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Friction coefficient of rod-like chains of spheres at very low Reynolds numbers. I. Experiment

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Abstract. — We present a new method to measure hydrodynamic friction forces on colloidal particles at very low ($\simeq 10^{-5}$) Reynolds numbers with high accuracy. Exploiting magnetic field induced aggregation of microcopic paramagnetic spheres into straight chains of calibrated length, we determine the anisotropic friction coefficient of the chains, with a range from one to 100 particles, by video-microscopic observation of sedimentation speed. The results are found to agree well with the Slender-body theory, even for small chain lengths. The effects of the proximity of a wall are also explored.

There are various analytical and numerical solutions of the Stokes equations, which are the limit of the Navier-Stokes equations for vanishing Reynolds number [1-4]. In particular, the friction coefficient of a large class of bodies has been studied, including very long and thin — but otherwise — arbitrary shapes, the so-called slender-bodies. These theories were tested with experiments using particles of a few millimeters in size suspended in fluids with high viscosity such as glucose ($\mu \simeq 2$ kg/ms) [5, 6]. In this fashion Reynolds numbers as low as 10^{-3} were obtained, but a typical problem in these experiments is the relatively large liquid volume required to eliminate effects of the container walls on the measurement of the friction coefficient. The latter corrections are of the order d/h or $l/(h \ln(2l/d))$ for a body of spherical shape or a slender-body, respectively, where d and l are, respectively, the diameter and length of the body and h its distance from the wall. Thus the typical container size is in the range of one meter [6].

In this paper we present a novel, handy, small scale and accurate experimental technique to measure anisotropic friction coefficients of well defined bodies at fixed orientation. We use

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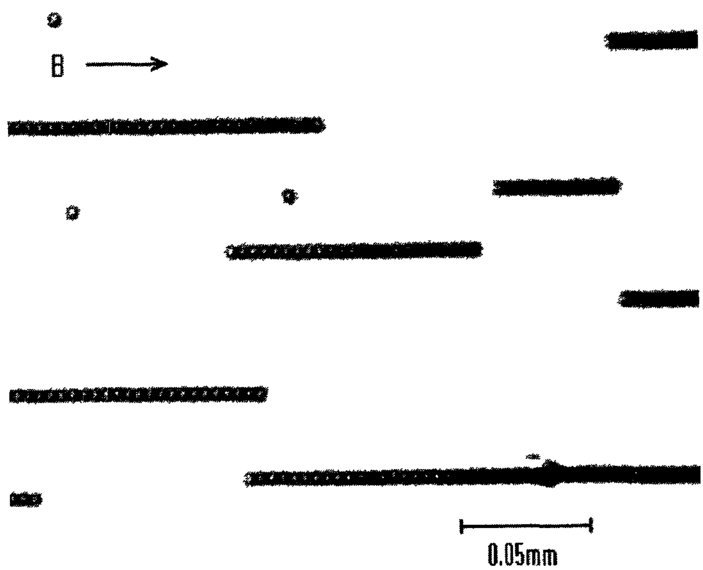

Fig. 1. — Microscopic picture of several colloidal chains of spheres; diameter of spheres 4.3 μm; the magnetic field $B \simeq 30$ mT is in the horizontal direction.

aqueous suspensions of μm-sized paramagnetic colloidal spheres, which are aggregated [7] into linear chains of calibrated length and fully aligned by application of a homogeneous magnetic field. Their sedimentation velocity is measured by video-microscopic observation and image processing. Exploiting the fact that both the characteristic velocity v and the linear dimension L are much smaller in colloidal systems, we naturally obtain single particle Reynolds numbers $R = v\,L\,\rho/\mu$ of 10^{-5} or below (where ρ and μ denote, respectively, the density and the viscosity of the liquid). In addition wall effects can be simply reduced because the required minimum container sizes are in the range of mm. We find that the sedimentation speed for motion both parallel and perpendicular to the chain axis agrees remarkably well with the Slender-body theory, when the chain is far enough from the container wall [3]. Moreover the slowing down due to the vicinity of a planar wall is found to agree well with numerical simulations [8].

The magnetic colloidal particles provided by the DYNAL company [9] were Fe_3O_4-doped polystyrene beads of diameter $d = 4.3\,\mu$m, stabilized with surface charges and suspended in water. Under the influence of an external homogeneous magnetic field they behave to the first order like induced paramagnetic dipoles. The dipole-dipole interaction leads to an attraction between two beads positioned along the magnetic field and, therefore, to the formation of perfectly regular chains of up to a hundred particles along this axis as shown in figure 1. In this way colloidal chains of various adjustable lengths were prepared. In fact, manipulations of chains with the help of small superimposed field gradients were used to obtain end to end aggregation, to position chains with respect to the wall, or to lift them up again for repeated sedimentation measurement of a given individual chain. By adding *horizontal* field gradients, alternatively to the left and the right, one single chain among those formed after injection was selected. Thus hydrodynamic interaction between chains could be excluded.

The particles were injected into a water-filled glass cell (of $10 \times 2\,\mathrm{mm}^2$ base and $10\,\mathrm{mm}$ height) through a capillary with an inner diameter of about 30 microns in order to minimize the resulting flow. Hereafter, the field was turned on. The cell diameter (2 mm) was large enough to avoid corrections due to wall effects for the chains with less than 30 beads. On the other hand, it was small enough so that the convective motion of the water due to thermal gradients across the cell was sufficiently damped.

However, even several hours after filling the cell, the residual of the induced motion of the liquid was still of the order of 1 μm/s, which had to be taken into account in the data analysis. This was done by adding a small concentration of undoped (diamagnetic) polystyrene beads to the suspension. They have a density of $\rho = 1.054$ kg/dm^3. Since this exceeds only slightly the density of water, their motion describes, after a small correction due to the weak sedimentation, the motion of the liquid and the measurements of the sedimentation speed of the chains can be corrected accordingly. The convective flow of the liquid was not constant over the cell. Therefore, immediately after each individual measurement of the sedimentation velocity of a given chain the thermal convection of the surrounding liquid had to be determined. Since the flow of the fluid in the vicinity of a moving chain is changed and these effects decrease, like wall effects, only with $1/r$ (r being the distance to the chain), the motion of the chains had to be stopped, for a reliable determination of the thermal convection. This was accomplished by introducing an additional force along the sedimentation axis of the chains by an appropriate magnetic field gradient. During the sedimentation speed measurement, the magnetic field was decreased to a point where it was just strong enough to maintain the chain structure and the orientation of the spheres. The remaining small perturbation of the measurements due to the magnetic field gradient cancels out in the final result, as discussed below.

The experimental results are plotted in figures 2 and 3. They show, respectively, for motion orthogonal and parallel to the chain axis the values of the sedimentation velocity v_n (n indicating the number of spheres in the chain) normalized by division through the velocity of a single sphere v_1. The error of v_n/v_1 is almost equal for all n-values and determined by the error of the value for one single sphere ($\simeq 1.5\%$). This error is mostly due to the fact, that the weight of the individual beads differs slightly because of the difference in the Fe_3O_4-doping. For the longer chains this error decreases at the rate of $1/\sqrt{n}$ and becomes negligible. The experimental error for v_n itself is about one percent.

The measured sedimentation velocities will now be compared with theoretical predictions. The equation of motion for a falling chain is $nm\dot{v}_n(t) + 6\pi\mu a\xi_n v_n(t) = n\frac{4\pi}{3}a^3\,\Delta\rho\, g + nF_{\mathrm{M}}$. Here v_n and ξ_n denote, respectively, the velocity and the dimensionless friction coefficient ($\xi_1 = 1$) of a chain of n spheres. m is the mass of a single sphere, a its radius, F_{M} the magnetic force exerted on it and $\Delta\rho$ the difference between the densities of the particle and the water. The time necessary for the system to reach its equilibrium (i.e. $\dot{v} = 0$) is equal to $nm/6\pi\mu a\xi_n$ and, in our case, of the order 10^{-6}s. This was of practical interest, because the chains were stopped several times during the measurement. The final velocity is $v_n .= v_n(\infty) = n\,(\frac{4\pi}{3}a^3\,\Delta\rho\, g + F_{\mathrm{M}})/6\pi\mu a\xi_n$. Normalizing the measured velocities by dividing through the velocity v_1 of a single sphere yields $v_n/v_1 = n/\xi_n$. Thus, the effect of a small magnetic field gradient cancels out and the final result can be directly compared with existing theories.

Since there are no analytical expressions for the friction coefficient of a chain of spheres with arbitrary n, we will limit ourselves to the Slender-body theory and compare the experimental results with expressions for ellipsoids and chains of spheres with a vanishing diameter-to-length ratio. Under this condition the friction coefficient of an ellipsoid and a chain of spheres can be

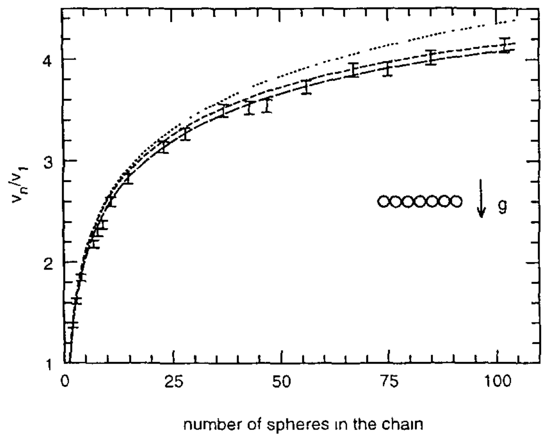

Fig. 2

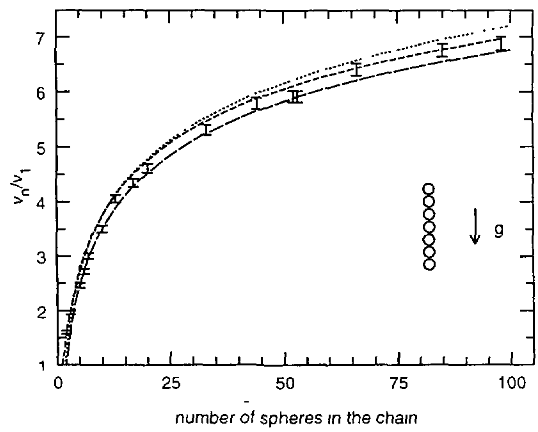

Fig. 3

Fig. 2. — Normalized sedimentation velocity v_n/v_1 for a chain of n spheres over the number n of spheres for sedimentation perpendicular to the chain axis ($v_1 = 5.8\,\mu$m/s); the dashed curves are theoretical predictions according to the Slender-body theory: (- - -) ellipsoid in an infinite volume; (– – –) ellipsoid including the effect of the container walls; (— — —) a chain of spheres with wall-effects included.

Fig. 3 — The same as in figure 2 but for sedimentation parallel to the chain axis.

expressed as [3]:

$$\xi_n^{\|} = \frac{2}{3}\frac{n}{\ln 2n - \gamma^{\|}} \qquad \xi_n^{\perp} = \frac{4}{3}\frac{n}{\ln 2n - \gamma^{\perp}} \tag{1}$$

$\xi^{\|}$ and $\xi^{\perp}$ are the friction coefficients for vertical and horizontal orientation of the body respectively. For an ellipsoid $n = l/2a$ is the ratio of the length l to the diameter $d = 2a$ and for a chain n is simply the number of spheres. For an ellipsoid the values for $\gamma^{\|}/\gamma^{\perp}$ are $+\frac{1}{2}/-\frac{1}{2}$ and for a chain of spheres Yamakawa [10] found the numerical values $+0.649/-0.418$. The curves (- - -) in figures 2 and 3 show the expressions for an ellipsoid in an infinite medium. They significantly deviate from the data, in particular at large n.

This is due to the effects of the walls of the glass cell on the sedimentation of the chains. To correct for this effect Brenner [2] gave the following expression for the ratio of the friction coefficient ξ in the presence of a boundary to the friction coefficient ξ_∞ in an infinite medium:

$$\frac{\xi}{\xi_\infty} = \frac{1}{1 - k\,\xi_\infty \frac{a}{h} + O(\frac{l}{h})^3} \tag{2}$$

h denotes the distance from the wall and k is a constant depending solely on the nature of the wall. For our setup, a body falling midway between two plane walls, we have $k = 1.004$ and h is the distance from the center of the particle to either plane. Combining equations (1) and (2) yields the final result shown as (– – – ellipsoid) and (— — — Yamakawa) in figures 2 and 3. Given our experimental error bars, both expressions agree with the measurements.

In fact, the overall agreement between the experimental values and the theoretical predictions is very good even for rather short chains. Hence, it appears that the Slender-body theory for chains of spheres gives a decent approximation even for n-values smaller than demanded by the

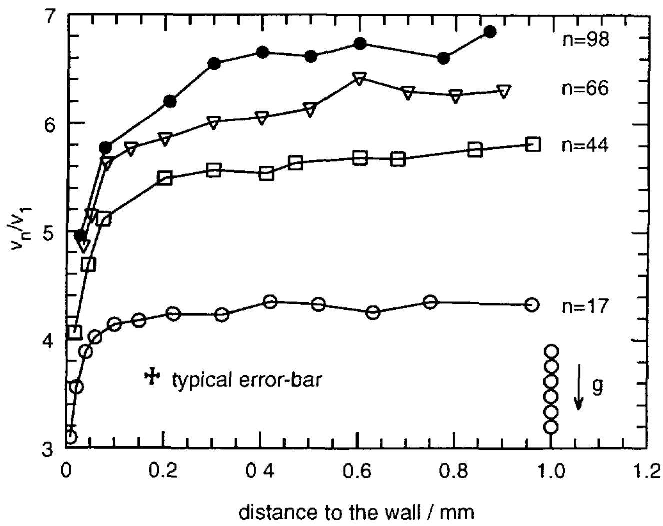

Fig. 4. — Normalized sedimentation velocity of the chains as a function of their distance to the closer wall; the measurements were performed for sedimentation parallel to the chain axis and for chains of four different lengths n (lines are guides to the eye); for comparison with the theory see [8].

range of validity of the theory $(1/(\ln n)^2 \ll 1)$. This is a well-know fact for ellipsoids, where an analytical expression for the drag coefficient is available [11], and its comparison with equation (1) yields a discrepancy of less than 1% for $n \geq 12$.

For sedimentation parallel to the chain axis the theoretical curves fit less well, but they lie well in the experimental errors. Compared with the discrepancies between theory and experiment which have been reported for this geometry [6, 12] (up to 25%), our result is completely satisfactory. The ratio of the sedimentation velocity of a chain for vertical orientation to its speed for horizontal orientation is about 7/4 for $n = 100$. This indirectly supports the well-established theoretical fact [13] that this value in an unbounded fluid approaches the limit 2 from below as $n \to \infty$. Unfortunately, the slow convergence of the series makes a closer approach to the value 2 experimentally impossible because, even with a chain of 1000 spheres, the ratio of the velocities would only be $\simeq 1.8$.

A second series of experiments was performed to investigate the interaction of the chains and the container walls more closely. Figure 4 shows the measured sedimentation velocity, for motion parallel to the chain axis for four different chain lengths as a function of the distance h to the closer wall, which could be determined by observation with the video system. A strong slowing down of the sedimentation is observed for distances smaller than the chain length. Equation (2) is no longer valid for $l/h \simeq 1$ and therefore there is no analytic expression to compare the sedimentation data near the wall. However, they turn out to be in excellent agreement with numerical simulations, as discussed separately [8].

In summary, we describe a new experimental set-up for accurate measurement of the hydrodynamic friction of individual spherical particles and rod-like chains down to very low Reynolds numbers. With the help of an applied magnetic field both position and orientation of the particles can be controlled and the chain length adjusted. Accurate measurements of velocities are presented for sedimentation parallel and orthogonal to the chain axis and as a function of the distance of the chain from the wall. The measurements of the friction coefficient of chains of spherical particles far from the wall show that the Slender-body theory is a very good approximation not only in the limit $1/(\ln n)^2 \to 0$ but even for small values of n.

The described concepts could also be applied to magnetic particles of other shapes or to diamagnetic particles suspended in paramagnetic solvents. Finally, with available high magnetic fields around 30 Tesla any kind of diamagnetic, small particles (typically below 100 μm) could be investigated.

Acknowledgments.

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