Colloid particles in microfluidic inertial hydrodynamic ratchet at moderate Reynolds number

František Slanina*
Institute of Physics, Czech Academy of Sciences, CZ-18221 Praha, Czech Republic

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The movement of spherical Brownian particle carried by an alternating fluid flow in a tube of periodically variable diameter is investigated. On the basis of our previous results [Phys. Rev. E **99**, 012604 (2019)] on the hydrodynamics of the problem, we look at the competition of hydrodynamics and diffusion. We use the method of Fick-Jacobs mapping on an effective one-dimensional problem. We calculate the ratchet current and show that is is strictly related to finite size of the particles. The ratchet current grows quadratically with particle radius. We also show that the dominant contribution to the ratchet current is due to inertial hydrodynamic effects. This means that Reynolds number must be at least of order one. We discuss the possible use for separation of particles by size and perspectives of optimization of the tube shape.

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I. INTRODUCTION

With the development of microfluidics [1,2], the laboratory-on-chip applications are becoming widely available for applications in technology, medical diagnostics [3], and so on. One of the basic tasks in construction of microfluidic devices still remains separation of micrometer and submicrometer particles according to physical properties including size, shape, and rigidity [4–9].

Sorting of large particles (typical size 10 μm) relies mostly on deterministic hydrodynamic techniques. Among them, the most widely used are deterministic lateral displacement [10] and lateral migration [11,12] based on the Segre-Silberberg effect [13]. The latter method found numerous medical applications [14–19].

For small particles (typical size under 1 μm) Brownian motion plays important role. In this case, sorting may rely on the well-known mechanism of Brownian ratchet [20,21]. Three ingredients are necessary for the Brownian ratchet to work. They are periodic driving, mirror-asymmetric geometry, and diffusion. The driving may be due to external potential or by hydrodynamics. Here we shall consider the latter variant. In this setting, particles are suspended in a fluid flowing in a tube and the flow is periodically varied so that total fluid flow over one time period is zero. The hydrodynamic Brownian ratchet was realized experimentally [22–24] and studied theoretically by direct simulations [25] and using the mapping on one-dimensional diffusion problem [26].

Brownian ratchets with purely hydrodynamic driving are more delicate than ratchets driven by external field. For example, infinitesimally small particles in stationary flow under the influence of Brownian motion are, in a stationary state, distributed uniformly over the interior of the tube. Therefore, if the alternation of the flow is so slow that adiabatic approximation is valid, then the flow of particles follows truly the flow of the fluid and the ratchet effect is exactly zero. Therefore, we need particles of either finite size or fast-enough oscillation of the flow in order to observe the ratchet effect.

In a series of articles [27,28] we solved the problem of movement of a spherical particle in axially symmetric tube with periodically varying diameter. In the first step, we solved the Navier-Stokes equations for undisturbed stationary flow in such tube, using expansion in powers in the amplitude of diameter variation. This problem was investigated previously using the slow-variation method introduced by Blasius [29] to periodically modulated tubes [30–37]. However, we rely on small-amplitude expansion, which was used less often [38–43]. In Refs. [27,28] we improved the earlier results by providing explicit analytic formulas for first few terms of the expansion.

In the second step we inserted spherical particle in the flow and established the velocity if its forceless movement. We found that the results are reliable up to Reynolds numbers about Re $\lesssim 4$. This is enough to see inertial hydrodynamic effects and still it is far from hydrodynamic instabilities. We come to such a conclusion on the basis of numerical studies of the flow in similar geometries [43–46], which show that the flow is stable up to Reynolds numbers about Re $\simeq 200$.

In our previous article [28] we showed that for relatively large particles of the size $\simeq$10 μm (which is about the size of blood cells) we can realize ratchet effect in purely hydrodynamic regime with infinitesimally slow diffusion. However, the purely hydrodynamic ratchet works only within a transient regime, which means that the frequency of alternation of the flow must be large enough and the length of the tube must be small enough. When we approach the quasistationary regime, the ratchet effect disappears.

In this work, we want to incorporate the effect of Brownian motion. This implies that we work with relatively smaller particles, of sizes $\simeq$1 μm or less. Our strategy will start with the ordinary advection-diffusion equation, where the drift term will be taken from our calculation of the velocity of a particle freely carried by the flow. Then the advection-diffusion

*slanina@fzu.cz

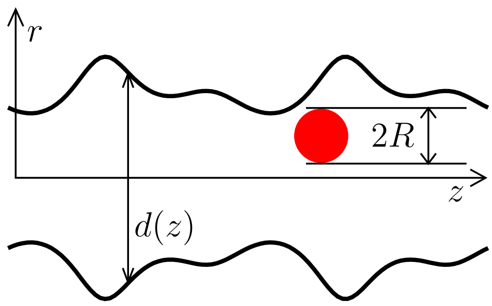

FIG. 1. Schematic picture of the system investigated here. The fluid flows through axially symmetric tube of periodically varying diameter $d(z)$. Within the fluid, there is a colloid particle of spherical shape and radius R.

equation will be mapped on an effective one-dimensional problem, using a simple variant of the Fick-Jacobs approximation [47,48]. This approximation was widely used to many problems involving diffusion and was developed systematically as a power series, which can be even partially summed [26,48–59]. We shall not employ the full power of this method here; we shall use just the most basic version of it. The mapping on one-dimensional problem will allow us to calculate the stationary current and hence the ratchet current in adiabatic approximation. This way we establish the properties of our version of Brownian ratchet.

At this point we must admit a weak point of our previous work [27]. We already tried to apply the Fick-Jacobs approximation there and obtained a compact analytic formula within certain range of parameters. However, when we checked later the values of physical quantities for which the approximation leading to such formula (Eq. (22) in Ref. [27]) holds, we came to a disappointing conclusion. If the liquid in question was water at room temperature, then the formula would hold only at spatial scales smaller than 10^{-15} m, which is completely beyond reality. Larger viscosity would make it even worse. To extend the result of Ref. [27] to wider and more realistic range of parameters, we must apply the Fick-Jacobs approximation in a more robust way and this is just the scope of the present work.

II. PARTICLE CARRIED BY FLOW IN TUBE

A. Geometry

We investigate movement of spherical particle of radius R carried by an incompressible fluid flow and performing Brownian motion. The flow occurs in an axially symmetric tube with spatially variable but periodic diameter. We use cylindrical coordinates, so that the coordinate z is oriented along the tube axis and r is the radial coordinate. Due to the symmetry of the problem, we assume no dependence on the angular coordinate ϕ in any measurable quantity. The system investigated here is sketched schematically in Fig. 1. The diameter of the channel at axial coordinate z is

$$d(z) = \frac{d}{1 + S(z)}, \tag{1}$$

where the parameter d will be called typical diameter, and the modulation function $S(z)$ is periodic, with spatial frequency Ω. Generally, we can expand it in Fourier series,

$$S(z) = \sum_{k=1,2,\dots} (A_k \sin k\Omega z + B_k \cos k\Omega z). \tag{2}$$

We shall often measure the lengths in terms of the spatial period of the modulation $L = 2\pi/\Omega$. This way, we define dimensionless parameters $\overline{d} = d\Omega$ and $\overline{R} = R\Omega$.

B. Hydrodynamics

We shall suppose that the volumetric flow of the fluid through the tube is Q. This is the basic parameter which quantifies the flow. It can be either positive or negative depending on the orientation of the flow with respect to the z axis. We denote η the dynamic and ν the kinematic viscosity of the fluid. The first step in investigating our problem is the solution of Navier-Stokes equations for the fluid in the tube with geometry defined by (1).

There are a few quantities which give rough but useful characteristics of the flow. Most importantly, we define the Reynolds number as

$$\mathrm{Re} = \frac{2\Omega|Q|}{\pi\nu}. \tag{3}$$

We are interested in regime with significant inertial effects but still far from any hydrodynamic instabilities. This implies that typical values of Reynolds number will be around $\mathrm{Re} \simeq 1$.

It is also useful to have some idea on the fluid velocities and pressures in the system. So we define the typical fluid velocity as $U = 4Q/\pi d^2$. The typical pressure drop per spatial period can be estimated using the Hagen-Poisseuille formula as

$$\Delta p = \frac{256\eta\Omega^3 Q}{\overline{d}^4}. \tag{4}$$

Of course, the actual pressure drop must be obtained by full solution of NS equations, which is a difficult task. The typical pressure drop (4) is just a rough, however useful, estimate for the actual value.

In recent works [27,28], we found the solution of the Navier-Stokes (NS) equations for the stationary flow in a tube with periodically varying diameter according to (1) and (2).

We used the expansion in the amplitudes of the modulation A_k and B_k. The solution is expressed in terms of the stream function ψ. The axial and radial coordinates of the fluid velocity $\mathbf{u}$ are related to the fluid stream function by

$$u_r = -\frac{1}{r}\frac{\partial\psi}{\partial z}, \quad u_z = \frac{1}{r}\frac{\partial\psi}{\partial r}. \tag{5}$$

The boundary conditions for NS equations were the usual no-slip conditions at the tube wall placed at $r = h(z) \equiv d(z)/2$, combined with the periodic boundary conditions in the z direction, $\psi(r,z) = \psi(r, z + 2\pi/\Omega)$ valid in the stationary state.

We look for the solution in the following general structure:

$$\psi(r,z) = \frac{2Q}{\pi}[\psi_0(r,z) + \psi_1(r,z) + \psi_2(r,z) + \dots], \tag{6}$$

where ψ_m contains mth powers of the amplitudes A_k, B_k. We proceed by inserting (6) into NS equations and collect all terms of the same order in the coefficients A_k, B_k. The terms of zeroth order in A_k, B_k lead to the contribution

$$\psi_0(r,z) = 2\left\{\frac{[1+S(z)]r}{d}\right\}^2 - 4\left\{\frac{[1+S(z)]r}{d}\right\}^4. \quad (7)$$

Using this term, we obtain equations for the contribution of first order in A_k, B_k. In principle we can proceed iteratively to any order, but already beyond the first order the equations become unmanageable. That is why we stopped at the first order in the expansion (6) (see Refs. [27,28] for details).

In the first order, we can separate contributions which are symmetric and antisymmetric with respect to the orientation of the flow, i.e.,

$$\psi_1(r,z) = \psi_{1\,\mathrm{even}}(r,z) \pm \psi_{1\,\mathrm{odd}}(r,z)\mathrm{Re}, \quad (8)$$

where the "+" sign applies for $Q > 0$ and "−" sign for $Q < 0$. The even and odd parts can be written as

$$\begin{aligned}\psi_{1\,\mathrm{even}}(r,z) = &\sum_{k=1,2,\dots}\sum_{l=0}^{\infty}(-1)^l(k\,\mathrm{Re})^{2l}\\ &\times g^{(2l)}\left(k\Omega[1+S(z)]r;\frac{k\Omega d}{2}\right)\\ &\times(B_k\cos k\Omega z + A_k \sin k\Omega z)\end{aligned} \quad (9)$$

and

$$\begin{aligned}\psi_{1\,\mathrm{odd}}(r,z) = &\sum_{k=1,2,\dots}\sum_{l=0}^{\infty}(-1)^{l+1}k(k\,\mathrm{Re})^{2l}\\ &\times g^{(2l+1)}\left(k\Omega[1+S(z)]r;\frac{k\Omega d}{2}\right)\\ &\times(A_k\cos k\Omega z - B_k \sin k\Omega z).\end{aligned} \quad (10)$$

The nontrivial part of the solution is contained in the functions $g^{(m)}(x;y)$ which can be expressed in terms of integrals containing Bessel functions. For details of the solution we refer the reader to Refs. [27,28].

The solution obtained in Refs. [27,28] was found reliable for Reynolds numbers $\mathrm{Re} \lesssim 4.5$ (see discussion in Refs. [27,28] for details).

When we insert a spherical, neutrally buoyant particle into an ambient flow described by velocity field $\mathbf{u}$, free movement of the particle is described by the velocity field $\mathbf{v}$ related to $\mathbf{u}$ by the well-known formula [28,60]

$$\mathbf{v} = \mathbf{u} + \frac{R^2}{6}\Delta\mathbf{u} + O(R^4). \quad (11)$$

The terms of the order R^4 and higher stem from the nonlinear terms in Navier-Stokes equations. In Stokes flow they are absent [60]. In this work we neglect the contributions of order higher than R^2. This approximation has an important technical consequence. We can see that the velocity field of particles $\mathbf{v}$ is incompressible (has zero divergence) as long as the flow $\mathbf{u}$ of the fluid itself is incompressible. Therefore, in analogy with the stream function for the fluid, we can define the function ψ_p (we shall call it particle stream function) from which the particle velocity $\mathbf{v}$ can be computed as

$$v_r = -\frac{1}{r}\frac{\partial\psi_p}{\partial z}, \quad v_z = \frac{1}{r}\frac{\partial\psi_p}{\partial r}, \quad (12)$$

in analogy with (5). The formula (11) taken up to quadratic order in R, induces linear relation between ψ and ψ_p, which is [28]

$$\psi_p = \psi + \frac{R^2}{6}\left(\frac{\partial^2}{\partial z^2} + \frac{\partial^2}{\partial r^2} - \frac{1}{r}\frac{\partial}{\partial r}\right)\psi. \quad (13)$$

Note that the differential operator present in (13) is *not* the Laplace operator expressed in cylindrical coordinates, as one might naively think looking at the form of (11).

Now we can insert the solution of the NS equations expressed by Eqs. (6) to (10) into the relation (13) and thus obtain the movement of the particle induced by the fluid flow. This movement was thoroughly discussed in Ref. [28], so let us stress only the most important finding. In fact, although the particle velocity field $\mathbf{v}$ is incompressible, the streamlines cross the tube walls. The physical meaning is that the fluid flow pushes the particles toward the wall at some points, while at other points the particles are repelled from the wall. Therefore, hydrodynamic traps are formed. In Ref. [28] we have shown that in transient regime of alternating flow, such hydrodynamic traps induce a ratchet effect, which may be used for particle separation based on purely deterministic hydrodynamic effects. However, such hydrodynamic ratchet works only in a specific window of parameters. First, it relies on transient effects, so that the frequency of the alternation of the flow cannot be too small. Second, Brownian motion of the colloid particles is neglected. The motion of the particles is totally deterministic.

C. Diffusion

The aim of the present work includes investigation of the influence of Brownian motion of the particles. Therefore, we study the competition between deterministic hydrodynamics and stochastic diffusion in their movement.

The tool used will be a usual advection-diffusion equation, into which the induced particle velocity (12) enters as an input. The diffusion coefficient of our spherical particle is

$$D = \frac{k_B T}{6\pi\eta R}. \quad (14)$$

We denote $\rho(r,z,t)$ the probability density for finding the particle at coordinates (r,z) in time t. In all what follows we assume that the probability density is axially symmetric, so the dependence on the azimuthal angle ϕ is absent. It satisfies the advection-diffusion equation in the form

$$\partial_t\rho = \left[\partial_z(D\partial_z - v_z) + \frac{1}{r}\partial_r r(D\partial_r - v_r)\right]\rho, \quad (15)$$

where we used abbreviated notation ∂_t, ∂_z, ∂_r for partial derivatives with respect to time and coordinates z and r, respectively. The advection velocities v_z and v_r are taken from the solution (6) of the flow according to relations (13) and (12).

The boundary conditions are

$$(D\partial_r - v_r)\rho(r,z,t)|_{r=0} = 0 \quad (16)$$

at the tube axis. This follows from the assumed axial symmetry of the solution which implies that the flow crossing the axis must be strictly zero. At the tube wall, the reflection boundary conditions imply

$$(D\partial_r - v_r)\rho(r,z,t)|_{r=d(z)/2} = \tfrac{1}{2}d'(z)(D\partial_z - v_z)\rho(r,z,t)|_{r=d(z)/2}. \tag{17}$$

To simplify the treatment of the diffusion problem, we perform mapping on a one-dimensional effective model, using the Fick-Jacobs approximation [47,48] in the lowest order. As there are several variants of such approximation [26,48–59] which differ in sometimes important details, we describe shortly the procedure used by us here. In fact, it is the simplest nontrivial version of the Fick-Jacobs approximation.

We want to write a closed equation for the projected density

$$p(z,t) = \int_0^{d(z)/2} 2\pi r\, \rho(r,z,t)\, dr. \tag{18}$$

Projecting the diffusion equation (15) and using the boundary conditions (17) we obtain an exact (but not closed) equation,

$$\partial_t p(z,t) = D\partial_z^2 p(z,t) - \partial_z\left[D\frac{\pi}{2}d'(z)d(z)\rho\left(\frac{1}{2}d(z),z,t\right) + \int_0^{d(z)/2} 2\pi r\, v_z(r,z)\rho(r,z,t)\, dr\right]. \tag{19}$$

To close the equation we assume that the diffusion is fast enough to quickly equilibrate the probability density in radial direction. Then we apply approximate backward mapping

$$\rho_{\text{approx}}(r,z,t) = \frac{p(z,t)}{A(z)}, \tag{20}$$

where $A(z)$ is fixed by consistency condition as $A(z) = \int_0^{d(z)/2} 2\pi r\, dr = \pi d^2(z)/4$. Replacing in (19) the true ρ by the approximate one (20), we obtain closed equation for $p(z,t)$. This equation has the general form

$$\partial_t p(z,t) = D\partial_z e^{-W(z)}\partial_z e^{W(z)} p(z,t). \tag{21}$$

Systematic improvement of this scheme is possible [26,48–59], using more sophisticated formulas for the backward mapping than (20), but we shall use only this lowest approximation. The limits of the Fick-Jacobs approximation and its generalizations are still under debate [52,54] and no easy criterion seems to be at hand. Generally, of course, quicker diffusion favors equilibration and therefore smaller particles (which have larger diffusion coefficient) are better described by the Fick-Jacobs scheme.

The function $W(z)$ represents effective one-dimensional potential in which the particle diffuses. All geometric and hydrodynamic effects are amalgamated into this function. We find that $W(z)$ satisfies

$$\partial_z W(z) = -2\frac{d'(z)}{d(z)} - \frac{1}{D}\frac{4}{\pi d^2(z)}\int_0^{d(z)/2} 2\pi r\, v_z(r,z)\, dr. \tag{22}$$

The integral on the right-hand side of (22) can be formally calculated using the particle stream function $\psi_p(r,z)$. Indeed,

$$\int_0^{d(z)/2} 2\pi r\, v_z(r,z)\, dr = 2\pi\{\psi_p[d(z)/2,z] - \psi_p(0,z)\} = Q_p(z). \tag{23}$$

The quantity $Q_p(z)$ introduced here will be called nominal particle flow. Note that this quantity depends on the position along the tube axis. For point particles the quadratic (and higher) corrections in (11) vanish and $Q_p(z) = Q$ for all z.

The reason $Q_p(z)$ is position dependent is related to the feature of the particle velocity field (11) which was amply discussed in our previous works [27,28] and already mentioned in the previous section. Indeed, the point is that the streamlines of the particle velocity field $\mathbf{v}$ cross the tube wall. Physically, it implies that the particles are pushed toward the walls at some places and repelled from the walls at other places, as we already mentioned. Formally, this effect results in position dependence of the nominal particle flow $Q_p(z)$. At the same time, we must keep in mind that the actual particle flow calculated from Eq. (15) as well as from its approximate projection (21) must obey particle-conservation law and in stationary state must not depend on position along the axis. The dependence of $Q_p(z)$ on position is reflected in the effective potential $W(z)$ as a sequence of traps and barriers, which represent the true physical phenomenon of pushing and pulling the particle to and from the tube wall.

Integrating (22) we obtain the following formula for the effective potential:

$$W(z) = -\ln d^2(z) - \frac{4}{\pi D}\int_0^z \frac{Q_p(z')}{d^2(z')}dz'. \tag{24}$$

Now we can solve the projected diffusion equation (21) in stationary state. We assume periodicity of the probability density $p(z) = p(z+L)$ and fix the normalization such that there is on average one particle per one period, $\int_0^L p(z)dz = 1$. Then, the particle current is given by [20]

$$J = D\frac{1 - \exp\left[-\int_0^L \frac{4Q_p(z')}{\pi D d^2(z')}dz'\right]}{\int_0^L\int_z^{z+L} \frac{d^2(z)}{d^2(z')}\exp\left[-\int_z^{z'} \frac{4Q_p(z'')}{\pi D d^2(z'')}dz''\right]dz'dz}. \tag{25}$$

This formula will be the starting point for further investigations.

III. RATCHET EFFECT

A. Adiabatic approximation

Rectification of particle current can occur in periodically driven diffusive systems if the static potential in which the particle moves lacks mirror symmetry. In our case there is no physical potential, as the particle moves freely within the tube, influenced only by hydrodynamic forces. However, effectively the particle feels tilted periodic potential $W(z)$. This effective potential has two sources, represented by two terms on the right-hand side of Eq. (24). The first term stems purely from the geometry of the tube. It can be interpreted as entropic term, equal to logarithm of the space available for particle movement. The second term is the most interesting one, because it contains hydrodynamic effects. In fact, hydrodynamic and geometric effects are inseparably mixed in this term, but we shall see later that the effect of hydrodynamic parameters, like the Reynolds number, can be singled out in the final formulas.

The ratchet effect requires time-dependent periodic driving. In our case it is achieved by periodic change of the volumetric flow $Q(t)$. We shall assume adiabatic approximation, i.e., the period of the change is much longer than any transient times present in the system. Therefore, at each moment the state of the system may be considered stationary. We assume rectangular pattern of the time dependence of $Q(t)$. In the first half-period the volumetric flow is constant and positive, $Q(t) = |Q|$; in the next half-period, it is constant and negative, $Q(t) = -|Q|$. Then, the ratchet current is the average of stationary currents $J = J_>$ taken at $Q > 0$ and $J = J_<$ taken at $Q < 0$. The ratchet current is then

$$J_{\text{rat}} = \tfrac{1}{2}(J_< + J_>). \tag{26}$$

The currents $J_\gtrless$ are calculated using the formula (25). Actually, we shall rather use the ratchet velocity

$$v_{\text{rat}} = \frac{2\pi}{\Omega} J_{\text{rat}}, \tag{27}$$

which is independent of the normalization of the particle density.

An important observation is due here. Within the adiabatic approximation, we suppose that the system is infinitesimally close to stationary state at all times, despite the fact that the fluid flow $Q(t)$ is time dependent. But the stationary state has an important property which is evident from the form of the advection-diffusion equation (15). If the particles are pointlike, i.e., $R = 0$, then the drift velocity $\mathbf{v}$ is equal to the fluid velocity and the stationary solution of (15) is uniform. Therefore, the true particle current is proportional to the volumetric flow $Q(t)$ at all times. Since the fluid flow is unbiased, the volumetric flow as well as the particle current is zero, when averaged over time period. This means that the ratchet current is exactly zero. The conclusion is that the ratchet effect is strictly related to finite size of the particles. In the following, we shall see explicitly how the ratchet velocity decreases to zero when $R \to 0$.

B. Series expansion

The direction of the current is contained in the nominal particle flow $Q_p(z)$. We shall write $Q_p(z) = Q\overline{Q}(z)$, where $\overline{Q}(z) = 1$ if the particle is pointlike. Nonzero particle radius brings z-dependent corrections of order R^2 to $\overline{Q}(z)$. Moreover, inertial effects in the NS equation add further corrections to $\overline{Q}(z)$ which depend both on z and on the orientation of the flow. Therefore, we shall distinguish $\overline{Q}(z) = \overline{Q}_>(z)$ if $Q > 0$ and $\overline{Q}(z) = \overline{Q}_<(z)$ if $Q < 0$.

In the following, we shall use dimensionless coordinate $\overline{z} = z\Omega$. We also introduce function

$$g(\overline{z}) = [1 + S(\overline{z})]^2, \tag{28}$$

which contains geometric effects and functions

$$G_\gtrless(\overline{z}, \zeta) = \int_0^{\zeta} g(\overline{z} \pm \zeta')\overline{Q}_\gtrless(\overline{z} \pm \zeta')d\zeta', \tag{29}$$

which will be useful later. Inserting (25) into (26) we obtain a closed equation for the ratchet velocity,

$$v_{\text{rat}} = \frac{4\pi\nu\Omega\text{Re}\int_0^{2\pi}\frac{V_1(\overline{z})}{g(\overline{z})}d\overline{z}}{\overline{d}^2\left\{\left[\int_0^{2\pi}\frac{V_0(\overline{z})}{g(\overline{z})}d\overline{z}\right]^2 - \left[\int_0^{2\pi}\frac{V_1(\overline{z})}{g(\overline{z})}d\overline{z}\right]^2\right\}}. \tag{30}$$

The central point is the calculation of functions $V_0(\overline{z})$ and $V_1(\overline{z})$. They are expressed as

$$\begin{aligned} V_0(\overline{z}) &= \tfrac{1}{2}[V_<(\overline{z}) + V_>(\overline{z})] \\ V_1(\overline{z}) &= \tfrac{1}{2}[V_<(\overline{z}) - V_>(\overline{z})], \end{aligned} \tag{31}$$

where

$$V_\gtrless(\overline{z}) = \frac{1}{\overline{Q}_\gtrless(\overline{z})} \pm \int_0^{2\pi} \frac{e^{-\beta G_\gtrless(\overline{z},\zeta)}}{1 - e^{-\beta G_\gtrless(\overline{z},2\pi)}} \frac{d}{d\overline{z}} \frac{1}{\overline{Q}_\gtrless(\overline{z} \pm \zeta)} d\zeta. \tag{32}$$

The dimensionless parameter

$$\beta = \frac{12\pi\eta\nu\overline{R}}{kT\overline{d}^2\Omega}\text{Re}, \tag{33}$$

which appears in the exponentials in (32) plays central role in the approximations we shall use in the following. Indeed, for reasonable values of physical variables, β can be considered very large and expansion in inverse powers of β is appropriate.

As an example, consider the spatial frequency $\Omega = 10^5$ m^{-1}, reduced tube diameter $\overline{d} = 0.3$, and reduced particle radius $\overline{R} = 0.05$. For such choice the the spatial period is $2\pi \times 10^{-5}$ m, tube diameter is 3 μm and particle radius 0.5 μm. If the flow velocity is adjusted to Re = 1, considering pure water at 293 K, then the typical fluid velocity is $U = 2.23$ ms^{-1} and the typical pressure drop per one spatial period, according to (4), is $\Delta p = 0.5$ MPa. For such parameters, we have $\beta = 5.20 \times 10^7$, i.e., a very large value. Note that even particles as small as $2R \simeq 1$nm (with other parameters unchanged) give $\beta \simeq 10^4$, which is still very large. Only at Reynolds numbers as small as Re $\simeq 10^{-3}$ may we expect that large-β expansion may come into problems. However, such a slow motion produces too weak ratchet effect to be of practical interest, as will be clear later. So we conclude that expansion on powers of β^{-1} seems reasonable in all physically interesting situations in devices of typical sizes $\simeq$10 μm and Reynolds numbers of orders Re $\simeq 10^{-1}$ to Re $\simeq 1$. This will be the range of Reynolds numbers investigated here.

So let us turn to calculation of the functions $V_\gtrless(\overline{z})$ in the limit $\beta \to \infty$. It consists of three steps. First, the term $e^{-\beta G_\gtrless(\overline{z},2\pi)}$ is neglected in the denominator of (32). Second, the upper limit of the integration in (32) can be prolonged to infinity. Third, the function $G_\gtrless(\overline{z}, \zeta)$ in the exponent can be expanded in Taylor series around the point $\zeta = 0$, thus generating a series in powers of the small parameter β^{-1}. In terms of the even and odd parts $V_\sigma(\overline{z})$ for $\sigma \in \{0, 1\}$, we can write

$$V_\sigma(\overline{z}) = \sum_{m=0}^{\infty}(\beta g(\overline{z}))^{-m}V_{\sigma,m}(\overline{z}). \tag{34}$$

For the first two terms, we can easily find

$$V_{\sigma,0}(\overline{z}) = \frac{1}{2}\left[\frac{1}{\overline{Q}_<(\overline{z})} + (-1)^\sigma \frac{1}{\overline{Q}_>(\overline{z})}\right]$$
$$V_{\sigma,1}(\overline{z}) = \frac{1}{2}\left[\frac{\overline{Q}_<'(\overline{z})}{\overline{Q}_<^3(\overline{z})} + (-1)^\sigma \frac{\overline{Q}_>'(\overline{z})}{\overline{Q}_>^3(\overline{z})}\right]. \quad (35)$$

We have computed the particle flow up to first order in the Reynolds number [28]. Within such approximation we can write for the nominal flow of particles

$$\overline{Q}_{\gtrless}(\overline{z}) = 1 + \overline{R}^2\,\overline{Q}_0(\overline{z}) \pm \overline{R}^2\mathrm{Re}\,\overline{Q}_1(\overline{z}). \quad (36)$$

The form of the functions $\overline{Q}_0(\overline{z})$ and $\overline{Q}_1(\overline{z})$ is related to the solution of Navier-Stokes equations through the relations (9), (10), (13), and (23). Detailed expressions are given later.

In order to simplify the formulas we denote the expression

$$N(\overline{z}) = [1 + \overline{R}^2\,\overline{Q}_0(\overline{z})]^2 - (\overline{R}^2\mathrm{Re}\,\overline{Q}_1(\overline{z}))^2, \quad (37)$$

which will always occur in the denominator. Using these functions we can write

$$V_{0,0}(\overline{z}) = \frac{1}{N(\overline{z})}[1 + \overline{R}^2\,\overline{Q}_0(\overline{z})]$$
$$V_{1,0}(\overline{z}) = \frac{1}{N(\overline{z})}\overline{R}^2\mathrm{Re}\,\overline{Q}_1(\overline{z}) \quad (38)$$

for the lowest order in β^{-1}.

We can see that in this order, the dependence of the ratchet velocity on both particle radius and the Reynolds number is quadratic, as long as $\overline{R}$ and Re are small enough but not so small that the large-β approximation becomes invalid. We can also immediately see that the contribution of this term vanishes if the inertial hydrodynamic effects are omitted.

In the next order in β^{-1} we obtain the following formulas:

$$V_{0,1}(\overline{z}) = \frac{1}{N^3(\overline{z})}(\overline{R}^2\,\overline{Q}_0'(\overline{z})\{3[1 + \overline{R}^2\,\overline{Q}_0(\overline{z})]^2\overline{R}^2\mathrm{Re}\,\overline{Q}_1(\overline{z}) + (\overline{R}^2\mathrm{Re}\,\overline{Q}_1(\overline{z}))^3\}$$
$$- \overline{R}^2\mathrm{Re}\,\overline{Q}_1'(\overline{z})\{[1 + \overline{R}^2\,\overline{Q}_0(\overline{z})]^3 + 3[1 + \overline{R}^2\,\overline{Q}_0(\overline{z})](\overline{R}^2\mathrm{Re}\,\overline{Q}_1(\overline{z}))^2\})$$
$$V_{1,1}(\overline{z}) = \frac{1}{N^3(\overline{z})}(\overline{R}^2\,\overline{Q}_0'(\overline{z})\{[1 + \overline{R}^2\,\overline{Q}_0(\overline{z})]^3 + 3[1 + \overline{R}^2\,\overline{Q}_0(\overline{z})](\overline{R}^2\mathrm{Re}\,\overline{Q}_1(\overline{z}))^2\}$$
$$- \overline{R}^2\mathrm{Re}\,\overline{Q}_1'(\overline{z})\{3[1 + \overline{R}^2\,\overline{Q}_0(\overline{z})]^2\overline{R}^2\mathrm{Re}\,\overline{Q}_1(\overline{z}) + (\overline{R}^2\mathrm{Re}\,\overline{Q}_1(\overline{z}))^3\}). \quad (39)$$

For moderate values of radius and Reynolds number, this contribution is negligible compared to lowest order in β^{-1}, as will be shown quantitatively in the next section.

On the other hand, if we neglect inertial hydrodynamic effects, then the second-order term in β^{-1} is the lowest term which gives nonzero contribution to ratchet velocity. In other words, if we used the Stokes equation instead of the Navier-Stokes one, then the lowest term in the large-β expansion is just given by the equations (39). We can artificially "switch off" the inertial effects by forcing $\overline{Q}_1(\overline{z}) = 0$ everywhere. We can immediately see that in this case the contribution of (38) to the ratchet current is zero. At the same time, the contribution of (39) to the ratchet current is proportional to particle radius if the radius is small. We can see this effect quantitatively in the inset of Fig. 2. We can see that the ratchet velocity produced by the hypothetical flow governed by Stokes equation is so small (in picometer per second range) that it is out of practical consideration. We also checked the importance of terms of higher order in β^{-1}. We calculated explicitly the terms $V_{\sigma,m}$ for $m = 2, 3$. The formulas are analogous to (39) but more complicated. We consider unnecessary to show them here. Indeed, we found that quantitatively, their contribution to the data in Figs. 2 and 3 is smaller than the line width in these figures.

Strictly speaking, the expansion (11) neglects all contributions of order R^4 and higher, so we ought to neglect them also in the formulas for ratchet velocity. This leads to expression (keeping just the lowest power of β^{-1})

$$v_{\mathrm{rat}} = \frac{4\pi\nu\Omega\mathrm{Re}^2\overline{R}^2}{\overline{d}^2}\frac{\int_0^{2\pi}\frac{\overline{Q}_1(\overline{z})}{g(\overline{z})}d\overline{z}}{\int_0^{2\pi}\frac{1}{g(\overline{z})}d\overline{z}} + O(\overline{R}^4), \quad (40)$$

in which the quadratic dependence on both radius and Reynolds number is explicit. However, we prefer to proceed somewhat inconsistently, keeping all terms in the full

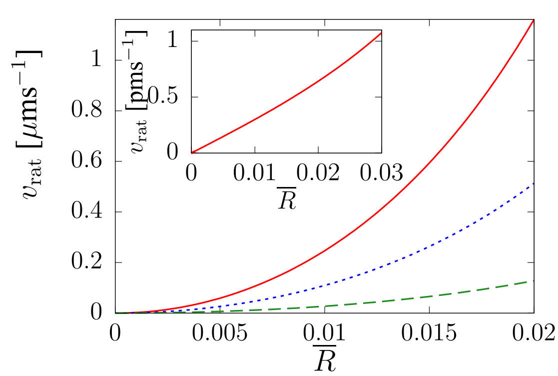

FIG. 2. Dependence of the average ratchet velocity of the particle on the reduced particle radius. The parameters of the tube are $\Omega = 10^5$ m^{-1}, $\overline{d} = 0.3$, $A = 0.15$, and $B = 0.2$. The Reynolds number is Re = 3 (solid line), Re = 2 (dotted line), and Re = 1 (dashed line). In the inset, average ratchet velocity for the same tube parameters and Re = 3, in the hypothetical case of neglected inertial effects.

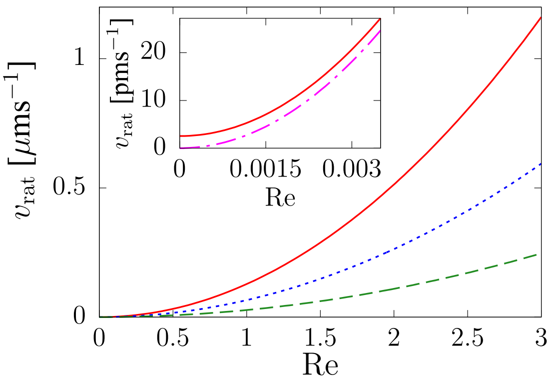

FIG. 3. Dependence of the average ratchet velocity of the particle on the Reynolds number. The parameters of the tube are $\Omega = 10^5\,\mathrm{m}^{-1}$, $\overline{d} = 0.3$, $A = 0.15$, and $B = 0.2$. The reduced radius of the particle is $\overline{R} = 0.02$ (solid line), $\overline{R} = 0.015$ (dotted line), and $\overline{R} = 0.01$ (dashed line). In the inset, average ratchet velocity for the same tube parameters and $\overline{R} = 0.05$. The result computed to the lowest (i.e., first) order in β^{-1} is drawn by dot-dashed line. The solid line shows the result computed up to second order in β^{-1}. Including third and fourth order in β^{-1} brings difference smaller than the width of the line.

expressions (37), (39), and (38) when calculating the ratchet velocity. One of the reasons is that for future we plan to calculate also higher terms in R in the expansion (11). Another reason is that particle diameter occurs also in the diffusion coefficient and through it in the parameter β, so the powers of $\overline{R}$ are mixed from several sources. Therefore, we consider more safe to keep all terms in the expressions.

C. Role of parameters

To calculate the ratchet velocity for given shape of the tube, we need the functions $\overline{Q}_0(\overline{z})$ and $\overline{Q}_1(\overline{z})$. Inserting the general solution (7)–(10) into (13) and (23), we finally obtain

$$\overline{Q}_0(\overline{z}) = \frac{2}{3}\Bigg\{-8\left[\frac{1+S(\overline{z})}{\overline{d}}\right]^2 - 2\left[\frac{S'(\overline{z})}{1+S(\overline{z})}\right]^2 + \sum_{k=1,2,\ldots} k^2\, g^{(0)\prime\prime}\left(\frac{k\overline{d}}{2};\frac{k\overline{d}}{2}\right) \times (B_k\cos k\overline{z} + A_k \sin k\overline{z})\Bigg\} \tag{41}$$

and

$$\overline{Q}_1(\overline{z}) = \frac{2}{3}\sum_{k=1,2,\ldots} k^3\, g^{(1)\prime\prime}\left(\frac{k\overline{d}}{2};\frac{k\overline{d}}{2}\right) \times (B_k \sin k\overline{z} - A_k\cos k\overline{z}). \tag{42}$$

Let us now specify the tube shape. If not stated differently, then we shall fix the spatial frequency $\Omega = 10^5\,\mathrm{m}^{-1}$ and reduced tube diameter $\overline{d} = 0.3$. Therefore, the spatial period of the modulation of the tube is $L = 62.8318\ldots\ \mu$m and the tube diameter oscillates around $d = 3\,\mu$m. We shall first work with a simple shape with just two Fourier components,

$$S(\overline{z}) = A\sin 2\overline{z} + B\cos\overline{z}. \tag{43}$$

This is the minimum model which yields nonzero ratchet current.

As we already stressed, the very occurrence of the ratchet effect is related to finite spatial extent of the particles. Therefore, the first question is in regard to how the ratchet velocity depends on the particle radius. We can see from (38) that $v_{\mathrm{rat}} \sim \overline{R}^2$ for small $\overline{R}$. Quantitatively, we can see this dependence in Fig. 2. We observe a dependence close to quadratic for all investigated sizes. To be precise, we neglected all contributions higher than quadratic in $\overline{R}$ from the beginning, see (11). So in order to include corrections to quadratic dependence consistently, we would need to go significantly deeper, starting with calculating corrections to (11).

For typical tube dimensions, $\Omega = 10^5\ \mathrm{m}^{-1}$ and particle size $R = 0.2\ \mu$m and for moderate Reynolds numbers $\mathrm{Re} \simeq 3$ the ratchet velocity is of order $v_{\mathrm{rat}} \simeq 1\ \mu\mathrm{ms}^{-1}$. This implies that in a real device consisting of tubes 10 spatial periods (i.e., about 0.6 mm) long, it takes about 10 min for a half-micrometer particle to traverse from one entrance to the other. Quadratic dependence on the size strongly enhances velocity of large particles, so that we can easily use such device for a "hydrodynamic chromatography," separating different fractions from a mixture.

In Fig. 2 we can also see that the ratchet velocity increases with Reynolds number. This dependence is shown in detail in Fig. 3. We can see that the dependence is quadratic, as hinted by equations (30) and (38). In fact, for the parameters as in Fig. 3, it is sufficient to consider just lowest term in the expansion in β^{-1}, i.e., $V_\sigma \simeq V_{\sigma,0}$. This is illustrated in the inset of Fig. 3. The effect of higher-order terms in β^{-1} becomes perceptible only for Reynolds numbers as small as $\mathrm{Re} \simeq 10^{-3}$. However, for such a slow flow, the ratchet velocity becomes practically irrelevant (picometers per second). Moreover, from the formal point of view, the large-β expansion breaks down for so small Re, as is also seen in the inset of Fig. 3. Indeed, the second term in the β^{-1} expansion gives unphysical nonzero limit for v_{r} when $\mathrm{Re} \to 0$, as can be deduced from (39) and as can be seen explicitly in the inset in Fig. 3.

A natural question is in regard to whether we can tune the ratchet velocity to desired level by the adjustment of the tube shape. We consider this question using somewhat more realistic sawtooth shape, in which the tube wall $h(\overline{z}) = \overline{d}/\{2[1+S(\overline{z})]\}$ is described by the function

$$h(\overline{z}) = \begin{cases} h_0 + \frac{a}{\pi-b}\overline{z} & 0<\overline{z}<\pi-b \\ h_0 + \frac{a}{\pi+b}(2\pi-\overline{z}) & \pi-b<\overline{z}<2\pi \end{cases}. \tag{44}$$

The constant h_0 is adjusted so that $S(\overline{z})$ is a sum of Fourier components with $k>0$ according to (2). [In practical calculations the Fourier expansion of the corresponding $S(\overline{z})$ was truncated at a quite large value $k = 100$.]

In our, rather simple, geometry, we can change two parameters, namely the parameter b, measuring the asymmetry, and a quantifying the amplitude of the corrugation of the tube. We show in Figs. 4 and 5 the dependence of the ratchet velocity on these parameters. Clearly, if either of them is zero, the ratchet effect is absent. Consistently, we observe that $v_{\mathrm{rat}} \to 0$ if either $b \to 0$ or $a \to 0$. For nonzero but small parameters, we observe that $v_{\mathrm{rat}} \sim a^2$ and $v_{\mathrm{rat}} \sim b$. Also in Fig. 4 we can compare the result for tubes of shapes (43) versus (44). We

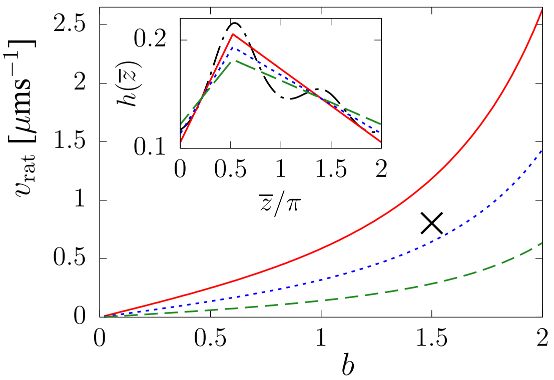

FIG. 4. Dependence of the average ratchet velocity of the particle on the parameter b determining the shape of the tube. Other parameters of the tube are $\Omega = 10^5$ m^{-1}, $\overline{d} = 0.3$, $a = 0.1$ (solid line), $a = 0.08$ (dotted line), and $a = 0.06$ (dashed line). Reynolds number is Re $= 2.5$, particle radius is $\overline{R} = 0.02$. The symbol $\times$ at $b = 1.5$ indicates the ratchet velocity for the same Re, $\overline{R}$, $\overline{d}$, and Ω, but in the tube with shape (43) with parameters $A = 0.15$, and $B = 0.2$. In the inset, function describing the tube wall. In dot-dashed line, we show the shape according to (43) with parameters $A = 0.15$, and $B = 0.2$. Other three lines are shapes according to (44) with $\overline{d} = 0.3$, $b = 1.5$, and $a = 0.1$ (solid line), $a = 0.08$ (dotted line), and $a = 0.06$ (dashed line).

can see that if the parameters A and B, or a and b, are adjusted so that the two shapes look similar, as compared in the inset of Fig. 4, the ratchet velocity is also similar in magnitude, as seen in the main plot of Fig. 4. This suggests that the minute details of the tube shape may be of little relevance for the ratchet transport. It is the overall form of the profile that counts.

The parameter b must lie within natural limits $b \in (-\pi, \pi)$. We always observed that the ratchet velocity increases with $|b|$. (Of course, for fixed a, $v_{\rm rat}$ changes sign when b changes sign.) From this point of view, maximally asymmetric sawtooth shape is the most efficient one. The parameter a is limited by requirement that there is nonzero aperture left in the tube, to allow passing particles. However, in our computations we observed that ratchet velocity always increases with a, even when a approaches this strict limit. This goes against natural expectation that the velocity must decrease when the aperture gets too narrow. The misleading behavior we observe at large a is related to the fact that the basic approximation made in solving the NS equations was keeping just terms which are linear in the Fourier coefficients A_k, B_k. For large a such approximation is unfounded. Hence, our calculations are unable to provide optimal value of the parameter a.

The second reason why our results for large a lose validity comes from the steric repulsion of the particles with tube walls. In this work, we neglect the steric effects completely. Large a implies small aperture of the tube at its narrowest position and further narrowing due to steric effects can be decisive. Not only the effective tube diameter is diminished for larger particles, but also the effective tube shape is altered. Therefore, the value of d, as well as all Fourier components in the expansion (2) depend on the particle radius. The effect of steric repulsion with walls was investigated, e.g., in Refs. [61,62], but we decided to neglect it, in order not to mix it with the inertial hydrodynamic effect, which is the focus of our work.

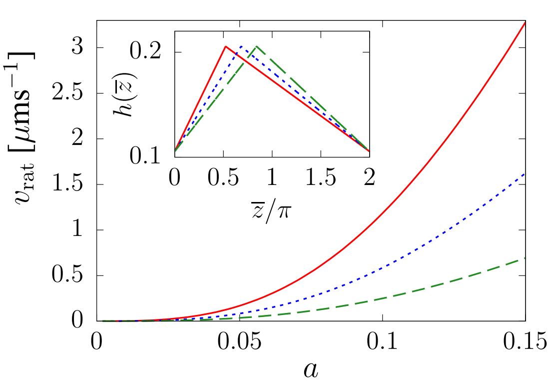

FIG. 5. Dependence of the average ratchet velocity of the particle on the parameter a determining the shape of the tube. Other parameters of the tube are $\Omega = 10^5$ m^{-1}, $\overline{d} = 0.3$, $b = 1.5$ (solid line), $b = 1.0$ (dotted line), and $b = 0.5$ (dashed line). Reynolds number is Re $= 2.5$, particle radius is $\overline{R} = 0.02$. In the inset, function describing the tube wall according to (44) with $\overline{d} = 0.3$, $a = 0.1$, and $b = 1.5$ (solid line), $b = 1.0$ (dotted line), and $b = 0.5$ (dashed line).

IV. CONCLUSIONS

Using our previous results [27,28] for the hydrodynamics of a spherical colloid particle in an axially symmetric tube with variable diameter we investigated the ratchet effect and possible particle separation in such setup. Particles are driven by periodically alternating fluid flow with no bias, so that the fluid flow averaged over one time period is zero. Note that this is not the same as driving by regularly oscillating pressure, because the nonlinear term in Navier-Stokes equations breaks the time-reversal symmetry. That is why the bias must be neutral in the flow, rather than in the applied pressure.

Our approach is based on the projection of the advection-diffusion equation on an effective one-dimensional diffusion problem. The mapping was performed using the simplest variant of the Fick-Jacobs approximation. The effective one-dimensional potential incorporates entropic effects due to the geometry of the tube, as well as hydrodynamic effects which are related to finite radius of the particle.

In order to calculate the particle current in the effective one-dimensional problem, we used the expansion in powers of the inverse of the large dimensionless parameter β, defined in (33). In fact, it is the Péclet number of our system, relating the magnitudes of hydrodynamic and diffusive transport. For typical values of parameters used in our work, it is of the order $\beta \simeq 10^7$. This means that hydrodynamic effects are decisive and diffusion plays an auxiliary role. However, as was also shown in our previous work [28] neglecting the diffusion completely would be a great mistake, because at least slow diffusion is indispensable for establishment of a nontrivial stationary state. Indeed, strictly pointlike particles do not exhibit any ratchet effect. The ratchet effect is possible only for nonzero particle radius. However, for such particles, hydrodynamic effects push the particles toward walls at some places and repel them from walls at other places. This induces an effective potential walls for the movement of particles. In absence of diffusion, these potential walls are unsurmountable and in stationary state all particles would be packed in potential traps. In reality this scenario is avoided due to at least weak diffusion. Alternatively, such an unphysical stationary state is bypassed by fast enough alternation of the fluid flow, i.e., keeping the system in a transient regime far from stationarity. The latter way was investigated in our previous work [28]. Here we looked at the influence of weak but finite diffusion in stationary state. Therefore, working at very large Péclet number β is appropriate here.

The results for stationary particle current found in this way was directly used for calculation of ratchet velocity in adiabatic approximation, i.e., in the limit of infinitesimally small frequency of the alternation of the fluid flow. Although in principle the entropic and hydrodynamic effects on the ratchet velocity are mixed, we found that hydrodynamics is dominant. We also found that inertial hydrodynamic terms, i.e., those originating from the nonlinear terms in Navier-Stokes equations, are several orders of magnitude larger than the terms present only in Stokes equations. This means that the use of bare Stokes equation in description of hydrodynamic ratchets is hopelessly insufficient.

We performed quantitative analysis of the ratchet velocity for tubes of typical diameter around $d = 3\,\mu$m at moderate Reynolds numbers Re $\lesssim 3$. The ratchet velocity was typically around $v_{\rm rat} \simeq 1\,\mu{\rm ms}^{-1}$. We found that the ratchet velocity grows quadratically with particle size, which allows practical application for particle separation.

As expected, the ratio of fluid velocity to ratchet velocity is of the same order as the Péclet number β. This is quite a large number, but the net particle velocity is still large enough to provide efficient separation of submicrometer particles at time scales of tens of minutes, considering a half-millimeter-thick membrane penetrated by pores of the size and shape investigated here.

We also tried to find optimal shape, which would provide maximum ratchet velocity. This effort was only partially successful. We investigated this question by varying parameters of sawtooth shape of the tube. Generically, we found that it is beneficial increasing the asymmetry of the tube as much as the geometry allows. We also found that the ratchet velocity increases when we increase the amplitude of the modulation of the tube, as expected. However, beyond certain value of the amplitude the velocity is expected to decrease again, due to narrowing the aperture of the tube. In our calculations we do not reach this regime, because the approximation used is principally limited to small amplitudes of the modulation. Indeed in the solution of Navier-Stokes equations just terms linear in the amplitude of the modulation are kept. Therefore, the attempt of optimization the shape must be taken with great care.

The method used in this work is open to improvements in several directions. First, it would be desirable to go beyond linear term in the amplitude of the tube modulation. We expect a lengthy but straightforward calculation. A new feature is that in linear approximation used here all Fourier components of the periodic modulation are independent, which simplifies the calculation a lot. Beyond linear approximation the Fourier components become coupled and an additional approximation will be necessary, keeping only a few lowest harmonics. The important gain from this effort would be much more reliable optimization of the tube shape with respect to ratchet velocity, as we discussed it before.

Second, in this work we completely neglected the effect of narrowing the available space within the tube due to finite particle size. This is a delicate task, because the shape of the available space is altered in a nontrivial way. In fact, it will affect all Fourier components of the tube modulation. Again, an approximation would be necessary, taking just finite number of these components and neglecting the rest. We believe there is no serious difficulty in doing that.

Third, we neglected also hydrodynamic interactions of the particles with walls. At this point, however, we are not aware of any directly applicable analytic procedure which would work in modulated tube. On the other hand, this effect is rather serious, because the ratchet phenomenon investigated here relies to great extent on the effect of pushing the particles toward the wall at some places. This effect will be surely influenced by hydrodynamic interactions to large extent. Therefore, there is a strong motivation to tackle this issue. We leave this problem open for future investigation.

Fourth, the formula for the velocity of sphere carried by flow can be improved by calculating the terms of fourth, or maybe even higher, order in particle radius.

Finally, all the theory presented here assumes small density of particles, so that the particle-particle interactions are neglected. Investigation of dense suspensions of colloids pose further challenge which goes beyond the scope of this work. Perhaps application of ideas borrowed from stochastic modeling of dense suspensions [63] could provide a path.

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