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Silvia Carlotto [a] & Antonino Polimeno [a]

[a] Dipartimento di Scienze Chimiche, Università degli Studi di Padova, Padova, Italy

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Time-Evolution Equations for Particle Dispersions in Nematic Liquid Crystal Media

SILVIA CARLOTTO AND ANTONINO POLIMENO

Dipartimento di Scienze Chimiche, Università degli Studi di Padova,
Padova, Italy

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A formal re-definition of the hydrodynamic equations for a compressible uniaxial nematic fluid in the presence of dispersed particles is presented, which is particularly amenable to computational treatment. Starting from the Parodi-Foster formulation of nematodynamics equations, it is shown that a functional formulation of the time evolution equations can be obtained which clearly separate conservative and dissipative terms. A simplified discretization scheme is also proposed, which leads to pseudo-particle representation of the fluid in combination with a standard description of the dispersed particles motion.

Keywords Dispersions; nematic liquid crystals; non-linear fluctuation hydrodynamics

1. Introduction

Colloidal dispersions are challenging objects of study, exhibiting non-trivial physico-chemical properties, which defy current theoretical/computational treatments for simple fluids and demand in many cases a multiscale approach. In general, colloidal dispersions are made of particles with the size in a range from 10 nm to 10 μm and they are present in food, in drugs, ink, paints and cosmetics so they are of considerable technological importance. In fundamental research they are ideal systems to study Brownian motion and hydrodynamic interactions [1], transition phase [2] problems, and phenomena connected with flocculation [3].

In particular, colloidal suspensions of particles in nematic environments are of particular interest, as shown by a number of articles on the subject [4–7]. Early experiments started in 1970 when Brochard and de Gennes studied a suspension of magnetic grains in nematic [8]; other experimental groups worked on silica spheres in nematic host [9], on latex particles in lyotropic liquid crystals or water droplets [10]. Experimental measures of translation diffusion coefficients of molecular probes

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Address correspondence to Antonino Polimeno, Dipartimento di Scienze Chimiche, Università degli Studi di Padova, via Marzolo 1, 35131, Padova, Italy. E-mail: antonino.polimeno@unipd.it

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in liquid crystals were obtained using several techniques. Poulin *et al.* [10] measured translational diffusion of latex spheres with dynamic light scattering, Lettinga *et al.* [11] used a digital microscopy to directly visualize the dynamics of fluorescently labelled *fd* virus in a nematic background of unlabelled *fd* virus, Yun and Fredickson [12] measured translation diffusion of molecules tagged with ^{14}C radioactive isotopes by liquid scintillation counting in various liquid crystals, determining the parallel and perpendicular diffusion with respect to an applied magnetic field. Moseley and Lowenstein [13,14] studied the diffusive motion of methane and chloroform molecules in liquid crystals. More generally Krüger [15] discussed several experimental techniques like NMR, MTR and QENS to measure diffusion coefficients in nematic and smectic phases, together with their theoretical interpretation. Recently, Spiegel *et al.* [16] employed forced Rayleigh scattering to study the diffusion of methyl-red in 5CB.

Definition of novel experimental observables is the subject of several investigations. For instance, Poulin *et al.* [17] presented a new method to measure attractive interactions between colloidal particles and determined the nature of the attraction between particle suspended in a nematic liquid crystal. van der Beek and Lekkerkerker [18] prepared charged colloidal platelets by variation of the ionic strength for tune the effective thickness-to-diameter ratio of the platelets. Nucleation and growth of colloidal crystals [19] were considered among others by Smalyukh *et al.* [20], who used laser tweezers to study the colloidal interaction of solid microspheres in the nematic bulk caused by elastic distortions around the particles with tangential surface anchoring. The defect structures that arise between two colloidal spheres immersed in a nematic liquid crystal were studied by molecular simulation and dynamic field theory [21]. Poulin evidenced novel colloidal interactions in anisotropic fluids arising from the orientational elastic energy of the anisotropic host fluid [22] and observed novel phases and colloidal assemblies in liquid crystals in which long-range-ordering and topological constraints imposed to the solvents allow controlled organization of the particles [23].

Theoretical and computational methods for the evaluation of translational properties of suspended particles and extensive mesoscopic treatments of colloidal nematic systems are also frequently discussed in the literature. In the past several hydrodynamic and kinetic interpretations of experimental data on translation friction (or diffusion) coefficients of solutes in nematic solvents were derived. Diogo [24] found analytical expressions for the friction acting on a spherical molecule in a nematic in the presence of an external field, under severe approximations. Franklin [25] used a modification of Kirkwood theory to relate translation parallel and perpendicular diffusion coefficients of probes in nematic to viscoelastic parameters, order parameter and molecular shape, deriving also an expression for rotation diffusion coefficients, and lately [26] employed hydrodynamic theory to interpret experimental findings of diffusion coefficients of molecular probes in PAA. Khare *et al.* [27] presented a kinetic treatment to study translation diffusion in nematic fluids, and compared their results with computer simulations. Yamamoto [28] presented a method for mesoscopic simulations of particle dispersion in nematic liquid crystal solvents, which can be used in the case of charged colloidal suspensions. Stark [29] studied the director field around a spherical particle immersed in a uniformly aligned nematic liquid crystal based on the assumption of a preferred homeotropic orientation on the surface of suspended particles. Kuksenok *et al.* [30] analyzed the director distribution around a spherical colloidal particle with radial anchoring on its surface in

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the limiting cases of weak and rigid anchoring. Grollau *et al.* [31] considered the effect of confinement on the director configuration around a spherical particle. A numerical analysis using adaptive grids was made by Fukuda and Yokoyama [32] on the director configuration around a spherical particle. Ruhwandl and Terentjev [33] considered that the flow of a liquid crystal around a particle based on its shape and viscosity coefficients. He studied the resulting drag force on a sphere moving in a nematic liquid crystal. Shape size effects were investigated by Kim and de Pablo [34]. Interactions among particles were considered by Fukada *et al.* [35] and Borštnik *et al.* [36]. Fukada investigated numerically the interaction between two spherical particles in a nematic liquid crystal mediated by elastic dispersion in the orientational order. Borštnik studied the effective interaction of micro-size particles immersed in a nematic liquid crystal at temperature above the nematic-isotropic phase transition.

In this preliminary report, we aim to start defining a comprehensive approach for the computational description of colloidal suspension in nematic liquid crystalline environments, based on a modified macroscopic (nematodynamics) view. Basically, we aim to re-define, the classical equations for rigid bodies in a uniaxial nematic medium, in order to recover discretized time evolution equations convenient enough for computational purposes, in the form of a set of pseudo-particles (for the nematic medium) interacting with the dispersed particles in suspensions. The basic structure of the final set of time evolution equations is shown to be particularly amenable to direct introduction, according to standard fluctuation-dissipation arguments, of Brownian or stochastic forces. Our main goal is to show the intrinsical structure of the time-evolution equations; we do not include any explicit detailed formal derivation, which is presented elsewhere together with numerical applications.

The paper is organized as follows. In Section 2 the basic formalism for the deterministic hydrodynamic equations of dispersed micro-sized particles in a uniaxial nematic are revised. Discretization schemes are discussed in Section 3. A brief discussion is finally provided in Section 4.

2. Particle-Fluids Equations

The following notation conventions hold, unless otherwise specified: for a generic function f, $\partial f/\partial t \equiv \dot{f}$; $\mathbf{r} = (r_1, r_2, r_3)^{tr}$ is a point in space; for a generic function f_i, $f_{i,j} = \partial f_i/\partial r_j$. Einstein sum convention is employed.

Let us first consider for simplicity the case of a dispersion of particles in a isotropic compressible isothermal medium. In general, we can describe the fluid by defining four fields $q(\mathbf{r}_i)$ where $i = 0, \ldots, 3$, i.e., the mass density $\rho(\mathbf{r}) \equiv q_0(\mathbf{r})$ and the density of momentum components $g(\mathbf{r}) \equiv q_i(\mathbf{r}) = \rho(\mathbf{r})v_i(\mathbf{r})$ where $i = 1, 2, 3$ and $v_i(\mathbf{r})$ are the velocity field components. Classic hydrodynamic equations for the fluid are

$$\begin{aligned}\dot{\rho} &= -g_{j,j}\\ \dot{g}_i &= -\frac{\partial p}{\partial r_i} - \frac{\partial}{\partial r_j}\frac{g_i g_j}{\rho} - \frac{\partial \sigma_{ij}}{\partial r_j}\end{aligned} \tag{1}$$

The system free energy can be defined as

$$\mathcal{F} = \int_V d\mathbf{r}\left(\frac{\mathbf{g}^2}{2\rho} + f_\rho\right) \tag{2}$$

where the density free energy f_ρ is employed and V is the system volume; the pressure is $p = \rho\partial f_\rho/\partial\rho - f_\rho$. Finally the stress tensor is defined in terms of the bulk and shear viscosities

$$\sigma_{ij} = -2\eta\left(A_{ij} - \frac{1}{3}\delta_{ij}A_{kk}\right) - \zeta\delta_{ij}A_{kk} \tag{3}$$

and $2A_{ij} = v_{i,j} + v_{j,i}$. It is possible to write Eq. (1) in a more compact form, which can be (1) related to alternative formulations, for instance in terms of canonical Clebsch variables, (2) naturally extended to include coupling with dispersed bodies and (3) amenable to discretization schemes giving rise to a pseudo-particle formalism. By inspection, we can see that Eq. (1) can be written as

$$\dot{q}_i(\mathbf{r}) = -\frac{1}{k_BT}\int_V \hat{\xi}_{ij}(\mathbf{r},\mathbf{r}')\frac{\delta F}{\delta q_j(\mathbf{r}')} \tag{4}$$

here $\hat{\xi}_{ij}(\mathbf{r},\mathbf{r}')$ is a generalized tensor which can naturally be split into an anti-symmetric conservative term and a symmetric dissipative one; the latter being related to the stress tensor and viscosities η, ζ. In fact, one can write explicitly $\hat{\xi}_{ij} = \hat{\xi}^a_{ij} + \hat{\xi}^s_{ij}$ and the anti-symmetric components are

$$\begin{aligned}
\frac{\hat{\xi}^a_{i0}(\mathbf{r},\mathbf{r}')}{k_BT} &= -\rho(\mathbf{r})\frac{\partial\delta(\mathbf{r}-\mathbf{r}')}{\partial r'_i}\\
\frac{\hat{\xi}^a_{0j}(\mathbf{r},\mathbf{r}')}{k_BT} &= \rho(\mathbf{r})\frac{\partial\delta(\mathbf{r}-\mathbf{r}')}{\partial r_j}\\
\frac{\hat{\xi}^a_{ij}(\mathbf{r},\mathbf{r}')}{k_BT} &= -g_j(\mathbf{r})\frac{\partial\delta(\mathbf{r}-\mathbf{r}')}{\partial r'_i} + g_i(\mathbf{r}')\frac{\partial\delta(\mathbf{r}-\mathbf{r}')}{\partial r_j}
\end{aligned} \tag{5}$$

while the non-zero symmetric components are defined only for the momentum density and can be written

$$\frac{\hat{\xi}^s_{ij}(\mathbf{r},\mathbf{r}')}{k_BT} = -\frac{\partial}{\partial r_k}D_{ijkl}\frac{\partial}{\partial r_l}\delta(\mathbf{r}-\mathbf{r}') \tag{6}$$

and the Dirac delta function derivatives have meaning only in the context of (4). In the presence of N particles, which can be chosen spherical and of equal mass M for simplicity, we can write the additional equations of motions by assuming a standard classic formulation. If $\{\mathbf{R}_1,\ldots,\mathbf{R}_N\}$ and $\{\mathbf{P}_1,\ldots,\mathbf{P}_N\}$ are the positions and momenta of the dispersed particles, we simply write Hamilton equations

$$\begin{aligned}
\dot{\mathbf{R}}_\alpha &= \frac{\mathbf{P}_\alpha}{M}\\
\dot{\mathbf{P}}_\alpha &= -\frac{\partial\mathcal{F}}{\partial\mathbf{R}_\alpha}
\end{aligned} \tag{7}$$

where $\mathcal{F}$ takes the role of generalized Hamiltonian, as a function of the particles positions: terms accounting for fluid/surface interaction can be included in the free energy (see below). the integral in (2) runs over the whole solvent volume excluding the particles; α runs from 1 to N.

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Let us consider now the generalization of the previous formalism to the case of a uniaxial isothermal nematic fluid. We follow the original formulation of nematodynamics equations by Martin, Parodi, and Forster [37–39] well summarized by Pleiner and Brandt [40]. The uniaxial director is described by the unitary vector $n_i(\mathbf{r})n_i(\mathbf{r}) = 1$. Thus the fluid is described by seven coupled fields. Equations for $q_0 = \rho$ and $q_i = g_i$, $i = 1, 2, 3$, are as in the isotropic case, albeit with a much more complex expression for the stress tensor. The additional director equation, for the three additional fields $q_{i+3} = n_i$ is written in the Parodi formulation, following Pleiner and Brandt notation, as $\dot{n}_i = -v_i n_{j,i} + \mu_{ijk} v_{k,j} - \delta^{\perp}_{ij} h_j/\gamma_1$, where the flow alignment tensor is $2\mu_{ijk} = (\lambda - 1)\delta^{\perp}_{ij} n_k + (\lambda + 1)\delta^{\perp}_{ik} n_j$ and the molecular field vector is

$$h_i = \frac{\delta \mathcal{F}}{\delta n_i} \tag{8}$$

and $\delta^{\perp}_{ij} = \delta_{ij} - n_i n_j$ is the transverse Kronecker tensor. Finally the description is completed by adding an explicit expression for the free energy and the stress tensor. The free energy can assume the form (2), but now adding elastic and surface terms

$$\mathcal{F} = \int_V d\mathbf{r}\left(\frac{\mathbf{g}^2}{2\rho} + f_\rho + f_K\right) + \oint_S d\mathbf{S} f_S \tag{9}$$

The stress tensor has a considerably complex form, which can be summarized as $\sigma_{ij} = -\mu_{kji} h_k + (\partial f/\partial n_{k,j}) n_{k,i} - \upsilon_{ijkl} A_{kl}$ where υ_{ijkl} is Forster symmetric tensor

$$\begin{aligned} \upsilon_{ijkl} &= \upsilon_2\left(\delta_{jl}\delta_{ik} + \delta_{il}\delta_{jk}\right) + 2(\upsilon_1 + \upsilon_2 + \upsilon_3) n_i n_j n_k n_l \\ &\quad + (\upsilon_5 - \upsilon_4 + \upsilon_2)\left(\delta_{ij} n_k n_l + \delta_{kl} n_i n_j\right) + (\upsilon_4 - \upsilon_2)\delta_{ij}\delta_{kl} \\ &\quad + (\upsilon_3 - \upsilon_2)\left(\delta_{ik} n_j n_l + \delta_{il} n_j n_k + \delta_{jl} n_i n_k + \delta_{jk} n_i n_l\right) \end{aligned} \tag{10}$$

λ is the reactive material parameter. It is now a matter of straightforward algebra to show that the overall set of time evolution equations for q_i, $i = 0, 1, \ldots, 6$ have the form (4); moreover, the generalized friction tensor has the form (5) for the anti-symmetric terms related to fluid flow (i.e., density and momentum density), while the additional non-zero terms which couple flow and director are

$$\begin{aligned} \frac{\hat{\xi}^a_{ij+3}(\mathbf{r}, \mathbf{r}')}{k_B T} &= -\delta(\mathbf{r} - \mathbf{r}') n_{j,i}(\mathbf{r}) + \mu_{jkl}(\mathbf{r}') \frac{\partial \delta(\mathbf{r} - \mathbf{r}')}{\partial r'_k} \\ \frac{\hat{\xi}^a_{i+3j}(\mathbf{r}, \mathbf{r}')}{k_B T} &= \delta(\mathbf{r} - \mathbf{r}') n_{i,j}(\mathbf{r}') - \mu_{ikj}(\mathbf{r}) \frac{\partial \delta(\mathbf{r} - \mathbf{r}')}{\partial r_k} \end{aligned} \tag{11}$$

while the symmetric terms for the momentum density are given by (6) with $D_{ijkl} = \upsilon_{ikjl}$; finally the symmetric or dissipative term for the director field is

$$\frac{\hat{\xi}^s_{i+3j+3}(\mathbf{r}, \mathbf{r}')}{k_B T} = \frac{1}{\gamma_1} \delta(\mathbf{r} - \mathbf{r}') \delta^{\perp}_{ij}(\mathbf{r}') \tag{12}$$

Eqs. (4)–(6), (11), and (12) can now be coupled, through (9), to Eq. (7) to allow a description of dispersed particles in a nematic fluid; notice that the original number

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of seven fields can be reduced to six by exploiting the olonomic constraint $n_i(\mathbf{r})n_i(\mathbf{r}) = 1$, which is identically taken into account and can be employed to obtained a reduced set of equations for unconstrained fields.

3. Discretization Schemes

One of the main advantages of Eqs. (4)–(12) is to allow the natural implementation of convenient discretization schemes. In this section, we present some basic ideas which combine a collocation approach to spatial discretization combined with the definition of an unstructured Lagrangian grid for the fluid. The resulting set of time evolution equations for the combined grid points (pseudo-particles) can be integrated in time together with Eq. (7) for the dispersed particles, thus allowing an effective description in time of the colloidal dispersion properties.

Accordingly, we can define a generalized (unstructured) Lagrangian grid, by selecting N_g points such that $\dot{\mathbf{r}}_n = \mathbf{v}(\mathbf{r}_n, t) = \mathbf{v}_n$, with $1 \leq n \leq N_g$. Discretized equations for fields $q_i(\mathbf{r}_n, t) = q_{in}(t)$ can be written in the form

$$\dot{q}_{in} = -\frac{\xi_{injn'}}{k_B T}\frac{\partial F}{\partial q_{jn'}} \tag{13}$$

where $\xi_{injn'}$ is the discretized counterpart of kernel $\hat{\xi}_{ij}$, i.e., $\xi_{injn'} \propto \hat{\xi}_{ij}(\mathbf{r}_n, \mathbf{r}_{n'})$, and F is an approximant of the free energy calculated on the grid points, i.e., $F \propto \sum_n \left(\frac{g_n^2}{\rho_n} + f_n\right)$. Explicit forms of $\xi_{injn'}$ and F can be obtained based on proper choices for approximating numerical the volume integral in Eq. (4) and are presented elsewhere [37]. Here we stress that they can be cast in forms which resemble standard approaches for coarse grained description of fluids (e.g., dissipative particle dynamics approaches) but they are obtained from ‘first principles’, i.e., basic hydrodynamic equations. Notice that other discretization schemes can be employed, based for instance on a variational approach or finite volume treatments.

4. Conclusions

Hydrodynamics equations for compressible isothermal nematic fluids based on Poisson-like algebraic structures, can be obtained directly from the standard Parodi-Forster formulation of nematodynamics equations. The presence of dispersed particles is easily accounted for by adding classical Hamilton equations for the particles momenta and positions. Moreover, the rational form for fluid dynamics can be cast in a discretized form which is reminiscent of a “pseudo-particles” dynamics in terms of positions and velocity of fluids points. In the future, suitable numerical application will likely to profit from similar formulations, which are amenable to be integrated by standard approaches characteristic of molecular and Brownian dynamics studies.

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