Negative mobility and sorting of colloidal particles

Ralf Eichhorn,*[a] Jan Regtmeier,[b] Dario Anselmetti[b] and Peter Reimann[c]

DOI: 10.1039/b918716m

Transport in the realm of soft matter is strongly influenced by diffusion. Conditions far from thermal equilibrium and non-linear dynamics may give rise to unexpected transport phenomena, which are ruled out by the second law of thermodynamics under equilibrium conditions. Here, we highlight one of them, namely the migration of colloidal microparticles opposite to a static force and illustrate its application for particle sorting purposes.

1 Introduction

The dynamics of soft matter is dominated by energy scales that are comparable to thermal energy at room temperature.[1] Accordingly, thermal fluctuations play a key role, often giving rise to quite unexpected physical phenomena that may even be in striking contradiction to our everyday experience (which we gain by observing the macroscopic world, where thermal fluctuations are mostly irrelevant).

In this contribution we review such a counter-intuitive phenomenon, namely the steady movement of single soft matter "particles" – specifically colloidal beads suspended in an aqueous solution – in the *opposite* direction to an externally applied static force. More precisely, we consider such a particle exhibiting zero net velocity, perturb it by a static force F, and measure the resulting particle mobility

$$\mu := \left.\frac{\mathrm{d}v}{\mathrm{d}F}\right|_{F=0} \qquad (1)$$

where v represents the velocity of the particle's average motion in the long run as a response to the perturbation F. Movement opposite to F corresponds to *negative mobility* $\mu < 0$.† The distinct feature of (absolute)‡ negative mobility is therefore a negative slope of the characteristic v-F curve at $F = 0$ and $v = 0$, as exemplified with Fig. 1.

Before describing the experimental system and the physical mechanism behind the negative mobility effect from Fig. 1, we point out two necessary prerequisites, which emerge by confronting this phenomenon with the basic laws of physics: Newton's second law tells us that the velocity changes in proportion to the (total) applied force, and, in particular, in accordance with the direction of that force. Of course, this fundamental principle cannot be violated by the negative mobility phenomenon. It rather leads us to the conclusion that *non-linear* dynamics is indispensable for negative mobility to occur, so that the effect of the bias F does not simply add up to the other forces acting on the particle. Another important aspect arises from thermodynamics. Obviously, a colloidal particle with negative mobility performs work

[a]NORDITA, Roslagstullsbacken 23, 10691 Stockholm, Sweden. E-mail: eichhorn@nordita.org
[b]Experimental Biophysics & Applied Nanoscience, Bielefeld University, 33615 Bielefeld, Germany
[c]Fakultät für Physik, Universität Bielefeld, 33615, Bielefeld, Germany

† The term "negative mobility" is also used in the different context of electrophoresis, referring to a negative sign of the electrophoretic mobility $\mu_{\mathrm{EP}} := v/E$ with an electric field strength E. In these cases, however, the particle always follows the direction of the corresponding force.
‡ The term "absolute" is often added in order to distinguish from "differential negative mobility", see also Section 4.

Ralf Eichhorn

Ralf Eichhorn, born 1970, is an Assistant Professor at the Nordic Institute for Theoretical Physics (Nordita) in Stockholm (Sweden). He received his PhD in theoretical physics from the University of Augsburg (Germany) in 2000, where he also stayed as a Postdoc for two years. He then joined the group of Peter Reimann at Bielefeld University (Germany). His main research interests include transport processes in non-equilibrium systems with application to biophysical problems, noise-induced phenomena, microfluidics, and nonlinear dynamics and chaos.

Jan Regtmeier

Jan Regtmeier, born 1979, is a project leader in the Experimental Biophysics & Applied Nanoscience group at Bielefeld University. He received his PhD in 2007 about non-equilibrium migration mechanisms for microfluidic bioanalysis. His research interests include the development of new transport mechanisms, their application towards bioanalytical problems, bioimaging and single cell protein analysis in micro and nanofluidic systems.

against the force F, which could in principle be harvested, *e.g.*, by cyclic operation of F. According to the second law of thermodynamics such an extraction of useful work from equilibrium systems is impossible, as it would realize a *perpetuum mobile* of the second kind. Consequently, a second central condition for negative mobility is that the unperturbed system must be in a *non-equilibrium* state.

2 Negative mobility of colloidal beads

The main result that we present here is the occurrence of negative mobility for single colloidal spheres in an aqueous buffer solution when they are driven through a structured microfluidic channel.[3] The central part of the microchannel is displayed in Fig. 1. The micro-structuring, consisting of periodically arranged rectangular posts, induces non-linear effects in the particle dynamics.§ Non-equilibrium conditions are established by an unbiased, periodically switching ac-voltage applied to the channel. The static perturbation is realized by a static dc-voltage, U, which is superimposed on the non-equilibrium ac-driving. In Fig. 1, the measured response curve, *i.e.*, the average particle velocity v as a function of U, nicely demonstrates the negative mobility behavior of the colloidal beads. It is in very good agreement with the simulation results of the bead dynamics in the microchannel, which is modeled by an overdamped limit of Newton's equations of motion (Langevin equations), including the most prominent electrokinetic effects present in our microfluidic setup (electrophoresis and electroosmosis[2]) as well as thermal noise effects.[3]

For an intuitive explanation of the physical mechanism behind the observed negative mobility,[3] we note that the gaps between two neighboring posts are alternately larger and smaller in size than the particle's diameter. The small gaps can be considered as particle "traps" – the electrical field lines created by the applied voltage can pass through them, but the beads cannot. The rectangular ac-driving leads to a back-and-forth motion in the x direction between two such traps (passing through a large gap midway between the small gaps). Given the alternating arrangement of the gap sizes within the rows of posts (see Fig. 1), the particle may be able to avoid the trap and instead pass through an adjacent large gap, whenever it thermally diffuses sufficiently far into the lateral y direction during its motion in the x direction towards the trap. Due to the symmetry of the ac-driving and the microstructure, these processes do not lead to any net movement. However, when this "unperturbed" system, exhibiting zero net motion, is perturbed by the static dc-voltage U, the back-and-forth motion becomes "biased": the bead moves faster in the direction of the force F generated by U, and slower in the opposite direction. During the slow motion (opposite to F) the bead has more time to diffuse than during the fast motion (in the direction of F), so that it is more likely to avoid the trap by diffusing in the y direction during the slow motion, resulting in a net displacement in the direction opposite to F. In the case of purely deterministic dynamics, the "bias" due to the voltage U would not have any visible net effect: the symmetry of the periodic post array and thus of the deterministic force field created in that structure would dictate an unperturbed back-and-forth motion between the equilibrium positions in the two traps. It is by the additional (possibly small) diffusive motion due to thermal noise that a net movement of the colloidal bead is induced. For illustration, a typical experimentally observed particle trajectory showing the negative mobility behavior is included in Fig. 1. A more detailed quantitative analysis of this mechanism is given in ref. 5.

The design of the experimental microstructure in Fig. 1 has been inspired by previous theoretical studies of negative mobility in model systems for Brownian particles.[6] Indeed, most of the existing works on negative mobility for single Brownian particles are of a theoretical nature. They cover a wide variety of different systems, ranging from setups close in spirit to "realistic" microfluidic devices[6–12] to models that include some sort of memory[13] or internal state.[14,15] A

§ A similar structure to the one used here has been theoretically studied in ref. 4 in the different physical context of gel electrophoresis.

Dario Anselmetti

Dario Anselmetti, born 1963, is a Professor of Experimental Biophysics & Applied Nanoscience at Bielefeld University since 2000. After receiving his PhD in 1990 at Basel University, he became a Postdoc at the IBM Research Laboratory Zürich in Rüschlikon and Basel University. From 1994 to 2000 he was research staff member and project leader at Ciba-Geigy, Novartis and Solvias AG. He is the executive chairman of the collaborative research center SFB 613 at Bielefeld University and member of the North Rhine-Westphalian academy of science. His main research interests are single molecule biophysics, AFM imaging and spectroscopy, systems nanobiology and micro- and nanofluidics.

Peter Reimann

Peter Reimann, born 1964, is a Professor of Theoretical Physics at Bielefeld University since 2002. After receiving his PhD 1992 from the University of Basel (Switzerland), he was a Postdoc at the University of Hasselt (Belgium), the Eötvös Lorand University in Budapest (Hungary), and the University of Augsburg (Germany). His main research interests are complex systems far from equilibrium, noise- and transport-phenomena, nanotribology, and the basic questions of statistical physics.

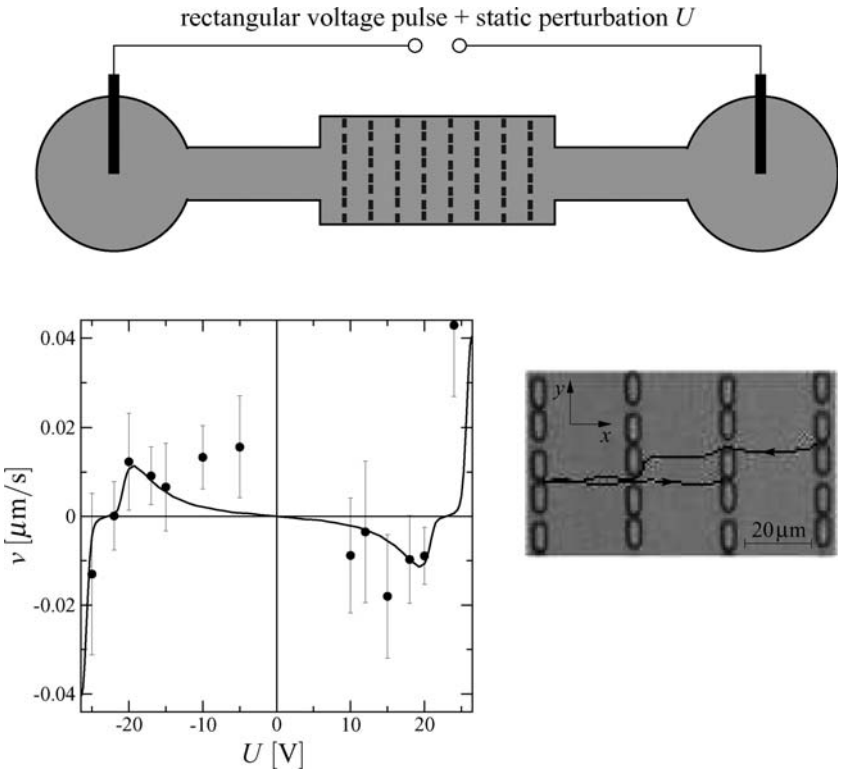

Fig. 1 (Adapted from ref. 3) **Upper part:** a schematic top view of the experimental setup. The two reservoirs (2 mm in diameter) are connected *via* inlet and outlet channels (each of length 2.5 mm, width 24 μm and height 9 μm) with the central microstructured part (length 6 mm, width 400 μm and height 9 μm), which contains a periodic array of rectangular posts (row distance 22.5 ± 0.1 μm). Electrodes are inserted into the reservoirs to apply electrical fields along the horizontal x direction (channel axis). Non-equilibrium conditions are generated by a rectangular ac-voltage. A static perturbation U is superimposed on the ac-driving, resulting in particle motion in a preferential direction. The sign convention is chosen such that a positive voltage U corresponds to a positive force F on the particles. Fabrication and operational details of the microfluidic chip are given in ref. 3. **Lower right part:** a partial view of the microstructure showing the periodic array of rectangular posts; in both the x and y directions, the gaps between adjacent posts are alternately smaller (1.7 ± 0.1 μm) and larger (3.1 ± 0.1 μm) than the size of the colloidal beads (between 1.9 and 2.9 μm in the experiments presented here). The black line with arrows represents a typical, experimentally observed particle trajectory for positive U over one period of the ac-drive (switching between ± 30 V every 25 s). The particle starts out in the rightmost trap and first experiences a negative value of the ac-drive, resulting in a slow motion in the negative x direction. Diffusion is relatively weak so that the particle escapes the "attraction basin" of the next small gap at the very last moment. Subsequently, it is quickly driven through the large gap and caught by the next small gap one row of posts further to the left (leftmost row in the picture), mainly by deterministic forces. The final fast motion in the positive x direction during the second half-period ends in the trap straight ahead. The overall result is a net displacement by one row of posts in the direction opposite to the static perturbation U. Note that the limited resolution in recording the particle trajectory smooths out details of the (diffusive) motion considerably. **Lower left part:** response of 2 μm beads to the static voltage U. The dots show experimentally measured velocities in the x direction, averaged over 40 beads and 200 s. Error bars are mainly of statistical origin, but variabilities in the beads and microstructure also contribute. The line shows the theoretical response characteristics obtained from numerical simulations.

particularly interesting setup, which could be readily realized, *e.g.*, by topographical microstructures or by light forces using optical tweezers, consists of a Brownian (colloidal) particle which moves in a "zigzag"-like, "meandering" structure.[8–12] Negative mobility effects are predicted for three prototypical sources of non-equilibrium:[12] time-dependent forcing (like in the example above),[8,9] time-dependent temperature (*i.e.* the system is alternately in contact with two heat reservoirs at different temperatures),[10] and temperature anisotropies[11] (*i.e.* the system is in simultaneous contact with two anisotropic heat reservoirs at different temperatures[16]).

In all of these systems, particle movement is "hindered" by a mechanism, which is symmetric in the unperturbed case ($F = 0$) and – as an effect of the static perturbation F – becomes more efficient in the direction of F than in the direction opposite to F. This principle seems to be the key to negative mobility in the regime of strong damping (the regime which also models colloidal systems). In contrast, a completely different physical mechanism, which is based on the interplay between thermal noise and transient chaos, is identified to induce negative mobility phenomena in underdamped Brownian dynamics, as detailed in more recent studies.[17] Yet another class of models exhibiting negative mobility exploits the collective effects of interacting Brownian particles.[18]

3 Particle sorting

After the first demonstration of negative mobility in a colloidal system, it was soon realized that this response phenomenon is very sensitive to particle properties. This opens the possibility of steering different particle species in opposite directions under identical experimental conditions. The proof-of-principle of such a separation scheme has been demonstrated in ref. 19 using the same microfluidic setup as described in Fig. 1. Focusing on two different species of colloidal beads, it has been possible to identify a suitable amplitude and frequency of ac-driving, such that one particle species exhibits negative mobility, whereas the other species responds to the dc-perturbation F "as usual", moving in the direction of F. The experimental results together with theoretical prediction are shown in Fig. 2.[19]

Negative mobility therefore opens up new, fascinating perspectives for the separation of micrometre-sized colloidal particles or biological compounds of comparable size, like cells or cell organelles. Compared to other methods proposed for particle sorting in microfluidic obstacle arrays,[20–25] it offers the possibility to not only "deflect" different particle species along different transport angles in the array, but even to steer them in opposite directions. However, as can be seen in Fig. 2 (and also Fig. 1), the separation velocities achieved so far are far too small for any practical purposes. The reason for the slow net motion is found in

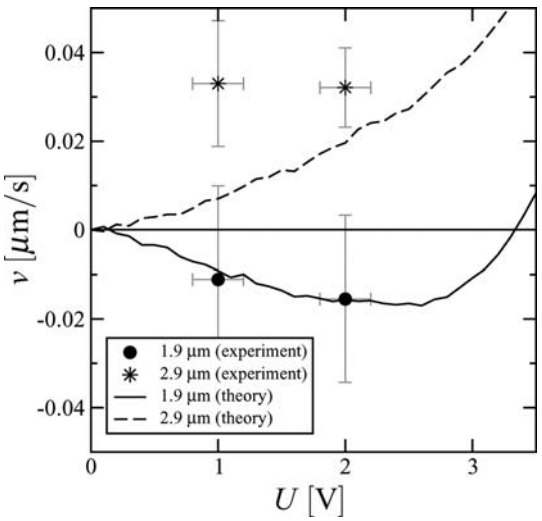

Fig. 2 (Adapted from ref. 19) Simultaneously observed average particle velocities for two different particle species: 1.9 μm beads (dots) move in the direction opposite to U (negative mobility), whereas 2.9 μm beads (stars) follow the direction of the static voltage U ("usual" response). The solid and dashed curves show results from numerical simulations for the two particle species. The experimental setup is identical to the one from Fig. 1, except that the ac-driving switches between $\pm$ 6 V every 70 s. The relatively large error bars and the differences between theory and experiment (particularly for the 2.9 μm particles) may be explained by bead-to-bead variations in size and surface charge and by deviations of the microstructure from strictly periodic conditions.[19]

the relatively slow diffusion of the colloidal beads (of the order of 0.1 $\mu m^2\ s^{-1}$), implying the need to use quite small ac-driving frequencies so that particle diffusion during one driving period is strong enough to induce the negative mobility effect (see above). As demonstrated in ref. 26, the bead velocity can be sped up by an order of magnitude by optimizing the post geometry. On the other hand, separation velocities will "naturally" benefit from the unavoidably increasing thermal noise effects, when replacing the micrometre-sized colloidal beads by smaller biomolecules like DNA or globular proteins. This, however, is a more challenging route to follow, as the trapping mechanism used for the micrometre-sized beads in Fig. 1 and 2 has to be substantially modified in order to work for (considerably) smaller particles.

4 Relation to other transport phenomena far from equilibrium

The negative mobility effect discussed here is often and more precisely named "absolute negative mobility" in order to make a distinction from the so-called "differential negative mobility". The latter term refers to $\mu < 0$ for $F \neq 0$ and therefore describes the decrease of the particle velocity upon increasing the applied force,[27] but not necessarily with v and F having opposite signs. It is, however, quite straightforward to generate absolute negative mobility in a symmetric system that exhibits differential negative mobility by applying an adiabatically slow non-equilibrium driving with an amplitude in that differential negative mobility regime.[12] Moreover, "extreme" cases of differential negative mobility have been reported,[8,10] where the particle motion in the direction of F slows down upon increasing F until it stops and indeed changes orientation to run opposite to F.

Whereas these negative mobility effects occur in *symmetric* systems, the so-called ratchet effects rely on a broken spatial and/or temporal symmetry.[28] Under non-equilibrium conditions, such types of *asymmetries* already induce a non-vanishing particle velocity in the absence of any systematic force ($F = 0$), as has been demonstrated in several experimental realizations for colloidal systems[29] and DNA fragments.[30] In other words, the defining feature of the ratchet effect is a characteristic v-F curve that passes through $F = 0$ at a finite value of v, usually with a positive slope ($\mu > 0$). A variant of the ratchet effect is often seen in many of the above mentioned separation methods with directional sorting,[20,23] as they make use of asymmetric obstacle arrays ("geometric ratchet").

5 Conclusions

The occurrence of negative mobility for colloidal beads in a microfluidic environment, as highlighted here, is the result of a subtle interplay between the following three indispensable prerequisites:[3,6] thermal noise (to induce diffusive motion of the colloidal particles), topographical structuring of the microchannel (to create a non-linear dynamics), and a pulsed external field (to drive the system away from equilibrium).

Concerning potential applications of this phenomenon, its viability for separation technologies still needs to be demonstrated. The first step beyond the proof-of-principle experiment in ref. 19 would be to realize negative mobility for a micrometre-sized biological compound. As already mentioned, for smaller biomolecules a suitable trapping mechanism or even a modified scheme for generating negative mobility needs to be developed. On the other hand, such a "separation induced by negative mobility" may be desirable as a further development of pulsed-field techniques in artificial micro- and nanostructures[25,31–33] in order to achieve most efficient sorting procedures with opposite transport directions for different particle species.

From a more fundamental perspective, the negative mobility effect discussed here may be viewed as part of the more general theme of specifically controlling particle motion by suitably tailoring the non-linearities and non-equilibrium conditions of the given system class. In transport problems along one dimension, on which we put our focus here, one can "adapt" the magnitude and sign of the mobility (eqn 1). More manifold possibilities occur when going to higher-dimensional situations like particle-transport on periodic surfaces or through two-dimensional obstacle arrays.[20–25] Then, the mobility μ becomes a tensor, and the particle velocity may reveal intriguing variations in direction upon changing the magnitude of the perturbation F or the amplitude and frequency of the ac-drive.[34]

Traditionally, noise is often considered as a nuisance that one has to fight and suppress as far as possible. Our present work represents one further step along an ongoing general change of paradigm, namely to utilize and channelize noise – which is practically unavoidable on the

micro- and nanometre scale anyway – for the purpose of analyzing and controlling nonlinear dynamics processes far from thermal equilibrium.

Acknowledgements

We thank Alexandra Ros and Thanh Tu Duong for fruitful collaboration, and the Deutsche Forschungsgemeinschaft (DFG) for financial support (Collaborative Research Center SFB 613).

References

1 M. Kleman and O. D. Lavrentovich, *Soft Matter Physics*, Springer, New York, 2002.
2 H. Bruus, *Theoretical Microfluidics*, Oxford University Press, Oxford, 2007.
3 A. Ros, R. Eichhorn, J. Regtmeier, T. T. Duong, P. Reimann and D. Anselmetti, *Nature*, 2005, **436**, 928; R. Eichhorn, A. Ros, J. Regtmeier, T. T. Duong, P. Reimann and D. Anselmetti, *Eur. Phys. J. Spec. Top.*, 2007, **143**, 159–164.
4 M. G. Gauthier and G. W. Slater, *Electrophoresis*, 2003, **24**, 441–451.
5 R. Eichhorn and P. Reimann, *Acta Phys. Pol. B*, 2006, **37**, 1491–1501.
6 R. Eichhorn, P. Reimann and P. Hänggi, *Phys. Rev. Lett.*, 2002, **88**, 190601; R. Eichhorn, P. Reimann and P. Hänggi, *Phys. Rev. E: Stat., Nonlinear, Soft Matter Phys.*, 2002, **66**, 066132.
7 B. Cleuren and C. Van den Broeck, *Phys. Rev. E: Stat., Nonlinear, Soft Matter Phys.*, 2003, **67**, 055101(R).
8 R. Eichhorn, P. Reimann and P. Hänggi, *Phys. A*, 2003, **325**, 101–109.
9 R. Eichhorn and P. Reimann, *Acta Phys. Pol. B*, 2004, **35**, 1407–1418.
10 R. Eichhorn and P. Reimann, *Phys. Rev. E: Stat., Nonlinear, Soft Matter Phys.*, 2004, **70**, 035106(R).
11 R. Eichhorn and P. Reimann, *Europhys. Lett.*, 2005, **69**, 517–523.
12 R. Eichhorn, P. Reimann, B. Cleuren and C. Van den Broeck, *Chaos*, 2005, **15**, 026113.
13 B. Cleuren and C. Van den Broeck, *Phys. Rev. E: Stat., Nonlinear, Soft Matter Phys.*, 2002, **65**, 030101(R).
14 B. Jiménez de Cisneros, P. Reimann and J. M. R. Parrondo, *Europhys. Lett.*, 2003, **64**, 599–605.
15 A. Haljas, R. Mankin, A. Sauga and E. Reiter, *Phys. Rev. E: Stat., Nonlinear, Soft Matter Phys.*, 2004, **70**, 041107.
16 R. Filliger and P. Reimann, *Phys. Rev. Lett.*, 2007, **99**, 230602.
17 L. Machura, M. Kostur, P. Talkner, J. Luczka and P. Hänggi, *Phys. Rev. Lett.*, 2007, **98**, 040601; D. Speer, R. Eichhorn and P. Reimann, *EPL*, 2007, **79**, 10005; D. Speer, R. Eichhorn and P. Reimann, *Phys. Rev. E: Stat., Nonlinear, Soft Matter Phys.*, 2007, **76**, 051110; M. Kostur, L. Machura, P. Talkner, P. Hänggi and J. Luczka, *Phys. Rev. B: Condens. Matter Mater. Phys.*, 2008, **77**, 104509; M. Kostur, L. Machura, J. Luczka, P. Talkner and P. Hänggi, *Acta Phys. Pol. B*, 2008, **39**, 1115–1124; J. Nagel, D. Speer, T. Gaber, A. Sterck, R. Eichhorn, P. Reimann, K. Ilin, M. Siegel, D. Koelle and R. Kleiner, *Phys. Rev. Lett.*, 2008, **100**, 217001.
18 P. Reimann, R. Kawai, C. Van den Broeck and P. Hänggi, *Europhys. Lett.*, 1999, **45**, 545; P. Reimann, C. Van den Broeck and R. Kawai, *Phys. Rev. E: Stat. Phys., Plasmas,Fluids, Relat. Interdiscip. Top.*, 1999, **60**, 6402; J. Buceta, J. M. Parrondo, C. Van den Broeck and F. J. de la Rubia, *Phys. Rev. E: Stat. Phys., Plasmas, Fluids, Relat. Interdiscip. Top.*, 2000, **61**, 6287; C. Van den Broeck, I. Bena, P. Reimann and J. Lehmann, *Ann. Phys.*, 2000, **9**, 713; S. E. Mangioni, R. R. Deza and H. S. Wio, *Phys. Rev. E: Stat., Nonlinear, Soft Matter Phys.*, 2001, **63**, 041115; B. Cleuren and C. Van den Broeck, *Europhys. Lett.*, 2001, **54**, 1; C. Van den Broeck, B. Cleuren, R. Kawai and M. Kambon, *Int. J. Mod. Phys. C*, 2002, **13**, 1195–1200.
19 J. Regtmeier, R. Eichhorn, T. T. Duong, P. Reimann, D. Anselmetti and A. Ros, *Eur. Phys. J. E*, 2007, **22**, 335–340.
20 D. Ertas, *Phys. Rev. Lett.*, 1998, **80**, 1548–1551; T. A. J. Duke and R. H. Austin, *Phys. Rev. Lett.*, 1998, **80**, 1552–1555; Z. Li and G. Drazer, *Phys. Rev. Lett.*, 2007, **98**, 050602.
21 A. M. Lacasta, K. Khoury, J. M. Sancho and K. Lindenberg, *Mod. Phys. Lett. B*, 2006, **20**, 1427–1442.
22 L. R. Huang, E. C. Cox, R. H. Austin and J. C. Sturm, *Science*, 2004, **304**, 987–990.
23 C.-F. Chou, O. Bakajin, S. W. P. Turner, T. A. J. Duke, S. S. Chan, E. C. Cox, H. G. Craighead and R. H. Austin, *Proc. Natl. Acad. Sci. U. S. A.*, 1999, **96**, 13762–13765; M. Cabodi, Y.-F. Chen, S. W. P. Turner, H. G. Craighead and R. H. Austin, *Electrophoresis*, 2002, **23**, 3496–3503; L. R. Huang, P. Silberzan, J. O. Tegenfeldt, E. C. Cox, J. C. Sturm, R. H. Austin and H. Craighead, *Phys. Rev. Lett.*, 2002, **89**, 178301; L. R. Huang, E. C. Cox, R. H. Austin and J. C. Sturm, *Anal. Chem.*, 2003, **75**, 6963–6967.
24 J. Fu, R. B. Schoch, A. L. Stevens, S. R. Tannenbaum and J. Han, *Nat. Nanotechnol.*, 2007, **2**, 121–128.
25 L. R. Huang, J. O. Tegenfeldt, J. J. Kraeft, J. C. Sturm, R. H. Austin and E. C. Cox, *Nat. Biotechnol.*, 2002, **20**, 1048–1051.
26 J. Regtmeier, S. Grauwin, R. Eichhorn, P. Reimann, D. Anselmetti and A. Ros, *J. Sep. Sci.*, 2007, **30**, 1461–1467.
27 S. R. White and M. Barma, *J. Phys. A: Math. Gen.*, 1984, **17**, 2995; G. A. Griess and P. Serwer, *Biopolymers*, 1990, **29**, 1863; V. Balakrishnan and C. Van den Broeck, *Phys. A*, 1995, **217**, 1; G. Cecchi and M. O. Magnasco, *Phys. Rev. Lett.*, 1996, **76**, 1968; G. W. Slater, H. L. Guo and G. I. Nixon, *Phys. Rev. Lett.*, 1997, **78**, 1170; R. K. P. Zia, E. L. Praestgaard and O. G. Mouritsen, *Am. J. Phys.*, 2002, **70**, 384–392.
28 R. D. Astumian, *Science*, 1997, **276**, 917; F. J. Jülicher, A. Ajdari and J. Prost, *Rev. Mod. Phys.*, 1997, **69**, 1269; P. Reimann, *Phys. Rep.*, 2002, **361**, 57; P. Hänggi and F. Marchesoni, *Rev. Mod. Phys.*, 2009, **81**, 387–442.
29 J. Rousselet, L. Salome, A. Ajdari and J. Prost, *Nature*, 1994, **370**, 446; L. P. Faucheux and A. Libchaber, *J. Chem. Soc., Faraday Trans.*, 1995, **91**, 3163; L. Gorre-Talini, J. P. Spatz and P. Silberzan, *Chaos*, 1998, **8**, 650; C. Marquet, A. Buguin, L. Talini and P. Silberzan, *Phys. Rev. Lett.*, 2002, **88**, 168301; S. Matthias and F. Müller, *Nature*, 2003, **424**, 53–57; K. Loutherback, J. Puchalla, R. H. Austin and J. C. Sturm, *Phys. Rev. Lett.*, 2009, **102**, 045301.
30 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.*, 1999, **96**, 13165–13169; R. W. Hammond, J. S. Bader, S. A. Henck, M. W. Deem, G. A. McDermott, J. M. Bustillo and J. M. Rothberg, *Electrophoresis*, 2000, **21**, 74–80.
31 T. A. J. Duke, R. H. Austin, E. C. Cox and S. S. Chan, *Electrophoresis*, 1996, **17**, 1075–1079.
32 O. Bakajin, T. A. J. Duke, J. Tegenfeldt, C.-F. Chou, S. S. Chan, R. H. Austin and E. C. Cox, *Anal. Chem.*, 2001, **73**, 6053–6056.
33 M. Cabodi, S. W. P. Turner and H. G. Craighead, *Anal. Chem.*, 2002, **74**, 5169–5174.
34 A. Gopinathan and D. G. Grier, *Phys. Rev. Lett.*, 2004, **92**, 130602; C. Reichhardt and C. J. Olson Reichhardt, *Phys. Rev. E: Stat., Nonlinear, Soft Matter Phys.*, 2004, **69**, 041405; M. Pelton, K. Ladavac and D. G. Grier, *Phys. Rev. E: Stat., Nonlinear, Soft Matter Phys.*, 2004, **70**, 031108; A. M. Lacasta, J. M. Sancho, A. H. Romero and K. Lindenberg, *Phys. Rev. Lett.*, 2005, **94**, 160601; J. P. Gleeson, J. M. Sancho, A. M. Lacasta and K. Lindenberg, *Phys. Rev. E: Stat., Nonlinear, Soft Matter Phys.*, 2006, **73**, 041102; D. Speer, R. Eichhorn and P. Reimann, *Phys. Rev. Lett.*, 2009, **102**, 124101.

