Diffusion enhancement in on/off ratchets

AIP | Applied Physics Letters

Diffusion enhancement in on/off ratchets

W. Chr. Germs, E. M. Roeling, L. J. van IJzendoorn, R. A. J. Janssen, and M. Kemerink

Citation: Appl. Phys. Lett. **102**, 073104 (2013); doi: 10.1063/1.4793198
View online: http://dx.doi.org/10.1063/1.4793198
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v102/i7
Published by the AIP Publishing LLC.

Diffusion enhancement in on/off ratchets

W. Chr. Germs, E. M. Roeling, L. J. van IJzendoorn, R. A. J. Janssen, and M. Kemerink[a)]
Applied Physics, Eindhoven University of Technology, PO Box 513, 5600 MB Eindhoven, The Netherlands

(Received 11 October 2012; accepted 7 February 2013; published online 20 February 2013)

We show a diffusion enhancement of suspended polystyrene particles in an electrical on/off ratchet. The enhancement can be described by a simple master equation model. Furthermore, we find that the diffusion enhancement can be described by a general curve whose shape is only determined by the asymmetry of the ratchet repeat unit. The scaling of this curve can be explained from an analytical expression valid for small off-times. Finally, we demonstrate how the master equation model can be used to find the driving parameters for optimal particle separation. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4793198]

In a Brownian ratchet, the random thermal motion of particles is used to create directed transport. By subjecting the particles to an asymmetric periodic potential and driving the system out of equilibrium, e.g., by superposing an oscillating force, the random motion is rectified into directed motion. Many different types of Brownian ratchets have been realized.[1] Ratchets have been demonstrated for suspended particles using structured arrays,[2–4] optical fields,[5,6] magnetic fields,[7–9] and electric fields.[10–13] Here, we focus on the on/off ratchet for suspended particles, where the periodic potential itself is oscillating in time, see Fig. 1(a).

Ratchets have been proposed as possible separators of different types of particles.[11–14] When using a ratchet as a particle separator, there are two important factors. One is the difference in average velocity of different types of particles, which should be large. Second, the spread of an ensemble of equal particles should be small. As depicted in Fig. 1(b), a mix of two types of particles can then eventually be extracted from a microchannel as two separated particle distributions.

The difference in average velocity can be achieved when the particles have a large enough difference in size. However, the second criterion can be more problematic. A ratchet can enhance the spreading of particles compared to the diffusion derived from the Einstein relation. This can be seen in Fig. 1(a) where at time t_2, when the potential has been restored, the spreading has increased as compared to thermal diffusion at t_1. This increase can be expressed as an increased effective diffusion constant, D_{eff}, of the whole ensemble. For rocking ratchets, this has been calculated[15–21] and has experimentally been confirmed.[22,23] Calculations on the diffusion in discrete multi-site on/off ratchets have been performed before in relation to the Péclet number, a figure of merit for the relative importance of drift over diffusion currents.[24,25] These works showed very low Péclet numbers. An increased diffusion has also been calculated[26] and measured[27] for particles between oscillating potential barriers of a non-ratchet device. However, apart from an anomalous increased diffusion perpendicular to the transport direction,[8] the enhanced diffusion along the transport direction has experimentally not been shown for on/off ratchets.

Here, we demonstrate diffusion enhancement for water suspended polystyrene particles in an electrical on/off ratchet.[28] We develop a master equation model with which the average movement and effective diffusion constant can easily be calculated numerically for on/off ratchets of arbitrary shape. In contrast with previous works,[24,25] each ratchet unit is represented by only one discrete site, resulting in a very simple model in which the transition rates are no longer explicitly dependent on the magnitude of the forces in the ratchet unit. We compare the results with measurements of the effective diffusion of suspended polystyrene particles. The model and the experiment show good agreement, and indicate relatively high Péclet numbers.

The water suspended polystyrene particles are measured in a microchannel above asymmetrically placed interdigitated finger electrodes. The ratchet has a spatial period of

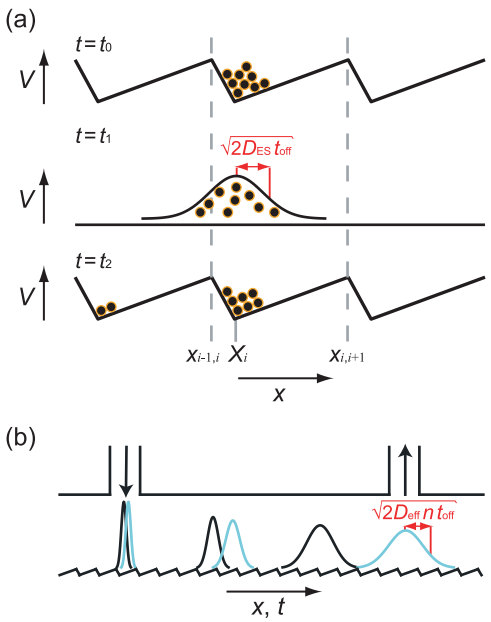

FIG. 1. (a) Particles in an on/off ratchet potential for three moments in time, showing the principle of an on/off ratchet. (b) The ratchet as a particle separator. Two types of particles are injected as a mix and extracted as two separated distributions. The effective diffusion coefficient, D_{eff}, is defined at the distribution on the utmost right, with n the number of on/off cycles and t_{off} the time per cycle for free diffusion.

[a)]Author to whom correspondence should be addressed. Electronic mail: m.kemerink@tue.nl. Tel.: +31 40 247 8321. Fax: +31 40 245 1036.

$L = 7\ \mu$m; geometrically, the short side of the ratchet unit equals $x_{\text{short}} = 2\ \mu$m. Particle displacement is recorded with an optical microscope and a high speed camera. A full description of the on/off ratchet and measurement setup is given in Ref. 28, which focuses on the interaction forces. We reanalyze the experimental data which was used to determine the average displacement of 300 nm and 500 nm particles vs. t_{off} in our previous work. Here, we calculate the effective diffusion coefficient, D_{eff}, using

$$D_{\text{eff}} = \frac{\sigma_{\text{eff}}^2}{2n_{\text{cycl}}t_{\text{off}}}, \text{with } {\sigma_{\text{eff}}}^2 = \sum_i p_i(x_i - \bar{x})^2, \tag{1}$$

where n_{cycl} is the number of on/off cycles and $\bar{x}$ the average position of the particles. Fig. 2 shows D_{eff} normalized by the Einstein-Stokes diffusion constant as symbols.

To describe on/off ratchet measurements, we developed a rather simple master equation model. The driving of the particles in the experiment has been shown to be dielectrophoretic in nature;[28] however, the model does not require any assumptions on the nature of the driving. It only assumes well defined locations of the potential well boundaries and potential minima. The time dependence of the ratchet potential is modeled as a block function, which is consistent with the experimental realization. The on-time is assumed to be long enough to trap all particles to a distribution that can be approximated by a delta function.

We define a probability, p_i, of finding a particle in a trap i, which is equal to the number of particles in the trap, n_i, divided by the total number of particles present in the device, N

$$p_i \equiv \frac{n_i}{N}. \tag{2}$$

After switching the ratchet potential off, the particles diffuse from a delta-shaped distribution to a Gaussian distribution. The rate per on/off cycle, γ_{ij}, at which a particle displaces from trap i to j is calculated from the integral of the Gaussian distribution of particles as

$$\gamma_{ij} = \int_{x_{j-1,j}}^{x_{j,j+1}} \frac{1}{\sigma_{\text{ES}}\sqrt{2\pi}} \exp - \frac{(x - X_i)^2}{2\,\sigma_{\text{ES}}^2} dx, \tag{3}$$

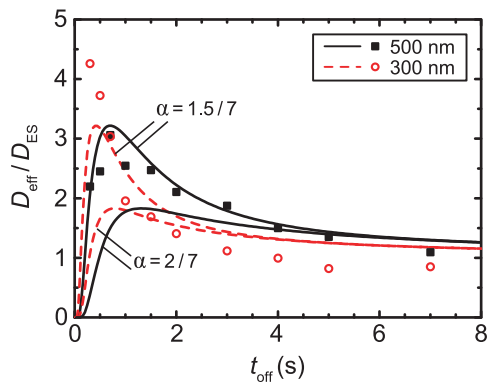

FIG. 2. Measurements (symbols) and calculations (lines) of the ratio of the effective diffusion coefficient and the Einstein-Stokes diffusion coefficient vs. the off-time per cycle for particle diameters of 300 nm and 500 nm.

with $x_{j,j+1}$ and X_i defined in Fig. 1(a). The width of the distribution is calculated as $\sigma_{\text{ES}} = \sqrt{2D_{\text{ES}}t_{\text{off}}}$, with t_{off} the time of the ratchet being in the off-state, and $D_{\text{ES}} = k_{\text{B}}T/6\pi\eta r$ the Einstein-Stokes diffusion coefficient with k_BT being the thermal energy, η the viscosity, and r the particle radius.

For the master equation model, the rate at which the trap occupation changes is written as

$$\frac{dp_i}{dt} = \sum_j (\gamma_{ji}p_j - \gamma_{ij}p_i). \tag{4}$$

The change per cycle in occupation for a whole array of traps can be calculated as

$$\begin{pmatrix} \Gamma_1 & \gamma_{21} & \gamma_{31} & \cdots \\ \gamma_{12} & \Gamma_2 & \gamma_{21} & \\ & & \ddots & \\ \gamma_{13} & \gamma_{12} & & \\ \vdots & & & \Gamma_{\text{final}} \end{pmatrix} \cdot \begin{pmatrix} p_1 \\ p_2 \\ \vdots \\ p_{\text{final}} \end{pmatrix} = \begin{pmatrix} \delta p_1 \\ \delta p_2 \\ \vdots \\ \delta p_{\text{final}} \end{pmatrix}, \tag{5}$$

with $\Gamma_i = -\Sigma_j \gamma_{i,j}$, from the right hand side of the sum in Eq. (4). Note that, apart from the middle diagonal, the matrix consists of diagonals of the same value, since $\gamma_{12} = \gamma_{23} = \gamma_{i\ i+1}$. By taking $p_{\text{final}/2}$ equal to one and all other traps empty, the average displacement per cycle and the effective diffusion coefficient of the entire particle distribution can be calculated by evaluating Eq. (5) only once. A few hundred subsequent evaluations for an array of a few hundred traps can easily be performed as well. Comparable to "normal" free diffusion, the calculations show that the width of the distribution σ_{eff} changes with $\sqrt{n_{\text{cycl}}}$, and D_{eff} is independent of n_{cycl}, i.e., independent of time.

Fig. 2 shows the results of the master equation calculations as lines for two different ratchet asymmetries, $\alpha = x_{\text{short}}/L = 1.5/7$ and $\alpha = 2/7$, with $T = 298$ K and $\eta = 8.9 \times 10^{-4}$ Pa s. For the 500 nm particles and an asymmetry of $\alpha \approx 1.5/7$, a good agreement between experiments and calculations is obtained. Both clearly show a peak in D_{eff} for intermediate t_{off}. The agreement for the 300 nm particles is of somewhat lower quality. The low D_{eff} at low t_{off} is due to the very limited diffusive spread ($\ll x_{\text{short}}$) of the particles for low t_{off}. This reduction of D_{eff} becomes smaller with increasing t_{off}, where at intermediate t_{off} we observe a D_{eff} that is larger than the Einstein-Stokes diffusion. This diffusion enhancement is explained by the process displayed in Fig. 1(a), where the ratchet potential displaces particles during the retrapping process, increasing the width of the total distribution. Finally, when t_{off} becomes very large, $D_{\text{eff}}/D_{\text{ES}}$ approaches 1, since the average displacement by the retrapping becomes small compared to the width σ_{ES}. The shape of the diffusion enhancement curve is very similar to the diffusion enhancement vs. tilting force in rocking ratchets,[15,17] where the enhancement can be explained by a similar reasoning.

Finally, we remark that the effective asymmetry $\alpha \approx 1.5/7$ found here on basis of the effective diffusion constant is rather close to $\alpha \approx 2/7$ found in Ref. 28 by analyzing the average displacement of the beads.

The number of particles that moves from trap i to trap j is described in Eq. (3) to depend on the width of the diffused particle distribution originating from trap i. Therefore, the

diffusion enhancement curve can be generalized to a curve that depends on this width, $\sqrt{2D_{ES}t_{off}}$. In fact, for each asymmetry α, a master curve can be found that describes the effective diffusion enhancement. These curves are shown as a function of the width relative to the trap length L and the asymmetry α for five different asymmetries in Fig. 3(a). The shape of the curve only depends on α, all other parameters are contained in the scaled axis variables which include the temperature, viscosity, and particle radius. The scaling can be explained by describing the diffusion with an expression for the fraction of particles that is displaced to the spatially nearest trap, for $t_{off} \ll (1-\alpha)^2 L^2/2 D_{ES}$,[11,29] as

$$s = \frac{1}{2}\mathrm{erfc}\left(\alpha L/\sqrt{4 D_{ES} t_{off}}\right) \tag{6}$$

from which the width of the distribution can be approximated as[11]

$$\sigma^2 = n_{cycl} L^2 s (1-s), \tag{7}$$

where erfc is the complementary error function. Using Eq. (1), the effective diffusion constant becomes

$$D_{eff} = \frac{L^2}{4t_{off}}\mathrm{erfc}\left(\frac{\alpha L}{\sqrt{4 D_{ES} t_{off}}}\right) \times \left[1 - \frac{1}{2}\mathrm{erfc}\left(\frac{\alpha L}{\sqrt{4 D_{ES} t_{off}}}\right)\right], \tag{8}$$

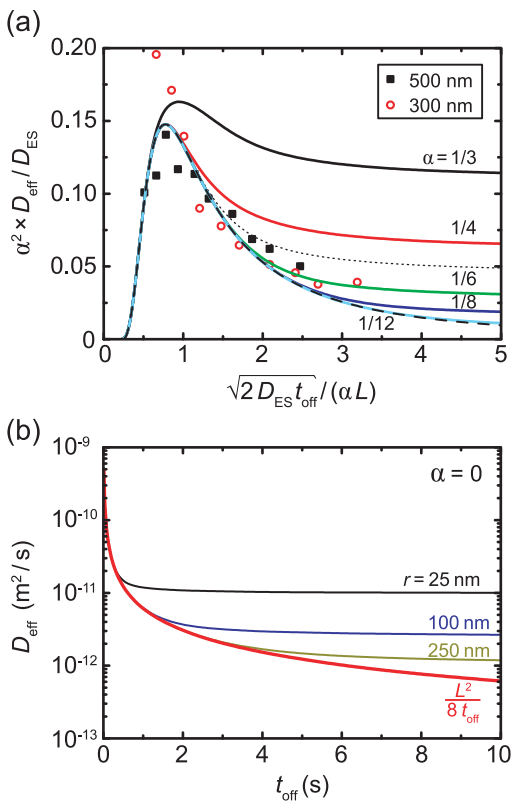

FIG. 3. (a) Calculations (solid lines) of the effective diffusion master curves for different asymmetries α. All other parameters are contained in the x,y-variables. The dashed line is calculated from Eq. (9). The measurements (symbols) are the same results as displayed in Fig. 2, the dotted line is calculated with $\alpha = 1.5/7$. (b) The analytical expression Eq. (10) (thick line) and the numerical calculations of the effective diffusion constant for three particle sizes (thin lines), all with $L = 7\,\mu$m and $\alpha = 0$.

which can be rewritten as

$$\begin{aligned}\frac{\alpha^2}{D_{ES}} D_{eff} &= \frac{\alpha^2 L^2}{4 D_{ES} t_{off}}\mathrm{erfc}\left(\frac{\alpha L}{\sqrt{4 D_{ES} t_{off}}}\right) \\ &\quad \times \left[1 - \frac{1}{2}\mathrm{erfc}\left(\frac{\alpha L}{\sqrt{4 D_{ES} t_{off}}}\right)\right] \\ &= \frac{1}{2\chi^2}\mathrm{erfc}\left(\frac{1}{\chi\sqrt{2}}\right) \times \left[1 - \frac{1}{2}\mathrm{erfc}\left(\frac{1}{\chi\sqrt{2}}\right)\right], \quad \text{with} \\ \chi &= \frac{\sqrt{2 D_{ES} t_{off}}}{\alpha L},\end{aligned} \tag{9}$$

explaining the choice of the x- and y-axis scaling. Equation (9) is displayed in Fig. 3(a) as the dashed line and is almost overlapping the low α curve. The χ value where Eq. (9) breaks down scales roughly with α^{-1}, leading to comparable t_{off} values for the breakdown. Even though the expression is valid for $\alpha \to 0$, the choice of the x-axis makes it impossible to actually display the curve for $\alpha = 0$. For $\alpha = 0$, Eq. (8) reduces to the simple size independent expression

$$D_{eff,\alpha=0} = \frac{L^2}{8\, t_{off}}, \tag{10}$$

which predicts the numerical results within 10% accuracy as long as $\sqrt{2D_{ES}t_{off}} < 0.4L$. Equation (10) is displayed in Fig. 3(b) together with numerical calculations for three different particle sizes. For large t_{off}, all asymmetries have D_{eff}/D_{ES} approaching 1; therefore, the curves in Fig. 3(a) approach α^2. The peak in the diffusion enhancement remains at the same coordinates for low α. We determined that for $\alpha \leq 1/4$ the diffusion peak sits at $\chi = 0.78 \pm 0.01$ with a height of $\alpha^2 D_{eff}/D_{ES} = 0.148 \pm 0.001$, which corresponds to the value found with Eq. (9). We, therefore, conclude that for intermediate and strong asymmetries, $\alpha \leq 1/4$, the effective diffusion peak occurs at

$$t_{off,peak} = \frac{(0.78\,\alpha L)^2}{2 D_{ES}} = \frac{3\,\pi\,\eta\, r (0.78\,\alpha L)^2}{k_B T}, \tag{11}$$

with

$$D_{eff,peak} = \frac{0.148\, D_{ES}}{\alpha^2} = \frac{0.148\, k_B T}{\alpha^2 6\,\pi\,\eta\, r}. \tag{12}$$

In view of effective particle separation, the diffusion enhancement is bad news. The Péclet number is often used as a figure of merit for ratchets and is defined as $Pe \equiv \langle v \rangle l / D_{eff}$, with $\langle v \rangle$ the average velocity and l a characteristic length. Pe can be calculated from Eqs. (6) and (8). By taking l equal to L, one finds

$$Pe = 4 \times \left(1 + \mathrm{erf}\left(\alpha L/\sqrt{4 D_{ES} t_{off}}\right)\right)^{-1}, \tag{13}$$

which is plotted in Fig. 4 together with numerical Pe curves for different α. In contrast to earlier work,[24,25] good Pe numbers are found, i.e., $Pe > 2$ for which drift wins over diffusion. Similar numbers have been calculated for tilted periodic potentials.[30] Figure 4 suggests that for the systems

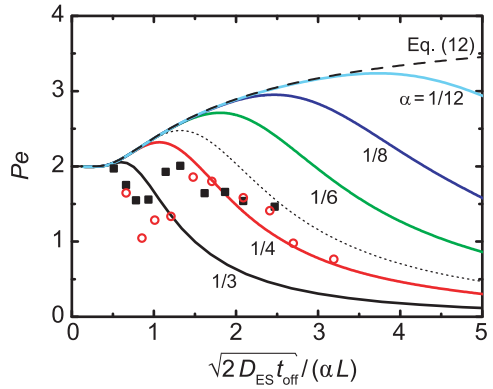

FIG. 4. Calculated Péclet numbers for different asymmetry α (solid lines) and analytically derived Péclet numbers (Eq. (12), dashed line). All device parameters are contained in the *x*-axis. The experimental points (symbols) are calculated from the diffusion data in Fig. 2 and the displacements data from Fig. 3 of Ref. 28.

investigated here Pe is always below 4. The limit $Pe = 2$ for $t_{off} \to 0$ reflects the fact that both drift and diffusion go to zero and are no longer affected by the ratchet. Unfortunately, the noise on the experiments is such that the expected value of Pe (slightly) above 2 for intermediate off-times cannot be confirmed.

In determining the optimal driving parameters for particle separation, the Péclet number is actually of limited use, since it gives information on one of the two particles only and gives no information on the desired large difference in average velocity between the particles. These parameters can be found using the same master equation model, which we will apply to calculate the optimal t_{off} for the system used here. The calculations are fast and a range of t_{off} can be calculated to find a maximum purity. Fig. 5 shows a calculation for the two particle sizes used in Fig. 2 with $\alpha = 1.5/7$ and $t_{off} = 0.32$ s. The solid lines are the average positions of the particle distributions after n_{cycl} cycles. The short dashed lines indicate the $\pm\sigma$ points. If enough cycles are allowed, any two particle sizes can, in principle, be separated, since the difference in position is linear in n_{cycl} while $\sigma \propto \sqrt{n_{cycl}}$.

The long dashed line shows the purity. The coordinate of separation is taken such that on each side of this coordinate both distributions have the same purity $n_1/(n_1 + n_2)$. The inset of Fig. 5 displays the purity after 5 min as a function of t_{off}. The highest purity was found for $t_{off} = 0.32$ s. For a realistic trapping time of 2 s, a 5 min interval is equal to 130 cycles, at which a purity of 0.87 can theoretically be obtained. Evidently, for larger (smaller) size differences, the same separation can be performed faster (slower). For example, separating 100 nm and 700 nm particles for 5 min with $t_{off} = 0.25$ s, a purity better than 0.9999 is calculated.

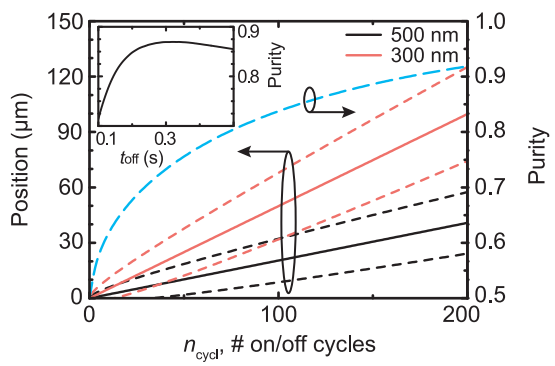

FIG. 5. Position of 300 nm and 500 nm (diameter) particle distributions as a function of number of on/off cycles (left axis). The solid lines are the average positions and the short dashed lines are the $\pm\sigma$ position. The long dashed line (right axis) shows the purity if the distributions are separated. Here, $t_{off} = 0.32$ s, determined to be the optimal value after 5 min, as shown in the inset.

In conclusion, we demonstrated a diffusion enhancement in an electrical on/off ratchet for polystyrene particles. The experiments show good agreement with a simple master equation model. Furthermore, we showed that the diffusion enhancement can be described via a general curve whose shape only depends on the asymmetry. An analytical approximation was derived and found to be in good agreement with the numerical model; its limitations are discussed. Finally, we showed that the master equation model can be used to find the optimal driving setting for optimal particle separation.

This work was funded by the Dutch Organization for Scientific Research NWO (Grant VIDI7575). We are grateful to B. Smalbrugge, T. de Vries, and E. J. Geluk for technical assistance.

[1]P. Hänggi and F. Marchesoni, Rev. Mod. Phys. **81**, 387 (2009).
[2]A. Oudenaarden and S. G. Boxer, Science **285**, 1046 (1999).
[3]G. Mahmud, C. J. Campbell, K. J. M. Bishop, Y. A. Komarova, O. Chaga, S. Soh, S. Huda, K. Kandere-Grzybowska, and B. A. Grzybowski, Nat. Phys. **5**, 606 (2009).
[4]S. E. Hulme, W. R. DiLuzio, S. S. Shevkoplyas, L. Turner, M. Mayer, H. C. Berg, and G. M. Whitesides, Lab Chip **8**, 1888 (2008).
[5]B. J. Lopez, N. J. Kuwada, E. M. Craig, B. R. Long, and H. Linke, Phys. Rev. Lett. **101**, 220601 (2008).
[6]L. P. Faucheux, L. S. Bourdieu, P. D. Kaplan, and A. J. Libchaber, Phys. Rev. Lett. **74**, 1504 (1995).
[7]A. Auge, A. Weddemann, F. Wittbracht, and A. Hutten, Appl. Phys. Lett. **94**, 183507 (2009).
[8]P. Tierno, P. Reimann, T. H. Johansen, and F. Sagués, Phys. Rev. Lett. **105**, 230602 (2010).
[9]L. Gao, M. A. Tahir, L. N. Virgin, and B. B. Yellen, Lab Chip **11**, 4214 (2011).
[10]E. Altintas, K. F. Böhringer, and H. Fujita, Jpn. J. Appl. Phys., Part 1 **47**, 8673 (2008).
[11]J. S. Bader, R. W. Hammond, S. A. Henck, M. W. Deem, G. A. McDermott, J. M. Bustillo, J. W. Simpson, G. T. Mulhern, and J. M. Rothberg, Proc. Natl. Acad. Sci. U.S.A. **96**, 13165 (1999).
[12]C. Marquet, A. Buguin, L. Talini, and P. Silberzan, Phys. Rev. Lett. **88**, 168301 (2002).
[13]L. Gorre-Talini, S. Jeanjean, and P. Silberzan, Phys. Rev. E **56**, 2025 (1997).
[14]J. Rousselet, L. Salome, A. Ajdari, and J. Prost, Nature **370**, 446 (1994).
[15]G. Constantini and F. Marchesoni, Europhys. Lett. **48**, 491 (1999).
[16]P. Reimann, C. Van den Broeck, H. Linke, P. Hänggi, J. M. Rubi, and A. Pérez-Madrid, Phys. Rev. Lett. **87**, 010602 (2001).
[17]P. Reimann, C. Van den Broeck, H. Linke, P. Hänggi, J. M. Rubi, and A. Pérez-Madrid, Phys. Rev. E **65**, 031104 (2002).
[18]E. Heinsalu, M. Patriarca, and F. Marchesoni, Phys. Rev. E **77**, 021129 (2008).
[19]P. S. Burada, P. Hänggi, F. Marchesoni, G. Schmid, and P. Talkner, ChemPhysChem **10**, 45 (2009).
[20]D. Dan and A. M. Jayannavar, Phys. Rev. E **66**, 041106 (2002).
[21]P. Reimann, Phys. Rep. **361**, 57 (2002).
[22]S.-H. Lee and D. G. Grier, Phys. Rev. Lett. **96**, 190601 (2006).
[23]M. Evstigneev, O. Zvyagolskaya, S. Bleil, R. Eichhorn, C. Bechinger, and P. Reimann, Phys. Rev. E **77**, 041107 (2008).

[24]J. A. Freund and L. Schimansky-Geier, Phys. Rev. E **60**, 1304 (1999).
[25]Y. Zhou and J.-D. Bao, Physica A **343**, 515 (2004).
[26]F. Müller, P. Romanczuk, and L. Schimansky-Geier, Stochastics Dyn. **11**, 461 (2011).
[27]S. Bleil, P. Reimann, and C. Bechinger, Phys. Rev. E **75**, 031117 (2007).
[28]W. Chr. Germs, E. M. Roeling, L. J. van IJzendoorn, B. Smalbrugge, T. de Vries, E. J. Geluk, R. A. J. Janssen, and M. Kemerink, Phys. Rev. E **86**, 041106 (2012).
[29]H. Linke, M. T. Downton, and M. J. Zuckermann, Chaos **15**, 026111 (2005)
[30]B. Lindner, M. Kostur, and L. Schimansky-Geier, Fluct. Noise Lett. **1**, R25 (2001).