particle motion investigations in a considerably idealized case of the periodic channel with perfectly harmonic dynamic oscillations is an important weakness of previous work recorded in [7]. In biologically related flexible systems, the transport should be induced by spatial fluctuations of the nonideal channel walls with a complex molecular structure. Randomness of the system was partially accounted for in the form of thermal noise acting on the moving particles. It has been shown that thermal noise, depending on the parameters of the system, can speed up or slow down the directed transport. However, for numerical simplicity of the

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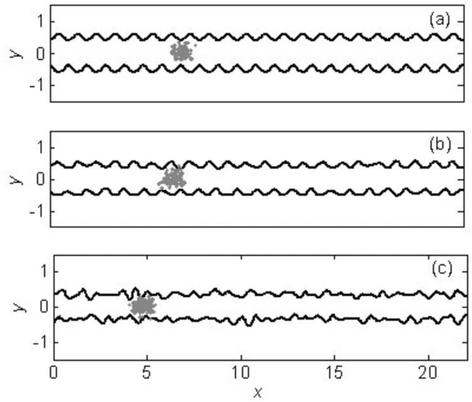

FIG. 1. Conceptual picture of directed transport in a symmetric channel with different fractions of randomness: (a) A two-dimensional snapshot of the embedded system obtained after ten oscillations in a purely periodic channel with $\nu=0$, (b) the same snapshot of the system with $\nu=0.25$, and (c) strongly randomized channel with $\nu=0.5$. The instantaneous positions of the particles of masses $m=1$ are shown by gray circles. Other parameter values are the same as in [7]: $\omega/\omega_0(m)=1$, $\eta/[m\omega_0(m)]=0.89$, $k_BT/U_0=0.02$, $\Delta B/l=1.3$, $\Delta A/A_0=0.85$, and $\lambda/A_0=0.5$.

model the randomness was completely ignored in the structure of the channel and in the fluctuations of the frequency and amplitudes of its oscillations.

In the present paper the model has been made more realistic. In particular, we tested the effect of nonideality of the channel and of its oscillations on the directed transport, proving that the phenomenon of directed transport in a fluctuating channel holds under strong (1) random perturbations of the channel's periodic structure, (2) stochastic fluctuations of the transversal channel wall vibrations, and (3) stochastic fluctuations of the longitudinal vibrations.

To perform these studies, random impact to the channel potential is generated numerically in the same manner as applied in [23,24]. Henceforth, this potential is combined with the periodic potential with changeable weight ν. This weight numerically describes a "*fraction*" of the statistically independent randomness contained in the total potential of each wall. At each fraction ν we repeat the same simulation procedure as performed in [7]. We first fix a set of parameters corresponding to the well pronounced transport phenomenon in the regular system, and then check the effect of the randomness at various ν. This allows the threshold ν_{crit} to be defined, up to which the effect of directed transport is preserved.

II. MODEL

In order to investigate the effect of randomness and temporal fluctuations on the characteristic properties of directed motion, we use essentially the same model as in [7]. The model incorporates a statistical ensemble of particles embedded between two walls. The walls oscillate in both the normal and the lateral directions (see conceptual structure of the model presented in Fig. 1). The dynamic behavior of each particle is described by the two-dimensional Langevin equation of motion [25]

$$m\ddot{\mathbf{r}} = -\eta\dot{\mathbf{r}} - \nabla_{\mathbf{r}}U(\mathbf{r}) + \mathbf{f}(t). \tag{1}$$

Here m and $\boldsymbol{r}=(x,y)$ are the mass and coordinate of a particle and η is the friction coefficient. The effect of thermal motion on the embedded particle is given by a random force $\boldsymbol{f}(t)$, which is δ correlated: $\langle f_i(t)f_j(0)\rangle = 2\eta k_B T\delta(t)\delta_{ij}$, where T is the temperature and k_B is the Boltzmann constant.

The interaction between the particle and the channel is represented by the potential $U(\mathbf{r})$. In contrast to the original model, the potential $U(\mathbf{r})$ now includes two parts: $U(\mathbf{r})=(1-\nu)U_1(\mathbf{r})+\nu U_2(\mathbf{r})$. The first term, $U_1(\mathbf{r})$, is regular and periodic along the channel, $U_1(\mathbf{r})=U_1(x)U(y,t)$, where

$$U_1(x) = U_0[\cos\{2\pi[x+B(t)]/l\} + \sigma]. \tag{2}$$

This potential represents the interaction with channel walls and l is the periodicity in x direction.

The second part of the potential, $U_2(\mathbf{r})$, is a numerically generated random potential with scale-invariant structure. It incorporates one of the important features of a realistic physical and biological system where the mesoscopic structure of surface is of almost random scale-invariant character and thereby cannot be characterized by a definite wave vector (or even few wave vectors as normally applied in simulations of Brownian molecular engines). In numerical simulation, $U_2(\mathbf{r})$ can be generated in the form [18,19] $U_2(\mathbf{r}) = U_2(x)U(y,t)$, where

$$U_2(x) = U_0\int_{q_1}^{q_2} dq\, c(q)\cos(qx+\zeta). \tag{3}$$

This follows the scale-invariant density of spectrum $c(q) = q^{-\beta}$, and characteristic cutoff wave vectors q_1 and q_2. The function $\zeta(x)$ represents a random phase that we assume to be δ correlated $\langle\zeta(q)\zeta(q')\rangle = 2\pi\delta(q-q')$.

The coefficient ν in the combination $U(\mathbf{r})=(1-\nu)U_1(\mathbf{r}) + \nu U_2(\mathbf{r})$ defines a fraction of the random term $U_2(\mathbf{r})$ in the total potential. When $\nu\to 0$ the problem naturally degenerates into regularity. In the opposite limit $\nu\to 1$, the channel walls become completely random. In this context, our problem can be reduced to the determining of a critical fraction value $\nu=\nu_{\mathrm{crit}}$ at which an effect of directed transport disappears.

In numerical study, the integral in Eq. (3) transforms into its discrete representation: $\int dq\, c(q)\to\Sigma_q$. Here a discrete step between the wave vectors Δq is determined by the smallest vector q_1 corresponding to an inverse maximal length $l_{\max}$ of the system which equals normally to its size $l_{\max}=L$. Total number of terms N in the sum is given by $N_{\mathrm{mode}\ s}=q_2/q_1\equiv q_2/\Delta q$. However, for theoretical generality and for accumulation of statistical data for the directed flux of particles, it is important to be able to extend a channel to "infinity" and continue the calculation procedure as long as necessary. This does not cause any problem for the analytically defined periodic potential term. That said, it does mean that numerically generated potential $U_2(\mathbf{r})$ should be extendable to an infinite run also. For this reason, instead of Eq. (3), one may use the following differential definition of random potential: $\partial U_2(x)/\partial x = U_0 \Delta x \Sigma_j q_j c(q_j) \sin(q_j x + \xi)$, where $j = 1, 2, \ldots, N_{\text{mode } s}$. This procedure naturally extends $U_2(x)$ infinitely each time the x coordinate runs out of the instant array bounds.

The possibility of regular channel width and lateral wall position oscillations is taken into account by introducing a time dependence into the width $A(t)$, and the phase of the potential $B(t)$:

$$U(y,t) = [\exp\{[y - A(t)/2]/\lambda\} + \exp\{-[y + A(t)/2]/\lambda\}]. \tag{4}$$

λ is the characteristic length of particle-wall interaction in the y direction.

For the sake of simplicity, it was assumed in [7] that both lateral and normal fluctuations follow harmonic law, $B = \Delta B \cos(\omega t)$ and $A = A_0 + \Delta A \cos(\omega t)$, with the same frequency, ω. The expansion of the channel is accompanied by lateral displacement of the walls to the right, while the narrowing of the channel occurs with the displacement to the left and leads to a modulation of the particle-wall interaction: the amplitude of the periodic potential $U(x, y = \text{const})$ reaches a maximum at the minimum width of the channel and a minimum at the channel's maximum width. As a result, temporally asymmetric forces acting on the particles are induced by coupling of normal and lateral oscillations. The time average of the external force during this process is zero.

The above correlation seems to be very important for the effect. In principle, one can expect a strong correlation between lateral and normal oscillations of the channel in biological (or artificial) systems which are controlled by external signals. As was found in [7] the effect of directed transport has a strongly pronounced resonant nature, which can in principle completely disappear, even at extremely small random impacts to the amplitude or frequency of the harmonic oscillations. It is therefore important to check its stability under such fluctuations.

To accomplish this, below we vary all the parameters in the relations $B = \Delta B \cos(\omega t)$ and $A = A_0 + \Delta A \cos(\omega t)$: $\Delta A \to \Delta A + \delta A(t)$, $\Delta B \to \Delta B + \delta B(t)$, and $\omega \to \omega + \delta\omega(t)$, with random functions $\delta A(t)$, $\delta B(t)$, and $\delta\omega(t)$. The last type of fluctuations makes the analytical relation $A = A_0 + \Delta A \cos(\omega t)$ senseless, because it formally redefines the amplitude of the oscillation not only for an instant in time, but for all the time elapsed from the very beginning of the process. In this case we have to redefine amplitude A using the same procedure applied for the infinitely extendable random potential: $\partial A/\partial t = -\Delta A \sin(\omega t)$ with initial condition $A(t=0) = A_0$.

III. RESULTS AND DISCUSSION

At small relative fluctuations of the periodic potential ν and oscillations $\delta A(t)$, $\delta B(t)$, and $\delta\omega(t)$, physical behavior of the system nearly coincides with that observed in [7]. One can reproduce all typical examples of the symmetric channel time evolution of an ensemble of noninteracting particles driven by spatial oscillations. Following the same procedure we take $N = 200$ particles of different masses placed at the initial time around the potential minimum at the point $r = (0,0)$, and reproduce the main features of the model. In particular it is found that (a) the spatial oscillations of the channel lead to directional motion of the embedded particles both to the right and to the left and (b) both direction and velocity of motion depend not only on the driving parameters ω and ΔB but also on the particle masses.

The system also reproduces the main characteristic property of this mechanism of directed motion: The average particle velocity is larger than the rate of growth of the particle spatial distribution's width. This is impossible in most ratchet systems where both velocities are of the same order. As usual, to characterize pumping we introduce an average current as $J = \lim_{t\to\infty} (1/Nt) \Sigma_{j=1}^{N} \int_0^t \dot{x}_j(t') dt'$. To extract numerical information about directed transport through the channel we also follow a time-dependent displacement averaged over the ensemble of realizations, $\langle x(t) \rangle = (1/N) \Sigma_{j=1}^{N} x_j(t)$. At negligible ν, $\delta A(t)$, $\delta B(t)$, and $\delta\omega(t)$ the dynamic behavior of the system can be characterized by the same dimensionless parameters: $k_B T/U_0$, $\Delta B/l$, $\Delta A/A_0$ and λ/A_0, ω/ω_0, $\eta/(m\omega_0)$, and $\omega_0 = (2\pi U_0/lm)^{1/2}$ as in the regular case. Further, it can be summarized in an analogous set of dynamic scenarios, presented previously in a set of phase diagrams in [7].

Now, to analyze the stability of these scenarios, one can choose any point in the appropriate diagram and vary one of the parameters ν, $\delta A(t)$, $\delta B(t)$, and $\delta\omega(t)$ (or a few of them at once). We examined different combinations of the parameters and found a general answer: The channel retains its ability to perform direct transport up to relatively large fluctuations of its structure and oscillation parameters. This ability has a threshold and remains almost unchangeable up to critical values of each of the parameters ν, $\delta A(t)$, $\delta B(t)$, and $\delta\omega(t)$. Upon reaching of any of the thresholds the effect of directed motion abruptly disappears.

The complete space of all new and old parameters has too many dimensions to be systematically accounted for by numerical simulations. To illustrate the results we limit ourselves by a particular combination of the parameters $\omega/\omega_0(m) = 1$, $\eta/[m\omega_0(m)] = 0.89$, $k_B T/U_0 = 0.02$, $\Delta B/l = 1.3$, $\Delta A/A_0 = 0.85$, and $\lambda/A_0 = 0.5$, which corresponds to the best driving ability of the channel at all zero corrections ν, $\delta A(t)$, $\delta B(t)$, and $\delta\omega(t) \to 0$. Keeping all other parameters fixed, we vary each fluctuation parameter independently and show the results in Figs. 2–5.

The conceptual picture of directed transport in a symmetric channel with different fractions of randomness is presented in Figs. 1(a)–1(c). Subplot (a) shows a two-dimensional snapshot of a purely periodic channel $\nu = 0$ for comparison. The instantaneous positions of the embedded particles of masses $m = 1$ after ten periods of oscillation are depicted by gray circles. The same snapshot of the system with $\nu = 0.25$ is shown in subplot (b). Subplot (c) presents a strongly randomized channel with $\nu = 0.5$, where the periodic portion becomes almost invisible.

Figures 2(a) and 2(b) presents dependence of the ensemble- and time-averaged displacement, $\Delta x = \langle x \rangle - x|_{t=0}$ after 40 oscillations [subplot (a)] and standard deviation $S_x = \sqrt{\langle (x - \langle x \rangle)^2 \rangle}$ [subplot (b)] on fraction ν, which characterizes

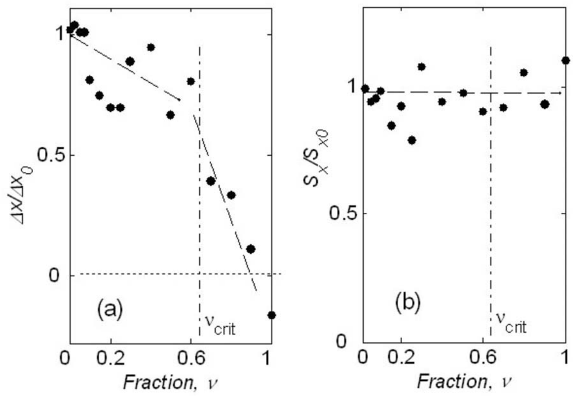

FIG. 2. Dependence of the ensemble- and time-averaged displacement $\Delta x = \langle x \rangle - x|_{t=0}$ after (a) 40 oscillations and (b) standard deviation $S_x = \sqrt{\langle (x - \langle x \rangle)^2 \rangle}$ on fraction ν, which characterizes net drift and dispersion of instantaneous particle positions, respectively. The results for Δx and S_x are normalized to their values Δx_0 and S_{x0} at $\nu = 0$. The critical fraction ν_{crit} is marked by the dashed-dotted line. The number of realizations is $N = 200$; other parameters as in Fig. 1.

net drift and dispersion of instantaneous particle positions, respectively. The results for Δx and S_x are normalized to their values Δx_0 and S_{x0} at $\nu = 0$. The critical fraction ν_{crit} at which the effect of directed motion decreases rapidly is marked by a dashed-dotted line. Our observations show that the channel behavior is much more resilient to structure variations than fluctuations of the oscillation parameters. It maintains its ability to drive particles even with variations in its structure of 50–60 %. However, even relatively small fluctuations (about 15–20 %) of $\delta A(t)$, $\delta B(t)$, and/or $\delta \omega(t)$ lead to fast and abrupt disappearance of the effect.

Corresponding results are presented in Figs. 3–5. All the figures show the ensemble- and time-averaged displacement, $\Delta x = \langle x \rangle - x|_{t=0}$ after 40 oscillations [subplot (a)] and standard deviation $S_x = \sqrt{\langle (x - \langle x \rangle)^2 \rangle}$ [subplot (b)] at all of the same parameters as Figs. 1 and 2.

Formally, a negative $\Delta x = \langle x \rangle - x|_{t=0}$ after crossing the dashed-dotted line depicting the critical value of each parameter in Figs. 2–5 represents that point of statistical realization losing systematic direction of motion. It can return after some number of oscillations and move later alternatively in negative or positive directions. However, even in such a scenario, the point maintains its compactness. This manifests itself in a finite (and almost equal to the regular case) value of the standard deviation $S_x = \sqrt{\langle (x - \langle x \rangle)^2 \rangle}$ in subplots (b) in each figure.

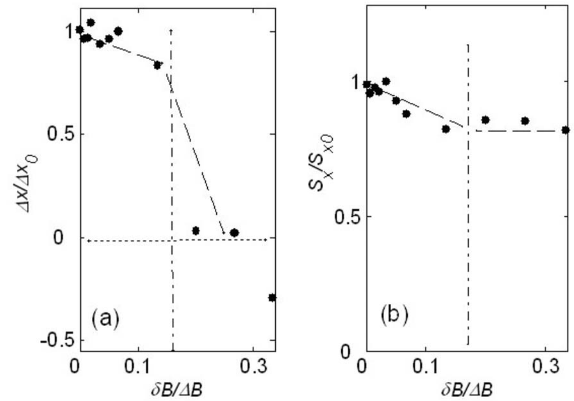

FIG. 4. Dependence of (a) the averaged displacement Δx and (b) standard deviation S_x on relative amplitude of transversal fluctuations $\delta B / \Delta B$. Parameters as in Fig. 2.

IV. CONCLUSION

We analyzed the stability of the directed transport and molecular separation mechanism in a dynamically controlled channel under random perturbations of its properties. The driving mechanism in the original system is based on a combination of lateral and normal harmonic channel oscillations. At an appropriately tuned resonance relation between these oscillations, the system exhibits very efficient directed transport. The system has an advantage over traditional ratchet systems due to its ability to produce directed transport even in the absence of thermal noise.

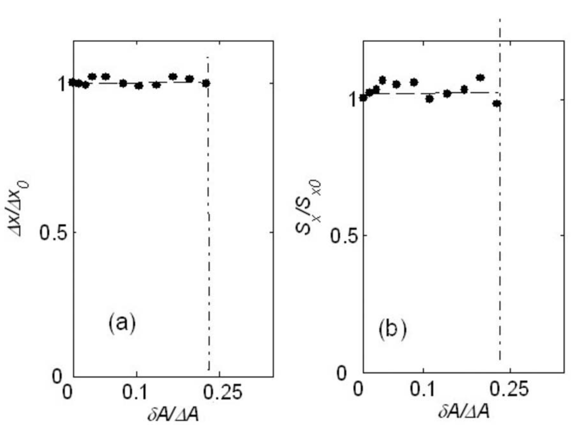

FIG. 3. Dependence of (a) the averaged displacement Δx and (b) standard deviation S_x on relative amplitude of lateral fluctuations $\delta A / \Delta A$. Parameters as in Fig. 2.

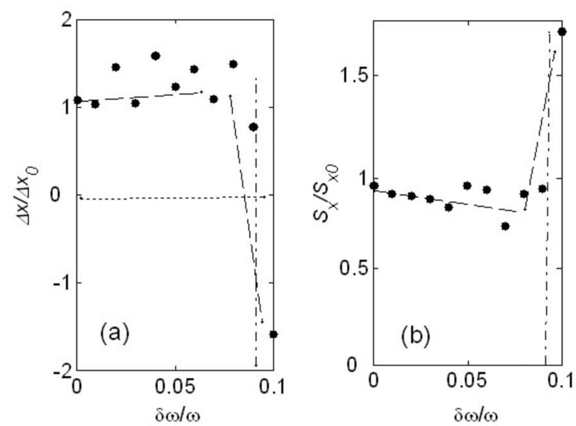

FIG. 5. Dependence of (a) the averaged displacement Δx and (b) standard deviation S_x on relative amplitude of frequency fluctuations $\delta \omega / \omega$. Parameters as in Fig. 2.

In the present paper the model is made more realistic. Nonideality of the channel and stochastic impacts to its oscillations are incorporated, and the stability of directed transport under random variations of all parameters is analyzed. It is proven that the phenomenon of directed transport holds under relatively strong fluctuations of all studied types. It is found also that the ability of the channel to transport particles has a threshold and remains almost unchangeable up to critical values of each of the fluctuating parameters. Upon reaching any of the thresholds the effect of directed motion abruptly disappears.

ACKNOWLEDGMENTS

We acknowledge M. Urbakh for fruitful discussion. A.E.F thanks DFG (PO 810/10-1) and ESF (reference no. 1661) scientific programs for financial support.

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