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Regular Article

Nematohydrodynamics for colloidal self-assembly and transport phenomena

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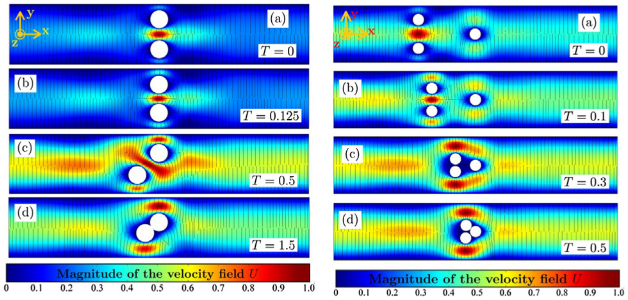
GRAPHICAL ABSTRACT

Time evolution of the multiparticle aggregate (left – dual system, right – triplet) in a nematic liquid crystal microchannel flow. Homeotropic anchoring is applied on the channel walls and particle surface. The fluid flow is in the positive *x*-direction. The solid black lines represent the director orientation in the channel and the coloured contour represent the magnitude of the axial (*x*- component) velocity field. The time and the velocity fields in the figures are dimensionless.



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ABSTRACT

Hypothesis: Colloidal particles in a nematic liquid crystal (NLC) exhibit very different behaviour to that observed in an isotropic medium. Such differences arise principally due to the nematic-induced elastic stresses exerted due to the interaction of NLC molecules with interfaces, which compete with traditional fluid viscous stresses on the particle.

Theory: A systematic mathematical analysis of particles in an NLC microfluidic channel is performed using the continuum Beris–Edwards framework coupled to the Navier–Stokes equations. We impose strong homeotropic anchoring on the channel walls and weak homeotropic anchoring on the particle surfaces.

Findings: The viscous and NLC forces act on an individual particle in opposing directions, resulting in a critical location in the channel where the particle experiences zero net force in the direction perpendicular to the flow. For multi-particle aggregation we show that the final arrangement is independent of the initial configuration, but the path towards achieving equilibrium is very different. These results uncover new mechanisms for particle separation and routes towards self-assembly.

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1. Introduction

Nematic liquid crystals (NLCs) are important examples of complex anisotropic fluids with locally preferred directions [1]. NLCs combine the intrinsic fluidity of liquids with long-range orientational ordering of the constituent rod-like molecules. The orientational order couples with the flow and induces novel effects compared with isotropic Newtonian fluids, such as backflow, anisotropic stresses and multiple viscosities. The study of NLCs in microfluidic environments is relatively new, with substantial experimental interest since around 2011. Subsequently, experimentalists have highlighted the immense potential of NLC microfluidics for transport, mixing and particle separation [2,3], while the ability of NLCs to spontaneously organize micron-size particles into regular patterns shows great promise [4]. For example, it is possible to generate defect or disclination lines in an NLC microfluidic set-up with an appropriate choice of boundary conditions, material parameters, temperature and flow effects and these defect lines can naturally attract colloidal particles or micro-cargo, which are subsequently transported along these lines as self-assembled chains [3,5]. Further, the forces facilitating spatial-reorganization of colloidal dispersions in an NLC medium are two to three orders of magnitude higher than in water-based liquids [6,7].

In the bulk NLC, additional long-range interactions between particles are present because of the competition between elasticity and the interaction between NLC molecules and surfaces (termed 'anchoring'), implying that colloids suspended in a nematic matrix are qualitatively different from their isotropic analogues. The particle sets a certain director distortion around itself, due to the surface anchoring conditions; the director distortions lead to long-range elastic interactions of the particle with the bounding walls (or neighbouring interfaces); and the nematic order leads to an anisotropy in the Stokes drag [8,9]. These features mean that rich self-ordering phenomena can be observed, which is characterized by strong interplay between the colloidal size, NLC anisotropies and particle and surface anchoring properties [3,10–12].

There is a wealth of literature on nematohydrodynamics in the absence of particle inclusions [13,14]. Studies of a particle in an NLC have generally focused on how the NLC molecules reorder around a single particle that is held in position [15] or the transitions in the flow profiles [16,17]. More recent experimental studies have focused on the dynamic behaviour of (finite sized) suspended colloidal particles in a nematic-fluid flow [1,3,7,18].

Our work is motivated by the experiments conducted by Sengupta et al. in [2]. Here the authors study an NLC microfluidic set-up experimentally and numerically in three different flow regimes – weak, medium and strong – and report on both the flow profiles and the averaged local molecular alignment profiles, referred to as 'director' profiles in the continuum-modelling literature. The surfaces of the microfluidic channel are treated to induce homeotropic boundary conditions, so that the nematic molecules are preferentially anchored along the normal to the boundary surfaces, or equivalently the continuum 'director' is parallel to the normal to the channel walls. A flow is induced by applying a pressure gradient at the inlet and the observations seem to be invariant across the width of the cell.

In this paper we focus on three separate aspects: (i) a static particle at the centre with variable anchoring strength on its boundary, (ii) the forces experienced by a particle due to hydrodynamic effects, nematic stresses and attractive forces induced by the boundary conditions, and (iii) the dynamics of two and three particles in an NLC microfluidic environment including the transient dynamics.

We mathematically model the NLC microfluidic environment using the nematodynamics formulation employed in [19]. The state of nematic alignment is described by a two-dimensional (2D) Landau–de Gennes (LdG) **Q**-tensor, which is a symmetric traceless two-by-two matrix with two degrees of freedom: an angle θ that describes the preferred in-plane alignment of the nematic molecules or the direction of the nematic director $\boldsymbol{n}$, and a scalar order parameter, s, that is a measure of the degree of alignment about the director $\boldsymbol{n}$. We investigate how the particles interact with the NLC environment in the absence and presence of flow, for both static and moving particles. The first example concerns a static particle in the NLC microfluidic cell with no fluid flow. For a given anchoring strength on the particle boundary, we study the director profile around the particle as a function of its size and, for a given particle size, we investigate the surrounding director profile as a function of anchoring strength. In both cases, there is a narrow window of parameters within which the director orientation on the particle boundary switches from uniform to normal/homeotropic and we numerically explore the switch in different cases. We then systematically study the force experienced by the particle including the effects of a flow field, particle surface anchoring, and the particle size. In particular, for a given anchoring strength and flow velocity, there is a critical particle size (relative to the channel dimensions) such that, in contrast to conventional liquids, the force attains a maximum, decreasing for larger particles owing to the attractive forces exerted by the boundaries. We conclude by studying the motion of two and three colloidal particles in the microfluidic channel, including the transient re-alignment dynamics, how the particles get attracted to each other starting from different initial configurations and are transported through the channel as an agglomerate.

2. Theory

We consider a two-dimensional NLC microfluidic channel (parallel-plate geometry) as shown in Fig. 1. The nematic director $\boldsymbol{n} = (\cos\theta, \sin\theta)$, represents the locally preferred in-plane alignment of the NLC molecules relative to the horizontal axis. We consider a circular particle, whose boundary is parameterized by the angle ϕ to the horizontal axis. We apply strong homeotropic anchoring conditions on the channel walls (modelled by Dirichlet conditions) while the anchoring conditions on the colloidal particle are varied from weak to strong in terms of an anchoring coefficient. Provided the channel dimension into the page (z direction) is large compared with the channel height (in the y direction, i.e., $2L_2$ in Fig. 1) then this two dimensional approximation is valid [20,21]. We note that the three-dimensional analogue of this two-dimensional set-up would correspond to cylindrical particles. However, similar methods can also be applied to studied to spherical colloidal particles in an NLC microfluidic channel, though this requires further study. When these dimensions are comparable then the problem is fully three dimensional, as seen in [22,23]. Whilst we do not consider this scenario in this paper, we analyse this further in Section A of the Supplementary Information. The fluid flow in the device is driven by an external pressure difference and by the nematic ordering. We impose no-slip conditions on the channel walls and particle surface.

The flow hydrodynamics are described by the incompressible Navier–Stokes equations with an additional stress ($\boldsymbol{\sigma}$) due to the NLC orientational ordering [19,24],

$$\nabla \cdot \boldsymbol{u} = 0, \tag{1}$$

$$\rho\left(\frac{\partial \boldsymbol{u}}{\partial t} + \boldsymbol{u} \cdot \nabla \boldsymbol{u}\right) = -\nabla p + \nabla \cdot (\mu[\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})'] + \boldsymbol{\sigma}). \tag{2}$$

Here $\nabla = (\partial/\partial x, \partial/\partial y)$, ρ and μ are the density and viscosity of the fluid medium respectively, p is the hydrodynamic pressure, $\boldsymbol{u}$ is the