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Non-linear Stokes drag of spherical particles in a nematic solvent

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Abstract. – We present a numerical calculation for the Stokes drag of a particle in a nematic solvent. Due to the radial anchoring of the molecules at the particle surface and a uniform director field at infinity, the particle is accompanied by a hyperbolic point defect which gives rise to the so-called dipole configuration. We write the Ericksen-Leslie equation in a reduced form emphasizing the importance of the Ericksen number Er. Due to the coupling between director and velocity field, we find a highly non-linear Stokes drag. We discuss it in terms of an effective viscosity and as a function of Er. Director field and stream line patterns add to the understanding of our results. Finally, we suggest that the non-linear Stokes drag should be observable in a falling-ball experiment.

A particle of radius a moving with velocity v_0 in an isotropic fluid of shear viscosity η experiences the well-known Stokes drag force, $F_{\mathrm{S}} = 6\pi\eta a v_0$ [1]. This force serves as a starting point for dealing with such interesting phenomena as the Brownian motion [1] of dispersed particles and the long-time tail in their velocity autocorrelation functions [2] or the so-called hydrodynamic interactions between moving particles [1, 3].

One could ask the question what causes the Stokes drag to depend in a non-linear fashion on the particle velocity. Such a case occurs as soon as the non-linear convective term in the Navier-Stokes equations cannot be neglected, *i.e.*, when the Reynolds number Re, which is defined as the ratio of viscous and inertial forces in the fluid, is of the order of one [4]. A famous example is a moving rigid body which induces turbulent flow in the surrounding fluid. As a result, the drag force depends on the square of the velocity, an effect well studied in car industry. A second possibility arises from deformable bodies whose shape reacts on the flow of the surrounding fluid [5]. The behavior of "soft spheres" under the influence of shear flow ranging from liquid droplets and fluid vesicles to elastic capsules and inert ellipsoids is well-studied with the interest of modeling, *e.g.*, red-blood cells.

In this article we study the Stokes drag of a rigid spherical particle suspended in a complex fluid, which in our case is a nematic liquid crystal. In such a phase, rodlike organic molecules align on average along a common direction indicated by a unit vector $\boldsymbol{n}$ called director. Through the coupling between the director and the velocity field, the Stokes drag is driven non-linear.

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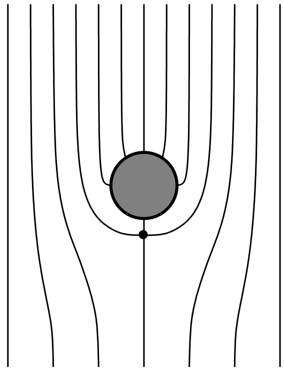

Fig. 1 – The dipole configuration of a sperical particle in a nematic solvent. At the particle surface a rigid radial anchoring of the molecules is chosen. At infinity the director points along the vertical direction. Due to topological constraints, the particle is accompanied by a hyperbolic point defect.

Stimulated by recent experiments on dispersions of particles in a nematic environment [6–8], which gave rise to the study of interesting collective phenomena [9,10], there are a number of articles addressing the director configuration around a single particle [6,11–13]. A complete list is given in ref. [14]. Here we concentrate on the dipolar configuration: a particle with rigid perpendicular anchoring of the liquid crystal molecules at its surface and uniform alignment at infinity is accompanied by a hyperbolic point defect in the director field to guarantee topological charge conservation as illustrated in fig. 1. This configuration is realized for sufficiently large particles [12,13].

The Stokes drag in liquid crystal solvents has some history whose experimental and theoretical work is reviewed in refs. [14,15]. Early experiments in nematic liquid crystals determined the temperature and pressure dependence of an effective viscosity η_{eff} in the Stokes drag [16]. Poulin *et al.* used it to measure the dipolar interaction between particles surrounded by the dipole configuration of the director field [17]. Ruhwandl and Terentjev performed numerical calculations of the Stokes drag for the Saturn-ring configuration where the particle is surrounded by a disclination ring at its equator [18]. The calculations were done for low Ericksen numbers Er, which means that the elastic deformation forces in the director field largely exceed the viscous forces in the fluid so that the director field is not affected by the flow field. This approximation considerably reduces the complexity of the problem. In the same limit, Stark and Ventzki extended the calculations to the dipole configuration and compared their results to the Saturn ring configuration and a uniform director field [15]. In all three cases the overall symmetry of the systems is uniaxial. Hence, the Stokes drag is generalized to $\boldsymbol{F}_{\mathrm{S}} = \boldsymbol{\gamma}\boldsymbol{v}_0$, where $\boldsymbol{\gamma}$ is a tensor with only two independent friction coefficients $\gamma^{\parallel}$ and $\gamma^{\perp}$ for respective motions parallel and perpendicular to the symmetry axis. Chono and Tsuji studied the velocity and director field around a cylinder for arbitrary Ericksen numbers [19]. However, for radial anchoring their director fields did not exhibit any topological defects required by the boundary conditions. Finally, Billeter and Pelcovits used molecular dynamics simulations to determine the Stokes drag of very small particles [20], and they observed that the Saturn ring is strongly deformed due to the motion of the particles.

In this article we perform a numerical study of the Stokes drag when the particle is moving along the symmetry axis of the dipolar configuration. We solve the full Ericksen-Leslie equations which describe the dynamics of a nematic liquid crystal [21,22]. We will demonstrate that the coupling between velocity and director field renders the Stokes drag highly non-linear. It crucially depends on whether the particle moves against the hyperbolic defect or leaves it behind. We will clarify the reason for such a behavior.

To calculate the Stokes drag, we consider the equivalent problem of determining the flow and director field around a particle at rest where we denote the uniform velocity at infinity by $\boldsymbol{v}_\infty$. Our problem is stationary ($\partial \boldsymbol{v}/\partial t = \partial \boldsymbol{n}/\partial t = \mathbf{0}$), we consider an imcompressible fluid ($\mathrm{div}\boldsymbol{v} = 0$), and restrict ourselves to small Reynolds numbers Re. By referring lengths and velocities to the radius a of the particle and to v_∞, respectively, the momentun balance of the Ericksen-Leslie equations reads [21, 22]

$$-\nabla_i p + \nabla_j (T^0_{ij} + Er\, T'_{ij}) = 0\,, \tag{1}$$

where ∇_i means partial derivative $\partial/\partial x_i$. The symbol T^0_{ij} stands for the rescaled elastic contribution to the stress tensor originating in deformations of the director field. It obeys the general formula, $T^0_{ij} = -(\partial f_b/\partial \nabla_j n_k)\, \nabla_i n_k$, where f_b denotes the Frank free energy of the director deformations. In the one-constant approximation ($f_b = (\nabla_i n_j)^2/2$), employed in the following, the elastic stress tensor reduces to $T^0_{ij} = -\nabla_j n_k \nabla_i n_k$. The average Frank elastic constant K, normally appearing in the Frank free energy, is subsumed into the Ericksen number [23] which is a measure for the ratio of viscous to elastic forces in the momentum balance. The quantity α_4 stands for one of the Leslie viscosities appearing in the viscous stress tensor of eq. (1),

$$T'_{ij} = \alpha_1 n_i n_j n_k n_l A_{kl} + \alpha_2 n_j N_i + \alpha_3 n_i N_j + 2A_{ij} + \alpha_5 n_j n_k A_{ik} + \alpha_6 n_i n_k A_{jk}\,. \tag{2}$$

Note that in eq. (2) the Leslie viscosities α_i are given in units of $\alpha_4/2$, which becomes the conventional shear viscosity of an isotropic liquid when all other Leslie viscosities are zero. The viscous stress tensor T'_{ij} couples the director field to the symmetrized velocity gradient, $A_{ij} = (\nabla_i v_j + \nabla_j v_i)/2$, and to the rate of change of the director relative to a fluid vortex, $\boldsymbol{N} = \partial \boldsymbol{n}/\partial t + \boldsymbol{v} \cdot \boldsymbol{\nabla} \boldsymbol{n} - \mathrm{curl}\boldsymbol{v} \times \boldsymbol{n}/2$, where in the stationary case $\partial \boldsymbol{n}/\partial t = \mathbf{0}$. Finally, the reduced hydrodynamic pressure p is given in units of K/a^2.

The second part of the Ericksen-Leslie equations, written in reduced form as

$$\boldsymbol{n} \times (\boldsymbol{h}^0 - Er\, \boldsymbol{h}') = \mathbf{0}\,, \tag{3}$$

balances the torques on the local director, where the cross product $\boldsymbol{n}\times$ ensures that $\boldsymbol{n}$ remains a unit vector. The torques consist of an elastic contribution, $h^0_i = \nabla_j(\partial f_b/\partial \nabla_j n_i) - \partial f_b/\partial n_i = \boldsymbol{\nabla}^2 n_i$, where in the second equality the one-constant approximation has been employed, and a viscous part, $h'_i = \gamma_1 N_i + \gamma_2 A_{ij} n_j$. Here, the γ_i are given in units of $\alpha_4/2$. The coefficient γ_1 denotes a true rotational viscosity of the director motion. It contributes to the viscous torque even in the stationary case whenever $\boldsymbol{\nabla}\boldsymbol{n} \neq \mathbf{0}$. Onsager relations require $\gamma_1 = \alpha_3 - \alpha_2$ and $\gamma_2 = \alpha_2 + \alpha_3 = \alpha_6 - \alpha_5$ [21, 22]. In our numerical calculations the Leslie viscosities of 5CB were used.

Finally, once the velocity and director field are known, the Stokes drag parallel to the symmetry axis of the dipole configuration is calculated with the help of the dissipated energy per unit time [18, 22, 24, 25]: $F^{\parallel}_{\mathrm{S}}\, v_\infty = \int (T'_{ij} A_{ij} + h'_i N_i)\, \mathrm{d}^3 r$. Note that a non-uniform director field contributes to the dynamic variable $\boldsymbol{N}$ through the convective term $\boldsymbol{v} \cdot \boldsymbol{\nabla}\boldsymbol{n}$ and therefore to the dissipated energy and the Stokes drag.

We add two comments. First, the divergence of the elastic stress tensor, $\nabla_j T^0_{ij}$, in eq. (1) contains the gradient of $\boldsymbol{n}$ to the third power. Close to the hyperbolic point defect in the dipole configuration of fig. 1, $\nabla_i n_j$ diverges as $1/\Delta x$, where $\Delta x \ll 1$ is the grid constant in our numerical treatment. Therefore, $\nabla_j T^0_{ij} \propto 1/\Delta x^3$ becomes very large. We observed that this rendered our numerical approach unstable. To overcome the problem, we used instead $\nabla_j T^0_{ij} = -\nabla_i f_b + h'_j(\nabla_i n_j)$, whose derivation in the static case can be found in ref. [22]. Its extension to the dynamic case is straightforward. The term $\nabla_i f_b$ just rescales the hydrodynamic pressure. The remaining term $h'_j(\nabla_i n_j)$ only diverges as $1/\Delta x^2$. After this replacement, our program was stable.

Secondly, from eq. (3) one concludes that for small Ericksen numbers the change of the director field relative to its static value scales as $\delta \boldsymbol{n} \propto Er$. This suggests the approximation that for $Er \to 0$ the director field is held fixed and only the momentum balance (1) is solved which was pursued in refs. [15, 18]. However, an inspection reveals that the change of the stress tensor T^0_{ij} due to $\delta \boldsymbol{n}$ scales as Er for an inhomogeneous director field. It is therefore of the same order as the viscous stress tensor, and strictly cannot be neglected. Nevertheless, our results in this paper support the approximation, since for $Er \to 0$ we reproduce the result from ref. [15].

We shortly summarize the numerical solution of eqs. (1) and (3) together with the incompressibility condition $\mathrm{div}\boldsymbol{v} = 0$. Details can be found in ref. [15], where the case of low Ericksen numbers is treated. We map the infinite space around a particle of reduced radius one onto the unit sphere by employing modified spherical coordinates with a radial coordinate $\xi = 1/r$, where r is the conventional radial distance. Furthermore, the velocity and director fields are expressed in the local spherical coordinate basis attached to each space point. Due to the rotational symmetry of the dipole configuration around the z-axis, all quantities do not depend on the azimuthal angle ϕ, and the director and velocity fields are situated in the symmetry planes through the z-axis. As explained in ref. [15], the incompressibility condition is treated via the method of artificial compressibility. The momentum balance equation is relaxed into its stationary solution via the Newton-Gauss-Seidel method obeying the following boundary conditions for the velocity and pressure fields. At infinity, $p = 0$ and $\boldsymbol{v}_\infty$ points along the z-axis. At the particle surface, we use the non-slip condition $\boldsymbol{v}(\rho = 1) = \boldsymbol{0}$. Due to the symmetry of our problem, the torques in eq. (3) can only point along the azimuthal direction perpendicular to the symmetry planes of the dipole configuration. Formula (3) then provides an equation for the tilt angle Θ which parametrizes the director field. It is numerically solved via a Newton-Gauss-Seidel relaxation. As starting field, we employ a static solution of eq. (3). The boundary conditions require the director field to point along the z-axis at infinity and in radial direction at the particle surface.

We now discuss our results for the Stokes drag along the symmetry axis of the dipole configuration. In fig. 2, the Ericksen number $Er \propto v_\infty$ is the abscissa coordinate, where $Er > 0$ means flow from below, and $Er < 0$ means flow from above, as indicated by the inset in fig. 2. The upper curve gives the distance r_d of the hyperbolic point defect from the center of the sphere in units of the particle radius a. When the fluid flows from above ($Er < 0$), the defect is slightly pulled towards the sphere, and the distance r_d/a changes from 1.26 at $Er = 0$ to 1.13 at $Er = -30$. On the other hand, for $Er > 0$ the defect very strongly moves away from the sphere reaching $r_\mathrm{d}/a = 3.04$ at $Er = 11$. In the lower curve, we plot the Stokes drag $F^{\|}_\mathrm{S}$ in terms of the effective viscosity $\eta^{\|}_\mathrm{eff} = F^{\|}_\mathrm{S}/(6\pi a v_\infty)$. It exhibits a corresponding behavior to r_d. For $Er < 0$, it decreases slightly from 0.48 poise at $Er = 0$ to 0.42 poise at $Er = -30$, whereas a strong increase occurs for $Er > 0$. Thus, the Stokes drag in a nematic environment is not only anisotropic, as studied in refs. [15,18], in addition it behaves strongly non-linearly. Furthermore, its magnitude crucially depends on whether the fluid first flows against the sphere ($Er < 0$) or against the point defect ($Er > 0$). Such a behavior is only possible through the coupling between the velocity and the director field. It is due to the

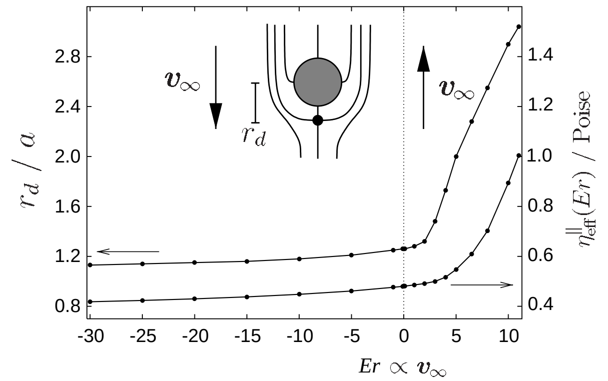

Fig. 2

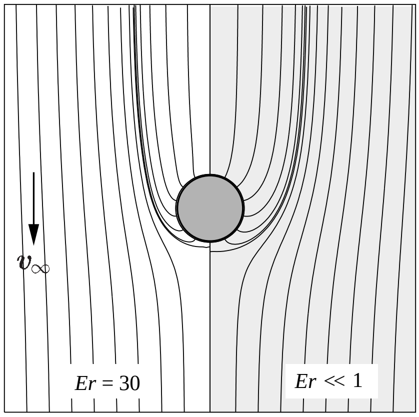

Fig. 3

Fig. 2 – The distance r_d of the hyperbolic point defect from the center of the sphere and the effective viscosity $\eta_{\mathrm{eff}}^{\parallel}$ of the Stokes drag $F_{\mathrm{S}}^{\parallel}$ as a function of the Ericksen number Er for the dipole configuration. $Er < 0$ and $Er > 0$ mean flow from above or below, respectively.

Fig. 3 – Director field patterns of the dipole configuration for the static case ($Er \ll 1$, right, shaded part) and for $Er = -30$ (left part).

fact that the torque balance equation (3) is not invariant under $\boldsymbol{v} \to -\boldsymbol{v}$. In a static dipole configuration ($Er \to 0$) it therefore cannot occur. We stress that the defect is not a solid object. Thus, the intuitive argument that the defect should be pushed against the particle for $Er > 0$ does not work here. The motion of the defect is determined by the non-trivial solution of the torque equation (3), therefore a simple explanation for the motion is not obvious.

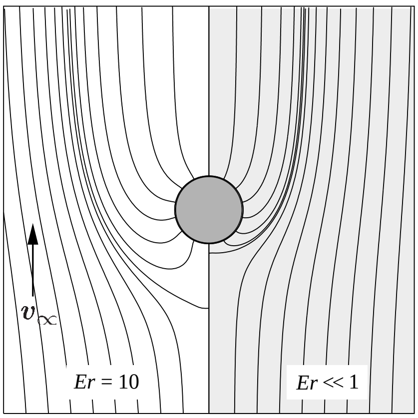

Fig. 4

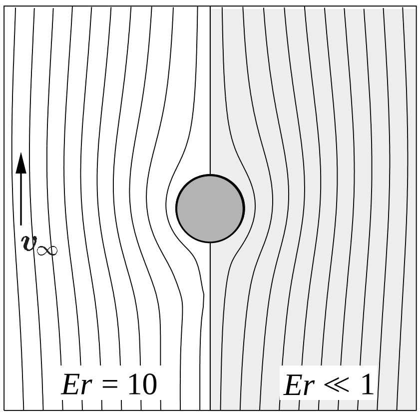

Fig. 5

Fig. 4 – Director field patterns of the dipole configuration for the static case ($Er \ll 1$, right, shaded part) and for $Er = 10$ (left part).

Fig. 5 – Stream line patterns of the dipole configuration for the static case ($Er \ll 1$, right, shaded part) and for $Er = 10$ (left part).

To improve our insight into the non-linear Stokes drag, we study the director and velocity field lines. In fig. 3 we compare the director field lines for $Er = -30$ (left part) to the static case for $Er \ll 1$ (right, shaded part). It is clearly visible that for $Er = -30$ the distance of the defect to the particle surface has decreased by a factor of two. As a result, strong director deformations occur closer to the particle. Compared to the static director field, the field lines for $Er = -30$ are straightened along the vertical direction due to flow alignment. The effective viscosity for $Er = -30$ is reduced since the volume with a non-vanishing convective term $\boldsymbol{v} \cdot \boldsymbol{\nabla n}$, which contributes to the dissipated energy through the dynamic variable $\boldsymbol{N}$, is reduced. On the other hand, when the defect moves away from the particle for $Er > 0$, the strong director distortions have to extend much further away from the particle, as illustrated in fig. 4 (left part) for $Er = 10$ preventing flow alignment close to the particle. The volume with a non-vanishing $\boldsymbol{v} \cdot \boldsymbol{\nabla n}$ increases strongly, and so does the effective viscosity. Finally, in fig. 5 we compare the velocity field lines for $Er = 10$ (left part) and $Er \ll 1$ (right, shaded part). Strong changes are only visible in the region where the defect has moved. Interestingly, the field lines first bend towards the symmetry axis when moving along the z-axis. The field lines are not completely smooth in this region which we attribute to the grid which becomes coarser with increasing distance from the sphere.

The reported non-linear Stokes drag should be observable in a falling-ball experiment [16], where the particle moves in a nematic solvent under the influence of gravity. Balancing the gravitational, the buoyancy, and Stokes's friction force, one arrives at a particle velocity of $v_0 = 2(\varrho - \varrho_{\mathrm{fl}})a^2 g/(9\eta_{\mathrm{eff}})$, where ϱ and $\varrho_{\mathrm{fl}} \approx 1\,\mathrm{g/cm^3}$ are the respective mass densities of the particle and the surrounding fluid, and g is the gravitational acceleration. For latex particles ($\varrho - \varrho_{\mathrm{fl}} = 0.01\,\mathrm{g/cm^3}$), the resulting Ericksen number is much smaller than one. However, if one takes gold particles with $\varrho = 19.3\,\mathrm{g/cm^3}$ [26], one arrives at $v_0 = 100\,\mu\mathrm{m/s}$ for particles of radius $a = 10\,\mu\mathrm{m}$ and $\eta_{\mathrm{eff}} = 0.5\,\mathrm{poise}$, which corresponds to $Er = 5$ if $K = 10^{-6}\,\mathrm{dyn}$ is employed. Variations in the particle radius then lead to the range of Ericksen numbers studied in this article. Furthermore, in recent beautiful observations of Saturn-ring configurations, Gu and Abbott used a special treatment of the gold surface of their particles to realize the required radial anchoring for the molecules [8].

To conclude, we have presented a numerical study of the Stokes drag for a particle in a nematic solvent surrounded by the dipole configuration. To implement the Ericksen-Leslie equations on the computer, we have formulated them in a scaled version which emphasizes the importance of the Ericksen number. We find a highly non-linear Stokes drag, which is the first example of such a behavior in colloidal science we know of. A falling-ball experiment is suggested to confirm our findings. It is possible that the hyperbolic defect becomes unstable for large positive Ericksen numbers and opens up to form a Saturn ring as predicted for high magnetic or electric fields [13]. However, due to a "numerical pinning" of the defect ring we are not able to verify this effect in a director theory [13]. An approach using the alignment tensor would clarify this question [27]. Further investigations are directed towards the Brownian motion for such a highly non-linear Stokes drag. Our findings might also be relevant for the interpretation of rheological studies on particle gels in a nematic solvent [9].

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