

Rectified Motion of Colloids in Asymmetrically Structured Channels

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(Received 3 October 2001; published 8 April 2002)

We set micron size particles into macroscopic motion by submitting them to a low frequency electric field (of zero mean value) in a microfabricated channel exhibiting a topological ratchetlike local polarity. Rectification is induced by the coupling between electrophoresis, electroosmosis, and dielectrophoresis. The macroscopic velocities of the particles are functions of the electric field and of the geometry of the channel; they strongly depend on their size which opens the way to potential separations.

DOI: 10.1103/PhysRevLett.88.168301

PACS numbers: 82.70.Dd, 05.40.-a, 07.10.Cm, 89.20.-a

A large number of theoretical studies have recently focused on the concept of “force-free” motion [1] for which an object can be set into macroscopic motion by coupling a local asymmetry in its environment with energy dissipation. No macroscopic force or gradient is then needed. This concept can be enlarged to situations in which a macroscopic force is applied to the object providing the time average of this force is zero. This covers the field commonly known as “rectification” or “tilting ratchets” [2,3].

In this Letter, we focus on water suspensions of colloidal particles in a channel whose lateral dimension is larger than but comparable to the diameter of the particles and that exhibits a modulation in width whose shape is periodic and polar along its axis. Figure 1 is a micrograph of such a channel. The particles are then submitted to an ac electric field along the channel whose mean value is zero.

The behavior of colloids in channels having this particular shape and submitted to an oscillating Poiseuille flow has been theoretically studied in [4] under the name of “drift ratchets.” These authors have shown by numerical simulations that not only does one expect to observe a rectification of the motion of these particles, but also that this rectification should be strongly size dependent.

The driving itself is quite different here as the electric field acts directly on the particles themselves on top of the global ac flow induced by electroosmosis. More precisely, and if we put electroosmosis aside, the electric field acts on the beads by two distinct effects. The first one which comes to mind is electrophoresis [5,6]: the particles are charged and an electric field E will drive them toward the corresponding electrode with a force f_{el} proportional to E .

However, because of the complex geometry of the channel, the electric field is not homogeneous: it is more intense in the constrictions than in the parts of wider section. This leads us to the second effect acting on the beads and which is dielectrophoresis (DEP): the contrast of polarizabilities between the beads and the surrounding medium drives the more polarizable of these two species towards the regions of high electric field [7] (here, the constrictions of the channel) with a force $f_{die} \propto a^3 \nabla E^2$ where a is the radius of the particle [8]. We work here at very low

frequencies for which the sign itself of f_{die} is notoriously unpredictable due to the frequency dependence of the dielectric constants [5]. However, from the above discussion, we know that f_{die} derives from a potential W reflecting the geometry of the channel (periodicity and local polarity); this potential itself is proportional to E^2 schematically represented on Fig. 1 (high electric fields at the necks and lower values in the wider regions). A negative dielectrophoretic behavior of the particle would correspond to a potential schematically described by this sketch, whereas a positive one would correspond to its mirror image. f_{el} drives the particle back and forth in this potential. In an energy plot, this appears by tilting the potential W with a slope f_{el} . However, the two effects are coupled: multiplying the electric field by a factor k also multiplies the potential by a factor k^2 . We thus expect the particles to be permanently trapped in the minima of W for high electric fields, whereas there might exist a more favorable low electric field regime for which they move back and forth in this potential.

To end this analysis on the influence of the electric field on the suspension, electroosmosis is also to be considered because of the intrinsic charge of the channel walls. Electroosmosis will induce a plug flow during one half-period [6] and the opposite flow during the second half-period. As we work at low Reynolds numbers (typically 10^{-2}), the flows are known to be fully reversible and hydrodynamics by itself will not generate any net motion even with

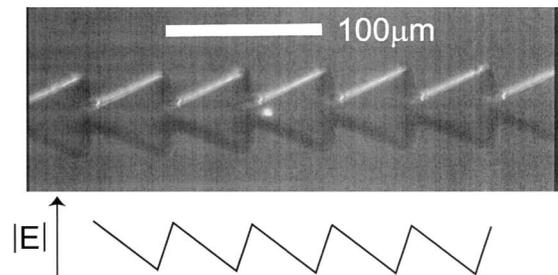


FIG. 1. Micrograph of a $4.5 \mu\text{m}$ latex bead in a PDMS channel. The blurred aspect of the bead is due to the time resolution of the camera.

a channel shape such as the one dealt with in this study. Still, this effect contributes to drive the particles back and forth in the dielectrophoresis-induced potential described above and, as pointed out in Ref. [4], other effects such as transverse diffusion of the particles in this laminar flow or shocks on the walls may cause rectification. Moreover, even at relatively modest Reynolds numbers, similarly to what is observed in smooth tubings for parabolic flows [9], the shape itself of the channel may also induce a distribution of the density of the beads due to a shear component of the flow.

Because of the much slower dependence of the electrophoretic effect with the size compared to DEP, we expect a nonlinear variation of the macroscopic velocity of the particles with their sizes. For the extreme cases, particles much smaller than the neck of the channel should not be sensitive enough to DEP to have their motion rectified. They should not be sensitive either to the electroosmosis-induced hydrodynamics effects just described. On the other hand, for diameters close to the size of the neck itself, the potential W becomes so steep that it is not overcome by the electrophoretic-electroosmotic driving, the macroscopic velocity should therefore also decrease. This non-monotonic behavior may be advantageous compared to the more usual laws for the velocity such as direct electrophoresis: For instance, getting intermediate fractions in a single step or being able to extract a few small particles among a lot of larger ones. It is worth noting that more conventional techniques such as electrophoresis or field flow fractionation, although extremely efficient for smaller or larger objects, show limitations precisely in the size range of a few μm .

Our second motivation in these experiments is to demonstrate their feasibility to transpose them in a second step to objects for which separation is more challenging and, in the first place, biological objects such as long DNAs, organelles or cells.

The microfabrication of such structures was performed using the strategy of micromolding [10]. Briefly, a negative imprint (a "mold") of the channel is microfabricated in a 15 μm thick negative photoresist (SU8-10, MCC) by conventional photolithography. A negative replica is then molded with a curable PDMS elastomer (Sylgard 184, Dow Corning) by pouring the mixture of polymer and curing agent on the mold and letting cure at 65 °C for at least 2 h. This replica is then plasma oxidized for 30 s in residual air and gently applied on a clean conventional glass slide in which two apertures have been previously drilled on both extremities of the channel. The plasma treatment promotes a strong adhesion between the glass and the PDMS, it also makes the channel walls hydrophilic and therefore easier to fill up with the colloidal suspensions. The channels were replaced every day. In most of the experiments described in this paper, the neck size was close to 12 μm . Channels with a neck size of 6 μm were also used. No particular further treatment of the surfaces to reduce elec-

troosmosis was performed. Although not strictly necessary, all the above steps were performed in a clean room (class 10 000) at controlled temperature and humidity.

After fabrication, the channels were filled up with a dilute suspension of carboxylate modified surfactant-free fluorescent latex beads of diameters ranging from 1 to 6 μm (Pierce and Molecular Probes) in ultrapure water (resistivity $\sim 18 \text{ M}\Omega \cdot \text{cm}$, Debye length $\sim 1 \mu\text{m}$). Differences of hydrostatic pressure were carefully balanced by adjusting the volumes in the two reservoirs until no residual flow could be observed. This absence of flow was periodically checked during the course of the experiments. A difference of potential generated by a signal synthesizer (Philips 5193) (either a sine or a square signal) was then applied between these two reservoirs via Pt electrodes after amplification by a factor 1000 (Trek Amplifier 609D). Typical electric fields were of the order of a few $100 \text{ V} \cdot \text{cm}^{-1}$. One of the electrodes was then grounded while the other was set at the high voltage. To eliminate any trace of a residual dc component, two precautions were taken: First, the mean value of the high voltage was carefully measured and adjusted to 0 by superimposing an offset with an additoner. Second, after each series of experiments, the two electrodes were exchanged (the grounded one was set to the high voltage and vice versa). The observations in the two configurations of the electrodes did not differ by more than 10% for frequencies lower than 15 Hz and the results we give here are the averages of these two measurements. The beads were observed by video microscopy, using fluorescence for the smallest beads, and velocities were measured by analyzing the motion of several single beads in each case.

Our first result is that the motion of the beads is indeed rectified. In all the cases studied here, the motion is directed toward the apex of the triangles, i.e., to the left on Fig. 1. Figure 2 represents the evolution of a 3 μm particle submitted to a sine driving (amplitude of motion ~ 2.5 periods, frequency 2 Hz). On top of the oscillatory "fast" behavior (Fig. 2, inset), there is a clear macroscopic drift corresponding to a $6 \mu\text{m} \cdot \text{s}^{-1}$ velocity in this particular example.

We have represented on Fig. 3, the evolution of the velocity of a 3 μm particle as a function of frequency for a sine signal. Square signals yielded most of the time to higher velocities. Here, the intensity of the applied voltage was adjusted in order to let the particle explore 100 μm (~ 2 patterns) for each period. As one can expect, we observe a quasilinear increase of the velocity with the frequency up to 15 to 20 Hz at which point the behavior of the beads becomes unstable: after a short while, they are ejected from the center of the channel toward its periphery and are then confined in the wider portions of the channel. We do not have yet a full explanation for this unstable behavior. It is not the signature of a negative DEP regime as experiments in dc field show trapping in the constrictions for high enough fields impairing left-to-right motion

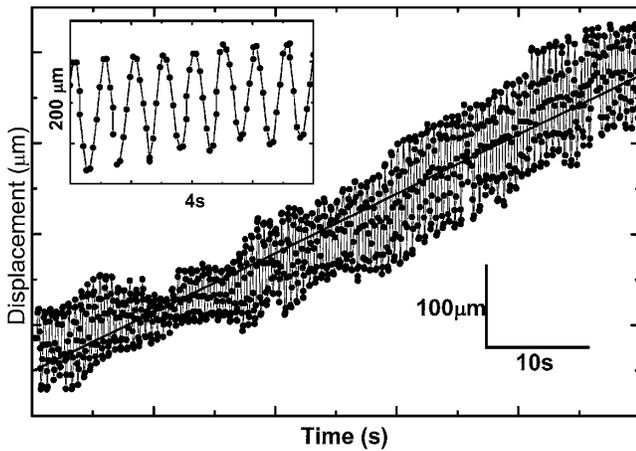


FIG. 2. Time evolution of a $3 \mu\text{m}$ bead driven at 2 Hz with a $100 \mu\text{m}$ amplitude. The inset details the “fast” oscillating regime reflecting the driving, whereas the macroscopic drift is clearly visible on the main curve. The solid line is the best fit giving an average velocity of $6 \mu\text{m} \cdot \text{s}^{-1}$.

(no such trapping was observed for right-to-left motion for similar intensities). A careful observation of the beads in ac field show that, above a threshold in velocity, DEP is counterbalanced by a force expelling the particles from the constrictions similarly to what can be observed around macro-ions aggregates [11]. The walls of the channel being in our case perfect insulators, we tend to believe that this hydrodynamic force is rather caused by electroosmosis. A direct evaluation of this hypothesis is, however, difficult precisely because of the polarity of the channel and also because of its heterogeneous surface chemistry with three walls made of oxidized PDMS and the fourth one made of glass.

At that point, let us make a side remark: DEP is usually performed with electrodes of complex shapes to create these high field regions [7], here we show that manipulation

of particles can be performed by a dc or a low frequency ac field along a structured channel avoiding therefore many of the usual technical difficulties (charge injection instabilities at electrodes, mirror effect tending to adsorb irreversibly charged particles on metal surfaces, etc.). The same concept has also been recently successfully applied to DNA in smaller structures [12].

The variation of the velocity with the electric field is plotted in Fig. 4. It reveals a nonmonotonic behavior with two zero-velocity regimes: the first one is intuitive and occurs for low electric fields (the particle is driven back and forth between two necks). The second one occurs for high electric fields when the local velocities becomes very high and expel the particles from the center toward the periphery of the channel similarly to what we observe at high frequencies. Between these regimes, the velocity increases linearly with the electric field. These two thresholds being size dependent, a curve such as Fig. 4 is quite appealing as it suggests that a careful tuning of the electric field intensity, in particular around the low electric field threshold, could set certain sizes into motion while others would stay permanently trapped.

As for the dependence of the velocity on the size of the particles, this behavior is represented in Fig. 5. This graph results from independent experiments (one series for each size) for which the voltage is adjusted to give a $100 \mu\text{m}$ oscillation amplitude for the motion of the particles. We found that it was the best way to compare beads of different sizes and therefore bearing different global charges. Although our measurements of the electrophoretic mobilities of the beads in straight channels showed the expected trend of a global increase of the charge with the size, they could not be theoretically described because of differences in the exact charge surface density of the beads. This analysis is a way to correct for this effect, it also enables a direct comparison with the simulations of Ref. [4]. Experiments were conducted between 2 and 15 Hz and all the

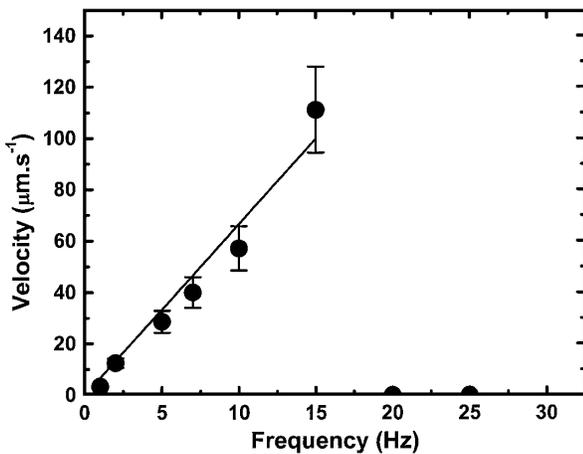


FIG. 3. Macroscopic velocity of a $3 \mu\text{m}$ particle as a function of frequency for a $100 \mu\text{m}$ fixed peak-to-peak amplitude of motion (sine electric field). The line is the best linear fit.

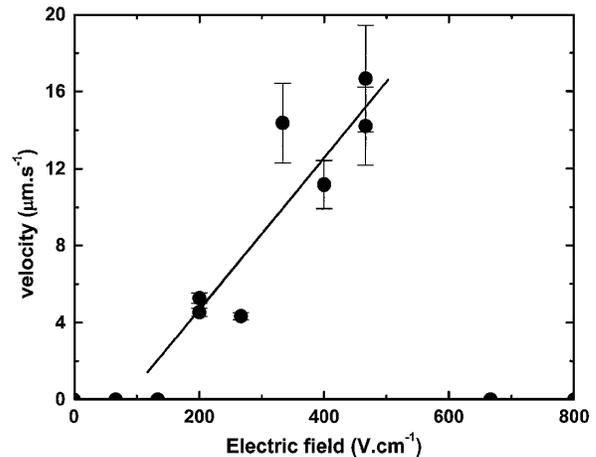


FIG. 4. Macroscopic velocity of a $3 \mu\text{m}$ particle submitted to a 2 Hz sine ac electric field as a function of the intensity of the applied electric field. The line is the best fit in the linear region.

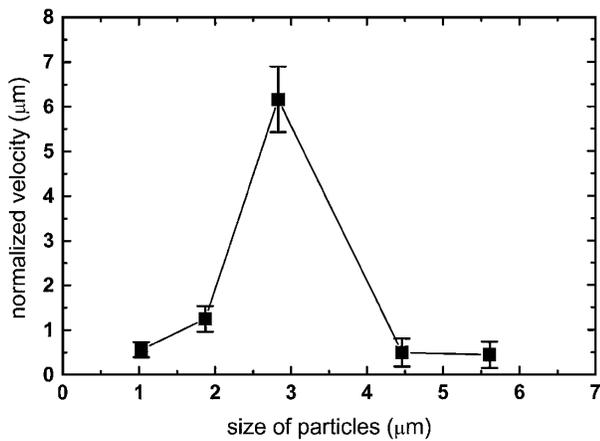


FIG. 5. Size discrimination for a 15 Hz square signal. The velocity is normalized by the frequency of the applied field. Each point corresponds to a separate experiment in which the particles of a given size are driven with an amplitude of ca. $100 \mu\text{m}$.

curves could be superimposed into a single one by normalizing the velocity by the frequency. This normalized velocity, that is plotted in Fig. 5 as a function of particle size, scales as a length: It is the average net distance over which a particle moves during a time period.

One can observe a strong maximum for $3 \mu\text{m}$ size particles: In the most extreme case ($100 \mu\text{m}$ oscillation, 15 Hz), macroscopic velocities up to $120 \mu\text{m} \cdot \text{s}^{-1}$ have been measured [13]. Moreover, experiments conducted with 3 and $4.5 \mu\text{m}$ beads together also showed that the velocities were different for the two sizes although the local excursions were somewhat different.

We have also made preliminary experiments with a channel whose constrictions are narrower (6 vs $12 \mu\text{m}$ in the experiments described above). We find that, in this case, the maximum is obtained for smaller particles. $2 \mu\text{m}$ particles move systematically faster than $3 \mu\text{m}$ particles. A complete study of the influence of the geometry of the channel on the optimal size is beyond the present Letter but there is no question that it is a powerful way to tune the precise size range of interest.

In conclusion, we have shown in this Letter that rectification is observed for colloidal particles in relatively large structures. Although we are still far from a real separation, and without any data concerning the dispersion of the observed motion, the contrast in velocities with the size is spectacular enough to offer a potential framework for a true separation device. With the simple microfabrication technology used here, we are already in the right size range for working with cells or even organelles. A technologically reasonable reduction in sizes (with neck sizes around $1 \mu\text{m}$) would let us go toward more molecular scales and

to regimes for which the sign itself of the mobility can be a function of the size [4].

It is a pleasure to thank A. Ajdari, J. Prost, P. Hänggi, T. O. Tegenfeldt, and R. H. Austin for discussions and support. Financial support from the ACI “physico-chimie de la matière complexe” is gratefully acknowledged.

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