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A new computational method for the solution of flow problems of microstructured fluids. Part 1. Theory

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The theoretical basis for a new computational method is presented for the solution of flow problems of microstructured fluids: examples include suspensions of rigid particles and polymeric liquids ranging from liquid crystals to concentrated solutions or melts of flexible chains. The method is based on a Lagrangian conservation statement for the distribution function of the conformation of the local structure, which can be derived from the conventional, Eulerian conservation statement and is exactly equivalent. The major difference, which is reflected in the numerical technique, is that the Lagrangian representation of the distribution function allows for computation of the Brownian contribution and of moments of distribution functions in ways that do not require explicit knowledge of the distribution function, and involve no approximation whatsoever. This suggests a new type of efficient, self-adaptive numerical scheme that is suitable for the solution of flow problems of microstructured fluids, in which macroscopic properties depend on the state of the microstructure.

1. Introduction

The flow of fluids with complex local structure (i.e. suspensions of particles, droplets, models of polymer molecules, polymer melts, etc.) has received much attention due to the technological importance of the subject. Drag reduction in dilute polymer solutions, emulsification, and the dynamics of filler particles in a flowing composite material are all consequences of the stretching and orienting effect which the flow has on microstructure.

Constitutive theories for these materials, based on a microstructural approach, attempt to relate the local stress in the fluid to the statistical distribution of stretch and orientation of the local structure. Macroscopic properties are generally determined by some moment(s) of the distribution function for microstructure conformation. A statistical description in terms of a distribution function is necessary in order to account for (i) a range of different possible initial states of the microstructure, and (ii) the randomizing effects of Brownian diffusion, when appropriate.

1.1. *The conformation distribution function*

The analysis of the mechanics of microstructured fluids normally begins with the development of the microdynamical equation for the state of a single microstructural element. The stretching and orienting tendencies of the surrounding flow must be

accounted for. Other deterministic effects such as internal viscous or elastic response must also be included. Finally, it is also possible to include Brownian diffusion when this is known to be important.

Microdynamical equations have been derived by many researchers, for many types of microstructure. In this paper, we shall fix the discussion by assuming that the complex fluid is a suspension with a microstructure that consists of uniformly distributed 'particles', whose instantaneous conformation can be described by a single (state) vector. Further, we assume that the inertia of the particles is unimportant to the dynamical response. In §4, we discuss a wide range of microstructured fluids to which the methods we develop may be applied.

Thus, for the purposes of the general discussion, we shall assume that the state vector of a particular microstructural element ($\boldsymbol{R}$) evolves according to an equation of the form

$$\dot{\boldsymbol{R}} = \mathscr{G}(\boldsymbol{R}; \boldsymbol{E}(t), \boldsymbol{\Omega}(t), \mu) - \nu \nabla(\log \phi). \tag{1.1}$$

In (1.1), the deterministic effects (of the surrounding flow, of the internal viscosity or elasticity, etc.) are lumped together in the first term $\mathscr{G}$, which might involve (smooth) nonlinear dependence on the director $\boldsymbol{R}$, and also depends on the rate-of-strain tensor $\boldsymbol{E}(t)$, the vorticity tensor $\boldsymbol{\Omega}(t)$, and on various micromechanical parameters $\mu = (\mu_1, \ldots, \mu_k)$. The Brownian diffusion is accounted for through the last term, which involves the diffusivity ν and the distribution function for the conformation of the microstructure, ϕ, to which we next turn our attention. Note that the Brownian term involves a gradient operator; we emphasize that this is an operator in *conformation space*. Furthermore, since (1.1) applies to a specific microstructural element, the time derivative is Lagrangian.

A number of elements of the microstructure reside in any small material volume of the suspension; we shall refer to the set of such elements as an *ensemble*. The state of an ensemble is conveniently described by a distribution function. These ideas are carefully presented in the present context by Bird *et al.* (1987). The distribution function specifies the probability that an element of the microstructure in the ensemble will have a particular orientation, stretched length, rotation rate and stretching rate. However, as discussed in Bird *et al.*, one normally approximates the rotation rate and stretching as having a Maxwellian distribution about the mean, i.e. as having the same distribution as if the flow system were in equilibrium. One is then left with a distribution function for conformation alone, which is called the conformation distribution function (CDF). The Brownian term in (1.1) is the velocity–space average of a randomly fluctuating Brownian force. Thus (1.1) really describes the dynamics of a velocity–space averaged element of microstructure. This point is discussed in detail by Bird *et al.* (1987), and also in §3.2, below.

The evolution of the CDF involves a competition between (i) the deterministic stretching and orienting tendencies of the flow and the internal micromechanics of the particle, which favour certain conformations over others, and (ii) Brownian diffusion, which favours a *relaxed* state. If the microstructure is composed of rigid particles, then the relaxed state corresponds simply to an isotropic distribution of orientations. If the microstructure is composed of stretchable as well as orientable particles (e.g. an elastic dumbbell model for a polymer molecule in solution) then the relaxed state is characterized by isotropy in orientation, and by a smooth equilibrium distribution of particle lengths that depends on the details of the internal micromechanics.

Mathematically, the evolution of the CDF is described by the Fokker–Planck (or

forward Kolmogorov) equation, which is a convection–diffusion equation over conformation space,

$$\frac{\partial \phi}{\partial t} + \nabla \cdot [\phi \dot{\boldsymbol{R}}] = 0. \tag{1.2a}$$

For the microdynamical equation (1.1), this specializes to

$$\frac{\partial \phi}{\partial t} + \nabla \cdot [\phi \mathscr{G}(\boldsymbol{R}; \boldsymbol{E}(t), \boldsymbol{\Omega}(t), \mu)] = \nu \nabla^2 \phi. \tag{1.2b}$$

We remind the reader that both the Laplace and gradient operators in (1.2) are operators in conformation space, while the time derivative is Lagrangian, following a specific material point through physical space.

In summary, understanding the dynamical evolution of the conformation of a suspension is a difficult problem. Associated with each material point in the flow is a distribution function that evolves according to the Fokker–Planck equation. Solution strategies for a large variety of physical problems involving the Fokker–Planck equation are reviewed by Risken (1989). In the mechanics of microstructured fluids, however, there is the additional complicating factor that the local structure has an effect on the macroscopic properties of the flow!

1.2. *The macroscopic stress*

To complete our description of the macroscopic properties of complex fluids, we require a relationship between the local state of stress in the suspension and the conformation of the microstructure. Constitutive theories have been developed for many microstructured fluids. These relate the state of stress at a point in physical space to the CDF, normally through *moments* of the distribution function.

An example is the well-known constitutive relation of Giesekus (1962) for linear dumbbell models of dilute polymer solutions. This constitutive relation may be written as a decomposition of the stress tensor into a Newtonian part and a non-Newtonian part:

$$\boldsymbol{\tau} = \boldsymbol{\tau}^{\mathrm{N}} + \boldsymbol{\tau}^{\mathrm{NN}}. \tag{1.3a}$$

The Newtonian part is

$$\boldsymbol{\tau}^{\mathrm{N}} = -p\boldsymbol{I} + 2\eta \boldsymbol{E}, \quad \boldsymbol{E} \equiv \tfrac{1}{2}[(\nabla \boldsymbol{u}) + (\nabla \boldsymbol{u})^{\mathrm{t}}], \tag{1.3b}$$

where p is the pressure and η is the (Newtonian) solvent viscosity, while the non-Newtonian part of the linear dumbbell model is

$$\boldsymbol{\tau}^{\mathrm{NN}} = \alpha \langle \boldsymbol{RR} \rangle, \tag{1.3c}$$

where α is a constant. The second moment $\langle \boldsymbol{RR} \rangle$ depends on the distribution function through the definition,

$$\langle \boldsymbol{RR} \rangle(t) \equiv \int \phi(\boldsymbol{R}, t)\, (\boldsymbol{R} \otimes \boldsymbol{R})\, \mathrm{d}\boldsymbol{R}. \tag{1.4}$$

Note that this moment function associated with a fixed material point contains only the Lagrangian dependence on time.

Thus, it should be clear that the solution of a flow problem for a microstructured fluid is difficult, involving the effect of the flow on the local structure and the coupled effect of the local structure on the flow. The suspension must satisfy the macroscopic balances of mass and momentum. A part of the momentum equation is the

divergence of the non-Newtonian stress, which requires calculation of one or more moment(s) of the distribution function associated to each material point. Generally, solutions of the flow problem can only be achieved numerically. The principal practical difficulty is that one must compute the distribution function for each material point.

To our knowledge, there are two techniques that eliminate the need to compute the distribution function in quite general flow problems. These are: (i) the solution of a direct moment evolution equation instead of a Fokker–Planck equation, and (ii) the double-Lagrangian technique we develop in the present work. In order to motivate the latter development, we first describe the technique of solution of direct moment evolution equations; this is a method with considerable shortcomings, as we show below.

1.3. *Direct moment evolution equations*

First we demonstrate by example how one derives a direct moment evolution equation. Then we discuss the solution of flow problems with this approach, which has been used in a number of recent studies including Lipscomb *et al.* (1988), Chilcott & Rallison (1988) and Rosenberg, Denn & Keunings (1990).

Because a moment of the distribution function associated with a material point depends only on time, its evolution in a Lagrangian framework is governed by an ordinary differential equation. As a simple demonstration, consider the example of a dilute suspension of rigid particles unaffected by Brownian diffusion. It is well known that an isolated, rigid axisymmetric particle rotates in a flow according to the equation

$$\dot{\boldsymbol{R}} = \boldsymbol{\kappa}\cdot\boldsymbol{R} - \boldsymbol{\kappa}:\boldsymbol{RRR}, \quad \boldsymbol{\kappa} = \boldsymbol{\Omega} + G\boldsymbol{E},$$

which involves an *equivalent* velocity gradient tensor, $\boldsymbol{\kappa}$, defined in terms of a shape factor G and the vorticity and rate-of-strain tensors $\boldsymbol{\Omega}$ and $\boldsymbol{E}$, respectively. The shape factor G lies (normally) between the extremes 0 (spherical particle) and 1 (infinite-aspect-ratio fibre). The orientation distribution function (ODF) f associated with a material point of the suspension evolves according to (1.2*c*) with ϕ replaced by f. For clarity of presentation, we shall use the symbol ϕ to denote the distribution function for stretchable microstructure, and f to denote the distribution function for rigid microstructure. The second moment of the orientation function $\langle \boldsymbol{RR}\rangle(t)$ is defined by (1.4) with ϕ replaced by f. Moment evolution equations are normally derived by taking the Fokker-Planck evolution for the distribution function, multiplying by $\boldsymbol{RR}$ (or $\boldsymbol{RRRR}$, or etc.), and integrating over conformation space. The result is an ordinary differential equation for the moment tensor associated with a material point of suspension. The derivation of the moment evolution equation in the present case yields

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \boldsymbol{RR}\rangle(t) = \boldsymbol{\kappa}\cdot\langle \boldsymbol{RR}\rangle + \langle \boldsymbol{RR}\rangle\cdot\boldsymbol{\kappa}^{\mathrm{T}} - \boldsymbol{\kappa}:\langle \boldsymbol{RRRR}\rangle. \tag{1.5a}$$

We emphasize that this equation describes the evolution of the second-moment tensor associated with a fixed material point of suspension; thus it must be solved simultaneously with the particle-path equation. However, it is more common to consider an Eulerian (field) description of the moment tensor; for this purpose, we replace the full time derivative following a fixed material point by a convective derivative, assuming that the microstructure moves with the fluid. This yields

$$\frac{\mathrm{D}}{\mathrm{D}t}\langle \boldsymbol{RR}\rangle(t) = \boldsymbol{\kappa}\cdot\langle \boldsymbol{RR}\rangle + \langle \boldsymbol{RR}\rangle\cdot\boldsymbol{\kappa}^{\mathrm{T}} - \boldsymbol{\kappa}:\langle \boldsymbol{RRRR}\rangle. \tag{1.5b}$$

where the moments may now be interpreted as *tensor fields* over the spatial domain. By such a tensor field representation of the moment, one avoids the need to solve (explicitly) for particle paths in the flow.

In order to solve a flow problem, one must have some means of accounting for the particle stress contribution in the macroscopic momentum balance, which is coupled with the solution of (1.5*a*) or (1.5*b*) at each point of space. There are important problems which may arise when one attempts to solve flow problems using a direct moment evolution equation approach. These are (i) the closure problem, and (ii) the possibility of steep spatial gradients or even discontinuities in the moment tensor field obtained from (1.5*b*). We shall discuss each in turn.

First, we discuss the closure problem. Note that in the present example, the evolution equation for the second moment ($\langle \boldsymbol{RR} \rangle$) (1.5*a* or *b*) involves the fourth moment. It is possible to derive an evolution equation for the fourth moment by the same procedure; however, the resulting evolution equation involves the sixth moment. Thus the system of moment evolution equations can never be closed, as higher-order moments are always required.

This closure problem is the typical situation in most microstructured fluids, but there are exceptions. For example, the second moment evolution equation is closed for the linear elastic dumbbell model of dilute polymer solutions that was considered earlier. The reason is that the dumbbell is assumed to rotate with the flow in an affine manner, and the elastic modulus of the dumbbell is modelled as linear. Thus the equation for $\mathrm{d}\boldsymbol{R}/\mathrm{d}t$ is linear in $\boldsymbol{R}$, leading only to second moments when one derives an evolution for $\langle \boldsymbol{RR} \rangle$.

In the usual case, where the moment evolution equations are not closed, some sort of approximation for higher-order moments is required if one wants to determine moments by integration of a *closed* set of moment evolution equations. Such approximations are referred to as *closure approximations*; the standard trick is to write higher-order moments in terms of lower-order moments. Two examples of this sort of *ad hoc* approximation are linear (see Hinch & Leal 1975, 1976)

$$\langle \boldsymbol{RRRR} \rangle (t) = \boldsymbol{A} + \boldsymbol{B} \langle \boldsymbol{RR} \rangle (t),$$

where $\boldsymbol{A}$ and $\boldsymbol{B}$ are constant tensors, and quadratic (see also Lipscomb *et al.* 1988 and Rosenberg *et al.* 1990):

$$\langle \boldsymbol{RRRR} \rangle (t) = [\langle \boldsymbol{RR} \rangle \langle \boldsymbol{RR} \rangle](t).$$

Both approximations lead to a closed set of equations, but both suffer from pathological behaviour even in very simple flow situations. The linear approximation has been shown to diverge in simple shear flows of suspensions of rigid particles with moderate to large aspect ratios by both Frattini & Fuller (1986) and Advani & Tucker (1987). The quadratic approximation fails to retain the tensorial symmetry of $\langle \boldsymbol{RRRR} \rangle$. Moreover, Altan *et al.* (1989) observe quite considerable quantitative discrepancies between these two closure approximations and the exact results obtained via the distribution function in simple shear, planar extensional and uniaxial extensional flows. Finally, in part 2 of this series (Szeri & Leal 1992), we report numerical results that demonstrate that some commonly employed closure approximations can lead to non-physical behaviour. It thus appears that the case for direct solution of moment evolution equations with closure approximations is weak, at best, even in the few very simple flows in which one can test the approximations.

The second problem associated with the solution of equations of the type (1.5*b*) is of a more practical nature. Specifically, if one considers a tensor field representation

of the moment, there may be very serious problems with spatial resolution of the tensor field, depending on the details of the flow situation. This subject has been investigated in detail by El-Kareh & Leal (1989). If the flow domain is simply connected, and if the initial moment field is smooth, then it is reasonable to expect that this smoothness will last indefinitely. However, if neighbouring material points have very different histories, e.g. they passed on either side of a submerged solid body, they may develop very different moments. In this way, one might imagine that a spatial discontinuity in the moment field could develop. Such a discontinuity would wreak havoc on standard numerical methods which assume continuity of the dependent variable so that convective derivatives such as the one in (1.5*b*) can be computed.

For the special problem of a suspension of finitely extensible nonlinear elastic dumbbells, El-Kareh & Leal showed that the correct moment (field) evolution equation must include a spatial diffusion term, which would heal any discontinuities that might otherwise develop. However, the corresponding diffusion constant is extremely small; thus very large gradients could still exist in an Eulerian field description, even with the addition of the spatial diffusion term.

In summary, the direct solution of moment evolution equations has the ambitious goal of eliminating the need to compute the distribution function associated with each material point of suspension. However, two important problems have arisen when pursuing this approach: (i) the closure problem, which depends on the details of the microdynamical equation, and (ii) the resolution of discontinuities or of large spatial gradients in an Eulerian field description, which depends on the details of the flow situation.

1.4. *The double-Lagrangian technique*

In the present paper, we shall develop the theory behind a new technique for the solution of flow problems for microstructured fluids. Like the approach of direct calculation of moments, our goal is to eliminate the need to compute the distribution function. We pursue this goal by rearranging the governing equations of the problem. This is accomplished by exploiting a doubly Lagrangian representation of the distribution function: the representation is Lagrangian in physical space because we follow material points of the suspension; as we shall see, the representation is also Lagrangian in *conformation space*. We shall demonstrate that this doubly Lagrangian point of view allows for an exact simplification of Brownian diffusion terms and of moment calculations. Moreover, a fast and efficient numerical scheme is naturally developed.

1.5. *Plan of the paper*

In §2 we derive an alternative representation of distribution functions that is Lagrangian in conformation space. In §3, we show how the Lagrangian representation of distribution functions can simplify: (i) the calculation of moments of the distribution function, and (ii) Brownian diffusion. We then discuss a numerical scheme for the solution of flow problems, and, in §4, show how it may be applied to different types of microstructured fluids. In §5, we give our conclusions.

Historically, our analysis stems from a recent theoretical study of the dynamics of a model microstructured fluid that is unaffected by Brownian diffusion (Szeri, Wiggins & Leal 1991, referred to hereinafter as SWL). In the Appendix, we briefly review the analysis of SWL and apply the idea of the present paper to give an analytic example of a Lagrangian representation of a distribution function. The example serves to expose the connection between the Eulerian and Lagrangian

points of view of conformation space. In part 2 of this series (Szeri & Leal 1992), we compute an example flow of a suspension of rigid, orientable particles using the techniques we develop here in part 1. The suspension is driven by a pressure gradient to flow between parallel flat plates.

2. Lagrangian representation of distribution functions in conformation space

The mathematical description of the state of the microstructure in terms of a distribution function is already *Lagrangian* in *physical* space because the distribution function is associated with a material point of the suspension. However, the usual representation of the distribution function is *Eulerian* in *conformation* space, i.e. the distribution function is represented as a scalar field that evolves in time and in 'position' in conformation space. In this section we introduce an alternative representation of the distribution function that is Lagrangian in conformation space. The result is a 'double-Lagrangian' representation that is (i) associated with a material point as it moves with the flow, and (ii) associated with a specific 'material' point in conformation space as it evolves according to the microdynamical equations.

The Lagrangian representation is easily approached from the Eulerian via the definition of ϕ^*, the Lagrangian representation of the distribution function:

$$\phi^*(t; \boldsymbol{R}_0) \equiv \phi(t, \boldsymbol{R})\,|_{\boldsymbol{R}=\hat{\boldsymbol{R}}(t;\boldsymbol{R}_0)}, \tag{2.1}$$

where $\hat{\boldsymbol{R}}(t; \boldsymbol{R}_0)$ is the solution of the associated microdynamical equation (1.1) with initial condition $\boldsymbol{R}_0$. Additionally, $\hat{\boldsymbol{R}}(t; \boldsymbol{R}_0)$ may be interpreted as the coordinate map between the initial (or reference) configuration of the local structure at time $t = 0$ and the deformed configuration at time t. The time derivative of ϕ^* is easily computed from the definition (2.1) and from the evolution equation for ϕ, equation (1.2a):

$$\begin{aligned}\frac{\partial}{\partial t}\phi^*(t; \boldsymbol{R}_0) &= \left[\frac{\partial \phi}{\partial t} + \nabla\phi \cdot \dot{\boldsymbol{R}}\right]\Bigg|_{\boldsymbol{R}=\hat{\boldsymbol{R}}(t;\boldsymbol{R}_0)} \\ &= [-\phi \nabla \cdot \dot{\boldsymbol{R}}]\,|_{\boldsymbol{R}=\hat{\boldsymbol{R}}(t;\boldsymbol{R}_0)} = -\phi^*(t; \boldsymbol{R}_0)\,[\nabla \cdot \dot{\boldsymbol{R}}]\,|_{\boldsymbol{R}=\hat{\boldsymbol{R}}(t;\boldsymbol{R}_0)}.\end{aligned}$$

This equation may be rearranged to read

$$\frac{1}{\phi^*(t; \boldsymbol{R}_0)}\frac{\partial}{\partial t}\phi^*(t; \boldsymbol{R}_0) = -[\nabla \cdot \dot{\boldsymbol{R}}]\,|_{\boldsymbol{R}=\hat{\boldsymbol{R}}(t;\boldsymbol{R}_0)}. \tag{2.2}$$

Now let us examine the right-hand side of (2.2). The divergence may be written as the trace of the derivative, hence

$$\nabla \cdot \dot{\boldsymbol{R}} = \mathrm{tr}\,[\nabla \dot{\boldsymbol{R}}]. \tag{2.3}$$

Next we multiply the term in square brackets by the *deformation gradient tensor* and its inverse

$$\nabla \cdot \dot{\boldsymbol{R}} = \mathrm{tr}\,[\nabla \dot{\boldsymbol{R}} (\nabla_0 \boldsymbol{R})\,(\nabla_0 \boldsymbol{R})^{-1}]. \tag{2.4}$$

Here ∇_0 is to be interpreted as the derivative with respect to the reference configuration. By use of the chain rule, (2.4) may be rewritten as

$$\nabla \cdot \dot{\boldsymbol{R}} = \mathrm{tr}\left[\frac{\partial}{\partial t}(\nabla_0 \boldsymbol{R})\,(\nabla_0 \boldsymbol{R})^{-1}\right]. \tag{2.5}$$

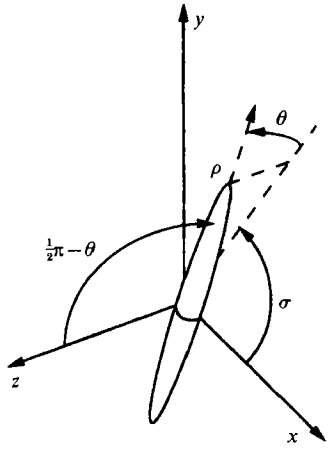

FIGURE 1. Definition sketch for the modified spherical coordinates used to describe the state of a microstructural element.

Finally, one can compute the time derivative of the determinant of the deformation gradient tensor to show that (2.5) is equivalent to

$$\nabla\cdot\dot{\boldsymbol{R}} = \frac{1}{\det(\nabla_0 \boldsymbol{R})}\frac{\partial}{\partial t}\det(\nabla_0 \boldsymbol{R}). \tag{2.6}$$

Thus, (2.2) may be rewritten

$$\frac{1}{\phi^*(t;\boldsymbol{R}_0)}\frac{\partial}{\partial t}\phi^*(t;\boldsymbol{R}_0) = -\left[\frac{1}{\det(\nabla_0 \boldsymbol{R})}\frac{\partial}{\partial t}\det(\nabla_0 \boldsymbol{R})\right]\Bigg|_{\boldsymbol{R}=\hat{\boldsymbol{R}}(t;\boldsymbol{R}_0)}.$$

This equation may be integrated to yield

$$\frac{\phi^*(t;\boldsymbol{R}_0)}{\phi^*(0;\boldsymbol{R}_0)} = \frac{1}{\det(\nabla_0 \hat{\boldsymbol{R}}(t;\boldsymbol{R}_0))}. \tag{2.7}$$

Thus, we have computed the Lagrangian form of the distribution function in terms of the coordinate map from reference to deformed configurations of the local structure.

In order to solve flow problems, it is convenient to work in terms of coordinates. It will be necessary to describe both the reference and deformed configurations using coordinates. We use *modified* spherical polar coordinates for conformation space (ρ, σ, θ) with basis vectors $(\boldsymbol{e}_\rho, \boldsymbol{e}_\sigma, \boldsymbol{e}_\theta)$. These coordinates (for the deformed configuration) are defined in figure 1. As shown in this figure, σ is an angle in the (x, y)-plane, measured counterclockwise from the x-axis. The angle measured from the z-axis is $\frac{1}{2}\pi-\theta$, and the radial coordinate is ρ. Note that when $\theta = 0$, the axial vector of the particle lies in the (x, y)-plane (this choice is convenient for the analysis of two-dimensional flows). The rectangular components of the axial vector are

$$\boldsymbol{R} = \begin{pmatrix} \rho\cos\theta\cos\sigma \\ \rho\cos\theta\sin\sigma \\ \rho\sin\theta \end{pmatrix},$$

In the modified spherical polar coordinates, the director of an element of microstructure is $\boldsymbol{R} = \rho \boldsymbol{e}_\rho$; thus

$$\dot{\boldsymbol{R}} = \dot{\rho}\boldsymbol{e}_\rho + \rho\dot{\sigma}\cos\theta\boldsymbol{e}_\sigma + \rho\dot{\theta}\boldsymbol{e}_\theta.$$

The gradient operator in conformation space is

$$\boldsymbol{\nabla} = \boldsymbol{e}_\rho\frac{\partial}{\partial\rho} + \frac{\boldsymbol{e}_\sigma}{\rho\cos\theta}\frac{\partial}{\partial\sigma} + \frac{\boldsymbol{e}_\theta}{\rho}\frac{\partial}{\partial\theta}.$$

Thus, the general microdynamical equation (1.1) may be written in component form as

$$\dot{\sigma} = \mathscr{G}_\sigma(\rho,\sigma,\theta;\boldsymbol{E}(t),\boldsymbol{\Omega}(t),\mu) - \frac{\nu}{\rho^2\cos^2\theta}\frac{\partial}{\partial\sigma}(\log\phi), \tag{2.8a}$$

$$\dot{\theta} = \mathscr{G}_\theta(\rho,\sigma,\theta;\boldsymbol{E}(t),\boldsymbol{\Omega}(t),\mu) - \frac{\nu}{\rho^2}\frac{\partial}{\partial\theta}(\log\phi), \tag{2.8b}$$

$$\dot{\rho} = \mathscr{G}_\rho(\rho,\sigma,\theta;\boldsymbol{E}(t),\boldsymbol{\Omega}(t),\mu) - \nu\frac{\partial}{\partial\rho}(\log\phi). \tag{2.8c}$$

Finally, the initial conformation $\boldsymbol{R}_0$ may be represented as $(\rho_0,\sigma_0,\theta_0)$, and the solution to (2.8) as $(\rho(t;\rho_0,\sigma_0,\theta_0),\ \sigma(t;\rho_0,\sigma_0,\theta_0),\ \theta(t;\rho_0,\sigma_0,\theta_0))$.

In order to describe the initial (reference) configuration, we use a similar coordinate system, although now we denote the coordinates $(\rho_0,\sigma_0,\theta_0)$, and the basis vectors are $(\boldsymbol{\varepsilon}_\rho,\boldsymbol{\varepsilon}_\sigma,\boldsymbol{\varepsilon}_\theta)$. The gradient operator with respect to reference-configuration coordinates is

$$\boldsymbol{\nabla}_0 = \boldsymbol{\varepsilon}_\rho\frac{\partial}{\partial\rho_0} + \frac{\boldsymbol{\varepsilon}_\sigma}{\rho_0\cos\theta_0}\frac{\partial}{\partial\sigma_0} + \frac{\boldsymbol{\varepsilon}_\theta}{\rho_0}\frac{\partial}{\partial\theta_0}.$$

The deformation gradient tensor in conformation space is easily computed:

$$\begin{aligned}\boldsymbol{\nabla}_0\boldsymbol{R} = {}& \boldsymbol{\varepsilon}_\rho\boldsymbol{e}_\rho\frac{\partial\rho}{\partial\rho_0} + \boldsymbol{\varepsilon}_\rho\boldsymbol{e}_\sigma\rho\cos\theta\frac{\partial\sigma}{\partial\rho_0} + \boldsymbol{\varepsilon}_\rho\boldsymbol{e}_\theta\rho\frac{\partial\theta}{\partial\rho_0}\\ & + \boldsymbol{\varepsilon}_\sigma\boldsymbol{e}_\rho\frac{1}{\rho_0\cos\theta_0}\frac{\partial\rho}{\partial\sigma_0} + \boldsymbol{\varepsilon}_\sigma\boldsymbol{e}_\sigma\frac{\rho\cos\theta}{\rho_0\cos\theta_0}\frac{\partial\sigma}{\partial\sigma_0} + \boldsymbol{\varepsilon}_\sigma\boldsymbol{e}_\theta\frac{\rho}{\rho_0\cos\theta_0}\frac{\partial\theta}{\partial\sigma_0}\\ & + \boldsymbol{\varepsilon}_\theta\boldsymbol{e}_\rho\frac{1}{\rho_0}\frac{\partial\rho}{\partial\theta_0} + \boldsymbol{\varepsilon}_\theta\boldsymbol{e}_\sigma\frac{\rho\cos\theta}{\rho_0}\frac{\partial\sigma}{\partial\theta_0} + \boldsymbol{\varepsilon}_\theta\boldsymbol{e}_\theta\frac{\rho}{\rho_0}\frac{\partial\theta}{\partial\theta_0}\end{aligned}$$

which has determinant

$$\det(\boldsymbol{\nabla}_0\boldsymbol{R}) = \frac{\rho^2\cos\theta}{\rho_0^2\cos\theta_0}\frac{\partial(\rho,\sigma,\theta)}{\partial(\rho_0,\sigma_0,\theta_0)}. \tag{2.9}$$

In (2.9), we have used the standard notation for the Jacobian of a transformation,

$$\frac{\partial(\rho,\sigma,\theta)}{\partial(\rho_0,\sigma_0,\theta_0)} \equiv J_3 = \det\begin{bmatrix}\dfrac{\partial\rho}{\partial\rho_0} & \dfrac{\partial\sigma}{\partial\rho_0} & \dfrac{\partial\theta}{\partial\rho_0}\\ \dfrac{\partial\rho}{\partial\sigma_0} & \dfrac{\partial\sigma}{\partial\sigma_0} & \dfrac{\partial\theta}{\partial\sigma_0}\\ \dfrac{\partial\rho}{\partial\theta_0} & \dfrac{\partial\sigma}{\partial\theta_0} & \dfrac{\partial\theta}{\partial\theta_0}\end{bmatrix}.$$

Thus the Lagrangian form of the distribution function may be written

$$\frac{\phi^*(t;\rho_0,\sigma_0,\theta_0)}{\phi^*(0;\rho_0,\sigma_0,\theta_0)} = \left[\frac{\partial(\rho,\sigma,\theta)}{\partial(\rho_0,\sigma_0,\theta_0)}\frac{\rho^2}{\rho_0^2}\frac{\cos\theta}{\cos\theta_0}\right]^{-1}. \tag{2.10}$$

In summary, the distribution function attached to a specific conformation time trace ϕ^* evolves in a way that is completely determined by the *map* from the *reference* configuration coordinates $(\rho_0,\sigma_0,\theta_0)$ to the *deformed* configuration coordinates $(\rho(t;\rho_0,\sigma_0,\theta_0), \sigma(t;\rho_0,\sigma_0,\theta_0), \theta(t;\rho_0,\sigma_0,\theta_0))$. The distribution function is related to a deformation gradient in conformation space.

Of course, the coordinate change from (ρ,σ,θ) to $(\rho_0,\sigma_0,\theta_0)$ is possible only when the determinant $J_3 \neq 0$. As we shall see in §3.2, it is the smoothness of the right-hand side and the nature of the Brownian force itself that prevents $J_3 = 0$, and so the coordinate change is not pathological.

It is of crucial importance to us that the distribution function is completely determined by the map of conformation space. It is this property that allows us to avoid calculating the distribution function when solving a flow problem. Instead, we compute the map, which evolves according to equations that are much more amenable to numerical solution than is (1.2*a*), as we show below.

2.1. *Lagrangian representation of the orientation distribution function for rigid microstructure*

Now we specialize the Lagrangian representation of the conformation distribution function to the case of microstructure consisting of rigid particles, where $\rho(t;\rho_0,\sigma_0,\theta_0) = \rho_0$ for all time. The distribution function (f) for a specific material point evolves according to a Fokker–Planck equation analogous to (1.2*a*), but conformation space is now described completely by only two orientation coordinates (σ,θ):

$$\frac{\partial f}{\partial t} + \nabla\cdot[f\dot{\boldsymbol{R}}] = 0, \tag{2.11a}$$

where

$$\dot{\boldsymbol{R}} = \mathscr{G}(\boldsymbol{R};\boldsymbol{E}(t),\boldsymbol{\Omega}(t),\mu) - \nu\nabla(\log f). \tag{2.11b}$$

In component form, we have

$$\dot{\boldsymbol{R}} = \dot{\sigma}\cos\theta\boldsymbol{e}_\sigma + \dot{\theta}\boldsymbol{e}_\theta,$$

where

$$\dot{\sigma} = \mathscr{G}_\sigma(\sigma,\theta;\boldsymbol{E}(t),\boldsymbol{\Omega}(t),\mu) - \frac{\nu}{\cos^2\theta}\frac{\partial}{\partial\sigma}(\log f), \tag{2.12a}$$

$$\dot{\theta} = \mathscr{G}_\theta(\sigma,\theta;\boldsymbol{E}(t),\boldsymbol{\Omega}(t),\mu) - \nu\frac{\partial}{\partial\theta}(\log f). \tag{2.12b}$$

The Lagrangian representation in conformation space of the ODF, f^*, is defined in a way that is analogous to (2.1). The Lagrangian representation of f evolves according to

$$\frac{f^*(t;\sigma_0,\theta_0)}{f^*(0;\sigma_0,\theta_0)} = \left[J_2\frac{\cos\theta}{\cos\theta_0}\right]^{-1}, \tag{2.13}$$

where

$$J_2 = \frac{\partial(\sigma,\theta)}{\partial(\sigma_0,\theta_0)}.$$

This equation is equivalent to (2.7) in the coordinate-free framework.

2.2. *Lagrangian representation of the orientation distribution function for rigid microstructure with directors* $\boldsymbol{R}$ *parallel to the* (x, y)*-plane*

Finally, we consider the special case of the ODF of microstructure consisting of rigid particles with all directors $\boldsymbol{R}$ lying parallel to the (x, y)-plane. This is a special case for two-dimensional flows that is useful for illustrative purposes. The ODF evolves according to (2.11a), but in this case,

$$\dot{\boldsymbol{R}} = \sigma \boldsymbol{e}_{\sigma}.$$

In coordinate form

$$\dot{\sigma} = \mathscr{G}_{\sigma}(\sigma; \boldsymbol{E}(t), \boldsymbol{\Omega}(t), \mu) - \nu \frac{\partial}{\partial \sigma}(\log f). \tag{2.14}$$

Once gain, we formulate and integrate a differential equation for f^* to yield a solution equivalent to (2.7),

$$\frac{f^*(t; \sigma_0)}{f^*(0; \sigma_0)} = [J_1]^{-1}, \tag{2.15}$$

where

$$J_1 = \frac{\partial \sigma}{\partial \sigma_0}.$$

In this section, we have developed a Lagrangian formalism for representation of the conformation distribution function of a microstructured fluid. Although we shall return to this point later, it is worth emphasizing that the Lagrangian representation would provide considerable computational advantage even if our goal were to calculate the distribution function, directly. Specifically, a numerical scheme based upon this approach is 'self-adaptive' in the sense that an equally spaced set of initial conditions will evolve following solution trajectories in conformation space. Thus, regions of conformation space with the highest conformational probability are ultimately represented by many material points, and conversely for regions of conformation space with the smallest conformation probability.

3. Exploitation of the Lagrangian representation of distribution functions in conformation space

Let us now consider the Lagrangian representation of moments of the distribution function, and of the Brownian contribution to the microdynamical equation. In the process, we shall see that the Lagrangian approach offers additional advantages of very considerable importance for computation of flows of microstructured fluids. Specifically, we demonstrate that moments of distribution functions that are required by the constitutive relation may be computed in the *reference* configuration by a change of variables that exploits the Lagrangian representation of distribution functions. In addition, we show how to rewrite the Brownian diffusion term in the microdynamical equation in a way that does not involve the distribution function. The map from reference to deformed coordinates for conformation space may be computed from the resulting equation. This is equivalent to, but considerably more efficient than, computing the distribution function.

3.1. *Moments of distribution functions*

The Lagrangian representation of the distribution function facilitates the calculation of moments by enabling integration of the moment in the original, undeformed configuration.

As an example, we consider the second moment of the ODF for rigid microstructure,

$$\langle \boldsymbol{RR} \rangle (t) = \int f(\boldsymbol{R}, t)\, \boldsymbol{R} \otimes \boldsymbol{R}\, \mathrm{d}\boldsymbol{R}. \tag{3.1}$$

The evolution of f is governed by a partial differential equation (2.11) in the Eulerian representation of conformation space. Alternatively, we have the Lagrangian representation of f, equation (2.13); the latter enables an easy change of variables as follows. Consider the second moment as being an integral in the *deformed* configuration. We can use the formal solution (2.13) to rewrite the moment as an integral in the reference (or initial) configuration as follows:

$$\begin{aligned} \langle \boldsymbol{RR} \rangle (t) &= \int f^*(t; \boldsymbol{R}_0)\, \boldsymbol{R} \otimes \boldsymbol{R}\, \mathrm{d}\boldsymbol{R} \\ &= \int f^*(t; \boldsymbol{R}_0)\, \boldsymbol{R} \otimes \boldsymbol{R} \det (\nabla_0 \boldsymbol{R})\, \mathrm{d}\boldsymbol{R}_0 \\ &= \int f^*(0; \boldsymbol{R}_0) [\boldsymbol{R}(t; \boldsymbol{R}_0) \otimes \boldsymbol{R}(t; \boldsymbol{R}_0)]\, \mathrm{d}\boldsymbol{R}_0. \end{aligned} \tag{3.2}$$

Depending on the initial state, (3.2) may simplify further. For example, for rigid microstructure with isotropic initial state, the second moment is simply

$$\langle \boldsymbol{RR} \rangle (t) = \frac{1}{4\pi} \int_0^{2\pi} \mathrm{d}\sigma \int_{-\pi/2}^{\pi/2} \mathrm{d}\theta_0 \cos\theta_0 (\boldsymbol{R} \otimes \boldsymbol{R})(t). \tag{3.3}$$

The form (3.3) is a simple matter to compute if we know how each initial orientation transforms into the current configuration, i.e. if we know the map from reference to deformed conformations of the microstructure that follows the material point with which f is associated.

In the same way, we can rewrite the fourth moment of the distribution function as

$$\langle \boldsymbol{RRRR} \rangle (t) = \int f^*(0; \boldsymbol{R}_0) [\boldsymbol{R}(t; \boldsymbol{R}_0) \otimes \boldsymbol{R}(t; \boldsymbol{R}_0) \otimes \boldsymbol{R}(t; \boldsymbol{R}_0) \otimes \boldsymbol{R}(t; \boldsymbol{R}_0)]\, \mathrm{d}\boldsymbol{R}_0. \tag{3.4}$$

Note that the same information is required to compute the second and the fourth moments of the distribution function; we require only the coordinate map from reference to deformed configurations.

In order to simplify moment calculations, we have used (2.7) to change variables. Because (2.7) also applies in the case where there is a Brownian contribution to the particle dynamics, the moment simplification works whether or not the Brownian contribution is included.

3.2. *Brownian diffusion*

The rheological properties of suspensions depend on the dynamical behaviour of individual elements of the microstructure which are carried along by the flow. Microstructural dynamics, in turn, are the result of the competition between the ordering tendencies of the surrounding flow, and the randomizing effects of Brownian motion. For larger microstructural elements, it may be possible to neglect the effects of Brownian diffusion.

In reality, the Brownian force on the microstructure is a rapidly and irregularly

fluctuating force that has an effect on the particle path, on the orientation and on the length of the microstructure. In this study, we are interested primarily in the relationship between the dynamical evolution of conformation of the microstructure and the rheology of the suspension. Therefore, we shall ignore the Brownian diffusion of microstructure across the pathlines of the flow.

We concentrate instead on the effects that Brownian forces have on the conformation of individual elements of the microstructure in a small material volume of suspension. It is possible to analyse the effects of a rapidly and irregularly fluctuating (Brownian) force on the solutions of the conformation evolution equations. However it is more convenient to make use of the usual assumption that the rate of change of the conformation has a Maxwellian distribution about the average. This is a natural assumption because it is possible to show, by statistical arguments, that equilibration of the distribution function in momentum space occurs on a timescale that is several orders of magnitude faster than that for equilibration in conformation space. As shown by Bird *et al.* (1987), this allows one to write down a velocity–space averaged Brownian force term which depends on the gradient of the logarithm of the conformation (or orientation) distribution function, as in (1.1).

An alternative route to the representation of the Brownian diffusion term as the gradient of the logarithm of the distribution function is by an equilibrium analysis of gradient diffusion. This approach was used by Einstein (1956) to obtain the diffusion coefficients that are associated with the smoothing of inhomogeneities in the concentration of spheres in suspension. More recently, Russel (1981) has given a rigorous statistical-mechanical derivation of a similar result, but with hydrodynamic interactions taken into account. For a survey of the research in this area, see the review article by Russel (1981).

Upon examination, (1.1) reveals that the Brownian term tends to smooth abrupt changes in the distribution function in conformation space. This is clear because the (averaged) Brownian diffusion pushes an individual element of the microstructure away from the direction of increasing conformational probability.

Although the dynamics of an element of the microstructure acted upon by a rapidly and irregularly varying (stochastic) Brownian force are unpredictable, at least in detail, it is of crucial importance in the present discussion that the dynamics of an element of the microstructure that is acted upon by a *velocity–space averaged Brownian force*, that is related to the distribution function, *is deterministic and predictable*. This statement seems, at first, to be paradoxical. The paradox is resolved when one realizes that the solution of (1.1) corresponds not to an actual time trace of the state vector of a particular element of microstructure, but rather to the *time-trace of the local velocity–space average of the state vectors* of elements of the ensemble which have a Maxwellian distribution about the mean rate of change of the conformation.

Normally, one solves (1.1) in conjunction with (1.2*a*), the Eulerian form of the conservation statement for the distribution function. In this section, we show how to solve (1.1) in conjunction with the Lagrangian representation of the conservation statement for the distribution function. Analytically, these two procedures are identical, i.e. no approximation is involved. Numerically, however, the use of the Lagrangian conservation statement allows for the development of fast, efficient, self-adaptive numerical algorithms for the solution of flow problems.

3.2.1. *Calculation of the Brownian force term*

For simplicity of exposition, we work first in the coordinate-free framework. The Brownian term is

$$-\nu\nabla\log\phi(\boldsymbol{R},t) = -\nu\frac{1}{\phi(\boldsymbol{R},t)}\nabla\phi(\boldsymbol{R},t).$$

Next, we substitute the Lagrangian form of the distribution function for the Eulerian form, using (2.1) and (2.7)

$$-\nu\nabla\log\phi(\boldsymbol{R},t) = -\nu\frac{\det(\nabla_0\boldsymbol{R})}{\phi^*(0;\boldsymbol{R}_0)}\nabla\left[\frac{\phi^*(0;\boldsymbol{R}_0)}{\det(\nabla_0\boldsymbol{R})}\right]. \tag{3.5}$$

The derivative ∇ may be recast as a derivative with respect to the reference configuration ∇_0 by changing variables; this yields

$$-\nu\nabla\log\phi(\boldsymbol{R},t) = -\nu\frac{\det(\nabla_0\boldsymbol{R})}{\phi^*(0;\boldsymbol{R}_0)}\nabla_0\left[\frac{\phi^*(0;\boldsymbol{R}_0)}{\det(\nabla_0\boldsymbol{R})}\right](\nabla_0\boldsymbol{R})^{-1}. \tag{3.6}$$

These expressions may also be written in terms of the coordinates introduced previously. For example, consider the case of initially isotropic, rigid microstructure confined to the (x,y)-plane. In coordinates, (3.6) takes the form

$$-\nu\nabla[\log f(\sigma,t)] = \boldsymbol{e}_\sigma\nu\left(\frac{\partial\sigma}{\partial\sigma_0}\right)^{-2}\frac{\partial}{\partial\sigma_0}\left(\frac{\partial\sigma}{\partial\sigma_0}\right). \tag{3.7}$$

Thus the full, rearranged microdynamical equation, which incorporates the Lagrangian conservation statement for the distribution function, is

$$\frac{\partial\sigma}{\partial t} = \mathscr{G}_\sigma(\sigma;\boldsymbol{E}(t),\boldsymbol{\Omega}(t),\mu) + \nu\left(\frac{\partial\sigma}{\partial\sigma_0}\right)^{-2}\frac{\partial}{\partial\sigma_0}\left(\frac{\partial}{\partial\sigma_0}\right). \tag{3.8}$$

Thus, by making use of the Lagrangian conservation statement for the distribution function, we have been able to rewrite the microdynamical equation in an exactly equivalent form that does not involve the distribution function. In doing so, we have constructed (3.8), which may be thought of as a partial differential equation for the coordinate map $\sigma_0 \to \sigma(t;\sigma_0)$. The coordinate map evaluated at a specific initial orientation σ_0^*, say, evolves in exactly the same way as the solution to (1.1) and (1.2*a*) with initial condition σ_0^*. All we have really done is to replace the formulation

$$\dot{\sigma} = \mathscr{G}_\sigma - \nu\frac{\partial}{\partial\sigma}(\log f), \quad \frac{\partial f}{\partial t} + \frac{\partial}{\partial\sigma}(f\dot{\sigma}) = 0$$

by the formulation (3.8). The advantages of this restructuring of the governing equations will become clear when we formulate a numerical method for the solution of (3.8).

Recall that the coordinate change from the current conformation coordinates (here σ) to the reference conformation coordinates (σ_0) is possible only when the relevant Jacobian (here J_1) is non-zero. Upon examination, (3.8) reveals that the Brownian term is proportional to J_1^{-2}, and in the direction that tends to increase J_1 where J_1 is low, and to decrease J_1 where J_1 is high. Thus, the pathological situation $J_1 = 0$ is avoided. Similar arguments apply in the two- and three-dimensional cases which we analyse below.

For rigid microstructure that orients in three dimensions, the Brownian term may be rewritten as

$$-\nu\nabla[\log f(\sigma,\theta,t)] = \frac{-\nu}{f^*(0;\sigma_0,\theta_0)}\left[\frac{\boldsymbol{e}_\sigma}{\cos\theta_0}\frac{\partial(\cdot,\theta)}{\partial(\sigma_0,\theta_0)}+\boldsymbol{e}_\theta\frac{\cos\theta}{\cos\theta_0}\frac{\partial(\sigma,\cdot)}{\partial(\sigma_0,\theta_0)}\right] \times\left(\frac{f^*(0;\sigma_0,\theta_0)}{J_2}\frac{\cos\theta_0}{\cos\theta}\right). \quad (3.9)$$

Finally, for stretchable, orientable microstructure, the Brownian term is

$$-\nu\nabla[\log\phi(\rho,\sigma,\theta,t)] = \frac{-\nu}{\phi^*(0;\rho_0,\sigma_0,\theta_0)}\left[\boldsymbol{e}_\rho\frac{\cos\theta}{\cos\theta_0}\frac{\rho^2}{\rho_0^2}\frac{\partial(\cdot,\sigma,\theta)}{\partial(\rho_0,\sigma_0,\theta_0)}\right.$$
$$\left.+\frac{\boldsymbol{e}_\sigma}{\cos\theta_0}\frac{\rho}{\rho_0^2}\frac{\partial(\rho,\cdot,\theta)}{\partial(\rho_0,\sigma_0,\theta_0)}+\boldsymbol{e}_\theta\frac{\cos\theta}{\cos\theta_0}\frac{\rho}{\rho_0^2}\frac{\partial(\rho,\sigma,\cdot)}{\partial(\rho_0,\sigma_0,\theta_0)}\right]\left(\frac{\phi^*(0;\rho_0,\sigma_0,\theta_0)}{J_3}\frac{\cos\theta_0}{\cos\theta}\frac{\rho_0^2}{\rho^2}\right). \quad (3.10)$$

Of course, if the initial state of the conformation is relaxed, then (3.9) and (3.10) will take simpler forms.

It is worth mentioning that this alternative way of computing the Brownian term involves no approximation whatsoever, except of course numerical approximation. Furthermore, the system (3.8) (or the analogous problems in two and three conformation space dimensions) is particularly advantageous in strongly orienting flows that lead to sharply varying distribution functions that are very difficult to resolve in the conventional Eulerian representation of the distribution function.

3.3. *The numerical solution of flow problems*

Let us now consider how one might solve a flow problem using the double-Lagrangian technique. First, let us fix some model equations assuming that we wish to solve a flow problem for microstructure consisting of 'particles' suspended in a Newtonian fluid. Assuming that the stress tensor is partitioned into Newtonian and non-Newtonian parts, the momentum and continuity equations for an incompressible suspension are

$$\rho\left(\frac{\partial}{\partial t}+\boldsymbol{u}\cdot\nabla\right)\boldsymbol{u} = -\nabla p+\eta\nabla^2\boldsymbol{u}+\nabla\cdot(\boldsymbol{\tau}^{\mathrm{NN}}), \quad \nabla\cdot\boldsymbol{u}=0.$$

These field equations over the spatial domain, together with appropriate boundary conditions, could be approximated numerically by any of the standard techniques. The divergence of the particle contribution to the stress is to be left as a body force.

The non-Newtonian stress for microstructured fluids is specified in terms of some moment(s) of the distribution function. Here, we advocate computing the non-Newtonian stress associated with a (sufficient) number of material points. Associated with each material point is a group of (a sufficient number of) elements of the suspended phase that evolves according to an equation like (1.1), but with the Brownian force rewritten by the method of §3.2. In reality, the ensemble of suspended elements is specified as a set of initial conditions in conformation space. These elements of the suspended phase evolve in a way that provides a discretized form of the coordinate map from reference to deformed (or current) conformations for each material point. Thus it is a simple matter to compute the non-Newtonian stress for each material point, by using the discretized form of the coordinate map

to rewrite the moment integral(s) over the reference configuration. Once the non-Newtonian stress is known for a set of material points distributed over the spatial domain, the body force in the momentum equation may be computed in a way that is appropriate for the numerical method being used. Of course, adequate resolution in conformation space and in physical space will, in general, depend on the details of the specific flow, as for any computational method.

There are a number of interesting features that make this method of accounting for the non-Newtonian stress very attractive from a numerical point of view. The most important aspect is that the scheme we have just described is self-adaptive. To understand this, note that within the ensemble at each material point, the elements of the suspended phase stretch and orient in a natural way in response to the changing flow conditions, Brownian diffusion, etc. Because the discretized form of the coordinate map from reference to deformed conformation is obtained from the natural motion of elements of the suspended phase, resolution of the map will be greatest where the conformational probability is greatest. Moreover, a minimum of resolution is 'wasted' on those regions of conformation space where conformational probability is least.

A second attractive aspect of the computational scheme is that the moment integrals for each material point care carried out in the reference configuration. This is especially convenient when the reference configuration is the relaxed state, for then fast and efficient algorithms for integration that rely on evenly spaced values of the integrand may be used.

Thirdly, it is expected that the scheme we have outlined will suffer less from the second major problem faced by direct numerical integration of moment evolution equations, namely the development of steep spatial gradients (or indeed discontinuity) in the moment tensor field. Because our scheme of accounting for the microstructure is Lagrangian in physical space, there are no spatial derivatives of the moment tensor field to discretize, as there are on the left-hand side of equation (1.5*b*), for example. Therefore, steep spatial gradients of the moment tensor field will not affect the integration of the microstructure evolution equations. Of course, steep spatial gradients in the non-Newtonian stress associated with the microstructure may still cause problems with the macroscopic mechanics, depending on what scheme is used to solve the flow equations.

Finally, we remark that the numerical scheme we have sketched here is especially amenable to vector and parallel solution. The ensemble of microstructural elements at each material point evolves over a time-step in a way that is independent of the ensembles at other material points, thus allowing for the evolution of each such ensemble to be calculated by a different processor. Within an ensemble, one could integrate the microdynamical equation(s) (of a type similar to (3.8)) in a way that allows for fast vector processing.

4. Examples of microstructured fluids to which our methods may be applied

There are three primary characteristics of microstructured fluids to which the present methods may be applied:

(i) distribution functions that evolve according to a classical conservation equation that involves a microdynamical equation;

(ii) microdynamical equations (that may involve the distribution function) that lead to smooth dependence of the conformation on initial conditions.

These two characteristics allow for a Lagrangian representation of the conservation statement, as in §2, and for restructuring the microdynamical equations so as to eliminate explicit dependence on the distribution function, as in §3. The third characteristic,

(iii) macroscopic properties of the suspension that involve the distribution function, possibly through moments, etc.,

allows for the calculation of the macroscopic properties of the fluid in terms of the Lagrangian representation of the distribution function. A large number of systems share these characteristics; we shall mention some representative examples in what follows, though this is not meant to be an exhaustive list. For detailed descriptions of the equations and models we mention below, see Bird *et al.* (1987), Larson (1988) or Doi & Edwards (1986).

4.1. *Dilute suspensions of rigid ellipsoidal particles*

A dilute suspension of rigid ellipsoidal particles evolves according to equations that are in the form of our model equations (1.1) and (1.2*a*):

$$\dot{\boldsymbol{R}} = \boldsymbol{\Omega}\cdot\boldsymbol{R} + \left(\frac{r^2-1}{r^2+1}\right)[\boldsymbol{E}\cdot\boldsymbol{R} - \boldsymbol{E}:\boldsymbol{RRR}] - \nu\nabla(\log f), \quad \frac{\partial f}{\partial t} + \nabla\cdot(f\dot{\boldsymbol{R}}) = 0. \tag{4.1}$$

Here $\boldsymbol{R}$ is the director (of constant length), r is the aspect ratio of the ellipsoids, and the Brownian diffusion coefficient is $\nu = kT/R_\omega$ (k is Boltzmann's constant, T is the absolute temperature and R_ω is the resistance coefficient for rotation of a particle about a transverse axis through its centre). These equations also describe the evolution of a dilute suspension of rodlike particles (or fibres) in the limit $r \to \infty$. Detailed expressions for the viscous and entropic stress contributions in terms of second and fourth moments are available in Leal & Hinch (1971) and in Hinch & Leal (1972). Application of the double-Lagrangian technique to flows of suspensions of rigid ellipsoidal particles is advantageous, in order to avoid the closure problem of the direct moment evolution equation approach.

4.2. *Theories for semi-dilute suspensions of rodlike particles*

Theories for semi-dilute suspensions of rodlike particles ($|\boldsymbol{R}| = 1$), such as the theory due to Doi & Edwards (1986), augment (4.1) in the limit $r \to \infty$ with an *orientation-dependent diffusion coefficient*, thus:

$$\dot{\boldsymbol{R}} = \boldsymbol{\Omega}\cdot\boldsymbol{R} + \boldsymbol{E}\cdot\boldsymbol{R} - \boldsymbol{E}:\boldsymbol{RRR} - \hat{\nu}(\boldsymbol{R})\nabla(\log f), \quad \frac{\partial f}{\partial t} + \nabla\cdot(f\dot{\boldsymbol{R}}) = 0, \tag{4.2a}$$

where the orientation-dependent diffusion coefficient is (note that $\boldsymbol{Q}$ is just a dummy variable).

$$\hat{\nu}(\boldsymbol{R}) = \nu\left[\frac{4}{\pi}\int|\boldsymbol{R}\times\boldsymbol{Q}|f(\boldsymbol{Q})\,\mathrm{d}\boldsymbol{Q}\right]^{-2}. \tag{4.2b}$$

In this case, the solution of a flow problem by the double-Lagrangian technique is advantageous because it allows one to avoid the closure problem. An additional advantage concerns the diffusion coefficient. The orientation-dependent diffusion coefficient is commonly pre-averaged over $\boldsymbol{R}$ in order to obtain a useful direct moment evolution equation, but this is not necessary when using the double-Lagrangian technique. The reason is that the integral over conformation space in the diffusion coefficient (4.2*b*) may be recast as an integral over the reference

configuration using the method of §3.1. In other respects, the solution of flow problems for these suspensions is a straightforward application of the methods we have outlined.

4.3. *Theories for nematic polymers*

In the theory for nematic polymers due to Doi (1981), the equations for a semi-dilute solution of rodlike particles are augmented by a potential $\mathscr{U}$ that involves the tensor order parameter $\boldsymbol{S}$, which may be expressed in terms of the second moment of the distribution function

$$\boldsymbol{S} = \langle \boldsymbol{RR} \rangle - \tfrac{1}{3}\boldsymbol{I}.$$

In this expression, $\boldsymbol{I}$ is the identity tensor. The equations that describe the evolution of $\boldsymbol{R}$ and f are

$$\dot{\boldsymbol{R}} = \boldsymbol{\Omega}\cdot\boldsymbol{R} + \boldsymbol{E}\cdot\boldsymbol{R} - \boldsymbol{E}:\boldsymbol{RRR} - \hat{\nu}(\boldsymbol{R})\,\nabla(\log f) - \hat{\nu}(\boldsymbol{R})\,\nabla\left(\frac{\mathscr{U}}{kT}\right),$$

$$\frac{\partial f}{\partial t} + \nabla\cdot(f\dot{\boldsymbol{R}}) = 0, \quad \mathscr{U} = \text{const.} - \tfrac{3}{2}U_0\,kT\boldsymbol{RR}:\boldsymbol{S}. \tag{4.3}$$

Application of the double-Lagrangian technique is straightforward, except that the integrals in the orientation-dependent diffusion coefficient and in the potential $\mathscr{U}$ are both written as integrals over the reference configuration by the methods of §3.1. Again, no pre-averaging is necessary.

4.4. *Dumbbell models of dilute polymer solutions (including internal viscosity, hydrodynamic interaction, excluded volume, finite extensibility, conformation-dependent friction, etc.)*

This is a large class of models for polymer molecules in dilute solution. The first and simplest member of this class is the linear elastic dumbbell, due to Kuhn (1934):

$$\dot{\boldsymbol{R}} = \boldsymbol{\Omega}\cdot\boldsymbol{R} + \boldsymbol{E}\cdot\boldsymbol{R} - \frac{4kT\beta^2}{\zeta}\boldsymbol{R} - \frac{2kT}{\zeta}\nabla(\log\phi), \quad \frac{\partial\phi}{\partial t} + \nabla\cdot(\phi\dot{\boldsymbol{R}}) = 0, \tag{4.4}$$

where ζ is a (constant) friction coefficient of the beads and $4kT\beta^2\boldsymbol{R}$ is the connector (Hookean) spring force. It is worth mentioning that this micromechanical model leads to an upper-convected Maxwell constitutive equation. Application of the double-Lagrangian technique to flows of suspensions of linear elastic dumbbells is straightforward. Note, however, that because the direct moment evolution equations for suspensions of simple linear elastic dumbbells are closed without approximation, the double-Lagrangian technique is not necessarily favoured for the solution of flow problems in this limiting case. On the other hand, a numerical scheme that is Lagrangian in physical space may address numerical difficulties associated with steep gradients in moment tensor fields.

The simple model (4.4) was later changed to include internal viscosity of the dumbbell, yielding the microdynamical equation

$$\dot{\boldsymbol{R}} = \left(\boldsymbol{I} - \frac{\epsilon}{1+\epsilon}\boldsymbol{RR}\right)\left[\boldsymbol{\Omega}\cdot\boldsymbol{R} + \boldsymbol{E}\cdot\boldsymbol{R} - \frac{4kT\beta^2}{\zeta}\boldsymbol{R} - \frac{2kT}{\zeta}\nabla(\log\phi)\right], \tag{4.5}$$

where the parameter ϵ is the internal viscosity divided by the friction coefficient of the beads. Here, the double-Lagrangian technique is recommended for the solution of flow problems, because the internal viscosity leads to a closure problem in the direct moment evolution equations.

Another modification that has been made, in order to account for bead–bead hydrodynamic interaction involves the Oseen tensor

$$\boldsymbol{O}(\boldsymbol{R}) = \frac{1}{8\pi\eta_s|\boldsymbol{R}|}\left(\boldsymbol{I}+\frac{\boldsymbol{R}\boldsymbol{R}}{|\boldsymbol{R}|^2}\right). \tag{4.6a}$$

Here η_s is the viscosity of the solvent. The microdynamical equation is

$$\dot{\boldsymbol{R}} = \boldsymbol{\Omega}\cdot\boldsymbol{R}+\boldsymbol{E}\cdot\boldsymbol{R}-\frac{2kT}{\zeta}(\boldsymbol{I}-\zeta\boldsymbol{O}(\boldsymbol{R}))\cdot(2\beta^2\boldsymbol{R}+\nabla(\log\phi)). \tag{4.6b}$$

Clearly, bead–bead hydrodynamic interactions will lead to a closure problem in deriving moment equations from (4.6*b*), and the double-Lagrangian technique can be used with advantage. An additional point in favour of our methods is that although the Oseen tensor is normally pre-averaged, this is not necessary when solving a flow problem using the double-Lagrangian technique.

Another modification to the simple dumbbell model is to include an excluded volume potential (Fixman 1966), which normally depends on the second moment $\langle\boldsymbol{R}\boldsymbol{R}\rangle$ and on $\boldsymbol{R}\cdot\boldsymbol{R}$. If the excluded volume potential ($\mathscr{E}$) is combined with the connector force potential ($\mathscr{S}$) into a total potential $\mathscr{V} = \mathscr{E}+\mathscr{S}$, then the microdynamical equations are

$$\dot{\boldsymbol{R}} = \boldsymbol{\Omega}\cdot\boldsymbol{R}+\boldsymbol{E}\cdot\boldsymbol{R}-\frac{2kT}{\zeta}\left(\frac{1}{kT}\nabla\mathscr{V}+\nabla(\log\phi)\right). \tag{4.7}$$

These equations again exhibit the closure problem. However, the solution of flow problems using the double-Lagrangian technique is a simple matter if the moments in the excluded volume potential are recast as integrals over the reference configuration, using the method of §3.1.

Finally, we mention the extension of the simple dumbbell to include finite extensibility and conformation-dependent friction. An important way to account for finite extensibility is to model the connector force by the Warner spring (Warner 1972),

$$\boldsymbol{F}^{\mathrm{C}} = \frac{3kT}{Na^2}\left[\frac{1}{1-(|\boldsymbol{R}|/|\boldsymbol{R}_{\max}|)^2}\right]\boldsymbol{R} = H(|\boldsymbol{R}|^2)\boldsymbol{R}, \tag{4.8a}$$

where N is the number of subunits making up the macromolecule (each of length a), and $|\boldsymbol{R}_{\max}| = Na$ is the total length of the extended macromolecule. The connector force limits the extension of the dumbbell through the asymptotic behaviour

$$|\boldsymbol{F}^{\mathrm{C}}|\to\infty \quad \text{as} \quad |\boldsymbol{R}|\to|\boldsymbol{R}_{\max}|.$$

Also we include the conformation-dependent linear isotropic friction law (Hinch 1977)

$$\boldsymbol{F}^{\mathrm{H}} = \zeta(|\boldsymbol{R}|)\,(\boldsymbol{u}_i-\dot{\boldsymbol{r}}_i). \quad \text{where} \quad \zeta(|\boldsymbol{R}|) = \zeta_0\frac{|\boldsymbol{R}|}{N^{\frac{1}{2}}a}. \tag{4.8b}$$

Here $\boldsymbol{F}^{\mathrm{H}}$ is the force on bead i, $\boldsymbol{u}_i$ is the undisturbed solvent velocity evaluated at the bead centre, $\boldsymbol{r}_i$ is the position vector of the centre of bead i, $\zeta_0 = 6\pi\eta_s N^{\frac{1}{2}}a$ is the equilibrium bead friction, where the equilibrium lengthscale of the dumbbell is $N^{\frac{1}{2}}a$. This leads to the microdynamical equation

$$\dot{\boldsymbol{R}} = \boldsymbol{\Omega}\cdot\boldsymbol{R}+G(|\boldsymbol{R}|)\,\boldsymbol{E}\cdot\boldsymbol{R}-\frac{2kT}{\zeta(|\boldsymbol{R}|)}\left(\frac{1}{kT}H(|\boldsymbol{R}|^2)\,\boldsymbol{R}+\nabla(\log\phi)\right). \tag{4.8c}$$

Here $G(|\boldsymbol{R}|)$ is a conformation-dependent shape factor. Normally, the direct moment evolution equation is pre-averaged in an attempt to elude the closure problem, but this is unnecessary when solving a flow problem using the double-Lagrangian technique.

4.5. *Bead–spring chain models of polymer molecules in solution*

Closely related to the examples discussed above, which all involve particles that may only stretch and orient as a whole, are the many-bead models for polymer molecules in solution. These theories attempt to account for the many degrees of conformational freedom of flexible-coil polymer molecules in solution. An example is the Rouse chain, which involves a chain of beads connected by Hookean springs. The confirmation distribution function for the jth link in the chain ψ_j (in 'normal coordinates' $\boldsymbol{R}'_j$) evolves according to the equation

$$\left.\begin{aligned} \dot{\boldsymbol{R}}'_j = \boldsymbol{\Omega}\cdot\boldsymbol{R}'_j + \boldsymbol{E}\cdot\boldsymbol{R}'_j - \frac{a_j}{\zeta}\left(H\boldsymbol{R}'_j - kT\frac{\partial}{\partial \boldsymbol{R}'_j}(\log\psi_j)\right), \\ \frac{\partial\psi_j}{\partial t} + \frac{\partial}{\partial \boldsymbol{R}'_j}\cdot(\psi_j\dot{\boldsymbol{R}}'_j) = 0, \end{aligned}\right\} \tag{4.9}$$

where H is the spring constant and $a_j = 4\sin^2(j\pi/2N)$ are the eigenvalues of the Rouse matrix of the chain of N beads. In this case, one can apply the double-Lagrangian technique to each link in the N-bead chain to rewrite the Brownian term in the microdynamical equation in a way that does not involve the distribution function, and to compute the stresses that depend on moments such as $\langle \boldsymbol{R}'_j\boldsymbol{R}'_j\rangle$.

4.6. *Reptation models of concentrated polymer solutions and melts*

In the theory of concentrated polymer solutions and melts, one must account for the fact that the mobility of an individual polymer molecule is severely constrained by its neighbours, with the result that a dominant mode of deformation is a 'snakelike' motion along the contour of the molecule, which has become known as reptation. As reviewed in Bird *et al.* (1987) and in Doi & Edwards (1986), one can derive a single-link diffusion equation for the distribution function of orientation along the contour of the chain, $f_\alpha(\boldsymbol{u}, s, t)$. Here $\boldsymbol{u}$ is a direction in orientation space, s is the arclength coordinate along the chain ($0 \leqslant s \leqslant 1$), and the subscript α denotes the species. The distribution function evolves according to an equation of the form

$$\frac{\partial f_\alpha}{\partial t} + \nabla\cdot\left[f_\alpha\left(\boldsymbol{\kappa}\cdot\boldsymbol{u} - \boldsymbol{\kappa}:\boldsymbol{uuu} - \frac{\epsilon' N_\alpha^2}{\lambda_\alpha}\nabla(\log f_\alpha)\right)\right] = \frac{(1-\epsilon')}{\lambda_\alpha}\frac{\partial^2}{\partial s^2}f_\alpha. \tag{4.10}$$

Note that the gradient operators in (4.10) are in orientation space ($\boldsymbol{u}$), but not in arclength space (s). Here the length of the chain is N_α, the associated characteristic time is λ_α, and ϵ' is the reputation coefficient. At equilibrium, $\epsilon' = 0$; in other words, in the absence of flow there is no reptation, only diffusion in orientation space. Away from equilibrium, $0 \leqslant \epsilon' \leqslant 1$ (see Bird *et al.*); thus when the material flows, there is generally diffusion associated with both reptation and orientation. Note that when $\epsilon' = 1$ there is no reptation, and (4.10) is identical to the evolution equation for the distribution function of rigid ellipsoidal particles in the limit of aspect ratio $r \to \infty$.

The double-Lagrangian technique is somewhat less straightforward to apply to theories of concentrated polymer solutions and melts due to the added complication of the arclength coordinate along the chain, s. The conformation space in which the

distribution function resides is rather more complicated than just the product of orientation and stretch spaces, say, as for dumbbell models of dilute polymer solutions.

For simplicity of presentation, let us consider, for the moment, chains which lie in the (x, y)-plane; thus orientation space ($\boldsymbol{u}$) at each link along the chain is described by the single angle σ. The distribution function under consideration is then a function of σ, s and t. The conformation space is the product of a line segment $\{s: 0 \leqslant s \leqslant 1\}$ and a circle $\{\sigma: 0 \leqslant \sigma < 2\pi\}$; i.e. the conformation space is a cylinder.

Equation (4.10) may be written into the form

$$\frac{\partial f}{\partial t}+\frac{\partial}{\partial \sigma}(f\dot{\sigma}) = \frac{(1-\epsilon')}{\lambda_\alpha}\frac{\partial^2}{\partial s^2}f, \tag{4.11}$$

where we have dropped the subscript α from f_α. The evolution equation for σ is just the σ-component of the term in parenthesis involving $\boldsymbol{u}$ in (4.10); we return to this point later. Now, we write the reptation term on the right-hand-side of (4.11) in a way that will facilitate application of the double-Lagrangian technique:

$$\frac{\partial f}{\partial t}+\frac{\partial}{\partial \sigma}(f\dot{\sigma})+\frac{\partial}{\partial s}(f\dot{s}) = 0. \tag{4.12a}$$

In (4.12a), we have introduced an evolution equation for the arclength coordinate:

$$\dot{s} = -\frac{(1-\epsilon')}{\lambda_\alpha}\frac{\partial}{\partial s}(\log f(\sigma, s, t)). \tag{4.12b}$$

The equation for s describes reptational motion, in exactly the same way that the equation for σ describes angular motion of links of the chain.

In a planar flow, we can write the vorticity and rate-of-strain tensors in components as (see the Appendix)

$$\boldsymbol{\Omega} = \begin{bmatrix} 0 & -\frac{1}{2}\omega & 0 \\ \frac{1}{2}\omega & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \boldsymbol{E} = \begin{bmatrix} e & \frac{1}{2}\gamma & 0 \\ \frac{1}{2}\gamma & -e & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

and the evolution equation for σ as

$$\dot{\sigma} = \frac{\omega}{2}-e\sin 2\sigma+\frac{\gamma}{2}\cos 2\sigma-\frac{\epsilon' N_\alpha^2}{\lambda_\alpha}\frac{\partial}{\partial \sigma}(\log f(\sigma, s, t)). \tag{4.12c}$$

Now, in applying the double-Lagrangian technique, we eliminate the distribution function $f(\sigma, s, t)$ from the system (4.12) in favour of the coordinate map $(\sigma_0, s_0) \rightarrow (\sigma(t; \sigma_0, s_0),\ s(t; \sigma_0, s_0))$, which is a solution of (4.12b, c). The Lagrangian form of the distribution function is

$$f^*(t; \sigma_0, s_0) = f(\sigma, s, t)|_{s=\hat{s}(t;\sigma_0, s_0),\ \sigma=\hat{\sigma}(t;\sigma_0, s_0)}.$$

By arguments similar to those presented in §2, one arrives at the integrated form of the Lagrangian representation of the single-link distribution function:

$$\frac{f^*(t; \sigma_0, s_0)}{f^*(0; \sigma_0, s_0)} = \left[\frac{\partial(\sigma, s)}{\partial(\sigma_0, s_0)}\right]^{-1} \equiv J^{-1}. \tag{4.13}$$

Equation (4.13) is exactly equivalent to (4.12a) provided the Jacobian is non-zero.

The relaxed state of the distribution function is characterized by isotropy in orientation space (at each link along the chain), and homogeneity in arclength. Using (4.13), and assuming that the chain is initially in a relaxed state, one can rewrite (4.12*b*, *c*) in the following way, analogous to the development of §3.2:

$$\dot{s} = \frac{(1-\epsilon')}{\lambda_\alpha} J^{-2} \frac{\partial(\sigma, J)}{\partial(\sigma_0, s_0)}, \tag{4.14a}$$

$$\dot{\sigma} = \frac{\omega}{2} - e \sin 2\sigma + \frac{\gamma}{2} \cos 2\sigma + \frac{\epsilon' N_\alpha^2}{\lambda_\alpha} J^{-2} \frac{\partial(J, s)}{\partial(\sigma_0, s_0)}. \tag{4.14b}$$

Of course, there are boundary conditions that must accompany (4.14), because the chain has ends, and because s must retain the sense of arclength. Assuming the initial state is relaxed, and that the ends of the chain are isotropic, the initial and boundary conditions for (4.14*a*, *b*) are, respectively,

$$\left.\begin{aligned} s(t=0;\sigma_0,s_0) = s_0, \quad s(t;\sigma_0,s_0=0) = 0, \quad s(t;\sigma_0,s_0=1) = 1, \\ \sigma(t=0;\sigma_0,s_0) = \sigma_0, \quad \sigma(t;\sigma_0,s_0=0) = \sigma_0, \quad \sigma(t;\sigma_0,s_0=1) = \sigma_0. \end{aligned}\right\} \tag{4.14c}$$

We have shown that the microdynamical equation for orientational and reptational modes may be integrated without computation of the single-link distribution function. In concentrated polymer solutions and melts, the stresses are computed as moments of the distribution function, integrated over orientation space and over arclength. These may be recast in the reference configuration in the manner demonstrated in §3.1. The extension of the present arguments to the case where the links in the chain are free to orient in three dimensions is straightforward.

5. Conclusions

In this paper, we have presented the theoretical basis for a new method for the solution of flow problems of microstructured fluids. Our method is centred on an equivalent representation of the conformation dynamics of local structure in a Lagrangian fashion in conformation space. There are several useful ramifications of this idea. The Lagrangian character of the method yields increased resolution in regions of conformation space where the conformational probability is high; thus the numerical scheme we propose is self-adaptive. Moreover, there is no need to compute the distribution function explicitly in order to calculate moments or to account for Brownian effects. Finally, the deformed configuration of the local structure at any particular time is obtained as part of the solution, making the method especially attractive to apply to manufacturing problems in which the final conformation distribution of the local structure in a material that once flowed is the critical unknown.

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Appendix. Analysis of a model microstructured fluid

In this Appendix, we attempt to fix ideas by consideration of a specific microstructured fluid. The microdynamical equations for the evolution of the conformation of stretchable, orientable particles described by a single axial vector

are considered in SWL. For reference, we review that analysis here, especially for the important general case of the dynamics of microstructure in two-dimensional flows that are *complex* (unsteady or spatially inhomogeneous velocity gradient tensor) as opposed to *simple* (steady and spatially homogeneous velocity gradient tensor). After we review the analysis of SWL (which does not include Brownian diffusion), we demonstrate that one may solve for the Lagrangian form of the distribution function in terms of the mathematical structures they develop.

A.1. *The conformation evolution equations*

Olbricht, Rallison & Leal (1982) demonstrate that conformation evolution equations for several types of microstructure may be collapsed down to the single vector equation of the form

$$\dot{\boldsymbol{R}} = \boldsymbol{\Omega}\cdot\boldsymbol{R} + G\left[\boldsymbol{E}\cdot\boldsymbol{R} - \frac{F}{F+1}\frac{\boldsymbol{R}\cdot\boldsymbol{E}\cdot\boldsymbol{R}}{\|\boldsymbol{R}\|^2}\boldsymbol{R}\right] - \frac{\alpha}{F+1}\boldsymbol{R}, \tag{A 1}$$

for each 'particle'. The orientation and length of the particle are given by the state vector $\boldsymbol{R}$. $\boldsymbol{\Omega}$ and $\boldsymbol{E}$ are the vorticity and strain-rate tensors, respectively, and the parameters G, α and F correspond to the shape factor, the elastic modulus of the (linear) internal spring and the internal damping of the particle, respectively. The reader may refer to Olbricht *et al.* (1982) for the parameter values which yield the particular microdynamical equations encompassed by this model equation. For our purposes, it is sufficient to note that $\alpha \geqslant 0$, $F \geqslant 0$ and G is generally between 0 and 1, although it may exceed 1 for certain exotic particles, as shown by Bretherton (1962). Note that the system (A 1) does not include the effects of Brownian diffusion. Also, the linear spring term causes certain pathological behaviour, as discussed in SWL. Nevertheless (A 1) is an interesting and instructive system to study.

We consider the evolution of the conformation of particles suspended in complex two-dimensional flows. Therefore, as in SWL, we define a rectangular coordinate system (x, y, z) where the flow occurs in the (x, y)-plane, although the particle is free to move out of the plane. In these coordinates, the vorticity and rate-of-strain tensors are

$$\boldsymbol{\Omega} = \begin{bmatrix} 0 & -\frac{1}{2}\omega & 0 \\ \frac{1}{2}\omega & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \boldsymbol{E} = \begin{bmatrix} e & \frac{1}{2}\gamma & 0 \\ \frac{1}{2}\gamma & -e & 0 \\ 0 & 0 & 0 \end{bmatrix}.$$

Here the flow parameters e (elongation), γ (shear) and ω (vorticity) depend on time through the motion of the particle in the unsteady, spatially inhomogeneous velocity field. In terms of the stream function the flow parameters are

$$\omega = -\frac{\partial^2\psi}{\partial x^2} - \frac{\partial^2\psi}{\partial y^2}, \quad e = \frac{\partial^2\psi}{\partial x\,\partial y}, \quad \gamma = \frac{\partial^2\psi}{\partial y^2} - \frac{\partial^2\psi}{\partial x^2}.$$

In the modified spherical polar coordinates of figure 1, the evolution equations for the conformation variables (ρ, σ, θ) are

$$\dot{\sigma} = \tfrac{1}{2}\omega - Ge\sin 2\sigma + \tfrac{1}{2}G\gamma\cos 2\sigma, \tag{A 2a}$$

$$\dot{\theta} = -\tfrac{1}{2}G(e\cos 2\sigma + \tfrac{1}{2}\gamma\sin 2\sigma)\sin 2\theta, \tag{A 2b}$$

$$\dot{\rho} = \left[\frac{G}{F+1}(e\cos 2\sigma + \tfrac{1}{2}\gamma\sin 2\sigma)\cos^2\theta - \frac{\alpha}{F+1}\right]\rho. \tag{A 2c}$$

The system (A 2) is normally a non-autonomous system of ordinary differential equations when the velocity-gradient tensor of the surrounding fluid flow is unsteady or spatially inhomogeneous. In the very special case of simple flows, (A 2) may be integrated by quadrature, as shown in SWL.

A.2. *Evolution of the conformation in complex flows*

When the surrounding flow is simple (and non-degenerate), then there is a single (or no) equilibrium orientation to which all initial orientations are asymptotic. This attracting orientation is steady, and therefore the meaning of attraction is based on the eigenvalues of the system (A 2) evaluated at the stable equilibrium orientation.

In complex flows, there may be analogous attracting orientation(s) of the microstructure. However, such attractors are generally time dependent rather than steady, and therefore we must expand on the notion of attraction. This observation lead SWL to consider directly the relative approach or separation of two distinct orientation time traces which are solutions of (A 2). It was found that two orientations experience a net approach (separation) over the time interval [0, T] whenever the quantity

$$\mathrm{CE}[\sigma_{10}, \sigma_{20}; T] \equiv \int_0^T 2Ge(t)\cos\left(\sigma(t;\sigma_{10})+\sigma(t;\sigma_{20})\right)+G\gamma(t)\sin\left(\sigma(t;\sigma_{10})+\sigma(t;\sigma_{20})\right)]\,\mathrm{d}t$$

is positive (negative). This quantity is called the *contraction exponent*. It depends on the initial orientation of the particles, on the time interval, and implicitly on the flow parameters. The time traces $\sigma(t;\sigma_{i0})$ of the two orientations are determined by integration of (A 2*a*). CE is called the contraction exponent because the difference of two orientations changes as

$$\tan\left[\frac{\sigma(T;\sigma_{10})-\sigma(T;\sigma_{20})}{2}\right] = \tan\left[\frac{\sigma_{10}-\sigma_{20}}{2}\right]\mathrm{e}^{-\mathrm{CE}[\sigma_{10},\sigma_{20};T]}$$

over the time interval. A natural extension of this concept is to the approach/separation of nearby orientations. In SWL the relationship

$$\epsilon(T;\sigma_0) = \epsilon(0;\sigma_0)\,\mathrm{e}^{-\mathrm{nCE}[\sigma_0;T]}, \tag{A 3}$$

is established, where the nearby contraction exponent

$$\frac{\mathrm{nCE}[\sigma_0;T]}{T} \equiv \frac{1}{T}\int_0^T [2Ge(t)\cos 2\sigma(t;\sigma_0)+G\gamma(t)\sin 2\sigma(t;\sigma_0)]\,\mathrm{d}t \tag{A 4}$$

distinguishes the approach or separation of two integral curves $\sigma(t;\sigma_0)$ and $\sigma(t;\sigma_0)+\epsilon(t)$, where $\epsilon(t)$ is small.

By computing the nearby contraction exponent for the full range of initial orientations $0 \leqslant \sigma_0 < 2\pi$, one may determine which integral curve(s) will attract the others. This procedure varies according to whether the flow parameters are periodic with period T (a special but important case), or not periodic; see SWL for the details.

The other degrees of freedom $\theta(t)$ and $\rho(t)$ also evolve in a way which is related to the nearby contraction exponent. To be specific, we have the discrete time maps

$$\tan\theta(T;\sigma_0,\theta_0) = \mathrm{e}^{-\frac{1}{2}\mathrm{nCE}[\sigma_0;T]}\tan\theta_0, \tag{A 5}$$

$$\rho(T;\rho_0,\sigma_0,\theta_0) = \rho_0\exp\left[\frac{T}{F+1}\left(\frac{1}{2}\frac{\mathrm{nCE}[\sigma_0;T]}{T}-\alpha\right)\right]\left[\frac{1+\mathrm{e}^{-\mathrm{nCE}[\sigma_0;T]}\tan^2\theta_0}{1+\tan^2\theta_0}\right]^{\frac{1}{2(F+1)}}. \tag{A 6}$$

These equations demonstrate that along attracting σ time traces, the axial vector of the microstructure moves into the plane of the two-dimensional flow, and that the particles may experience stretching provided that the history-dependent strong flow criterion

$$\frac{\mathrm{nCE}[\sigma_0;T]}{T} > 2\alpha \tag{A 7}$$

is satisfied.

Thus, through integration of (A 2*c*) over the time interval of interest and evaluation of the nearby contraction exponent, it is possible to characterize the dynamics of microstructure which follow the given particle path through the surrounding flow. One may identify the attracting and repelling orientations, and the evolution of the out-of-plane and stretch degrees of freedom of the particle directly from the nearby contraction exponent. Not surprisingly, the statistical distribution of microstructure conformation may also be related to the nearby contraction exponent, as we show in the next section.

A.3. *Solution for the distribution function*

We consider the model system, in which the Brownian diffusion is zero and $\mathrm{d}\sigma/\mathrm{d}t$ and $\mathrm{d}\theta/\mathrm{d}t$ are given by (A 2*a*, *b*). For rigid microstructure, the Lagrangian representation of the ODF is (2.13). For the microdynamical equations (A 2), equation (2.13) combined with (A 3)–(A 5), yields

$$f^*(t;\sigma_0,\theta_0) = f^*(0;\sigma_0,\theta_0)\,\mathrm{e}^{\frac{3}{2}\mathrm{nCE}[\sigma_0;t]}\left[\frac{1+\mathrm{e}^{-\mathrm{nCE}[\sigma_0;t]}\tan^2\theta_0}{1+\tan^2\theta_0}\right]^{\frac{3}{2}}. \tag{A 8}$$

Thus we see that the ODF evaluated on attracting (repelling) orientation time traces tends to increase (decrease). The Fokker–Planck equation need not be solved, as all the information is contained in the graph $(\sigma_0, \mathrm{nCE}[\sigma_0;T])$!

For the microdynamical equations (A 2), equation (2.10) yields the Lagrangian representation of the distribution function for stretchable, orientable particles:

$$\phi^*(t;\rho_0,\sigma_0,\theta_0) = \phi^*(0;\rho_0,\sigma_0,\theta_0)\left[\frac{1+\mathrm{e}^{-\mathrm{nCE}[\sigma_0;t]}\tan^2\theta_0}{1+\tan^2\theta_0}\right]^{\frac{3F}{2(1+F)}} \times \exp\left[\frac{3F}{2(1+F)}\mathrm{nCE}[\sigma_0;t]+\frac{3\alpha t}{F+1}\right]. \tag{A 9}$$

In the limit $F\to\infty$, we recover the rigid-particle result. Even in the full conformation evolution problem, we see that the distribution function evolves in a manner which is completely determined by the nearby contraction exponent.

A.4. *A specific flow example*

Finally, we give an example of an unsteady, spatially homogeneous flow for which the evolution equation for the ODF is integrable, both in the classical Eulerian framework and in the Lagrangian framework. At the end of the example, we show that the two approaches yield the same result.

The example flow is a time-dependent stagnation-point flow, with stream function $\psi(x,y,t) = xye(t)$, where $e(t)$ is the (unspecified) time-dependent elongation flow parameter. The shear and vorticity flow parameters are both zero in this example flow. We consider the ODF for rigid microstructure confined to the plane, for simplicity of exposition. Thus $f(\sigma,\theta,t) = f(\sigma,t)$ in this example.

In the classical formulation, the Fokker–Planck evolution equation for the ODF is

$$\frac{\partial f}{\partial t} - Ge(t)\sin(2\sigma)\frac{\partial f}{\partial \sigma} = 2Ge(t)\cos(2\sigma)f(\sigma,t). \tag{A 10}$$

We solve this classical equation by the method of characteristics. To find the characteristics one must solve the associated equations

$$\frac{\mathrm{d}\sigma}{-Ge(t)\sin(2\sigma)} = \frac{\mathrm{d}t}{1} = \frac{\mathrm{d}f}{2Ge(t)\cos(2\sigma)f(\sigma,t)}.$$

The first and second part yield the solution

$$u_1(\sigma,t) = \tan(\sigma)\exp\left[2G\int_0^t e(\tau)\,\mathrm{d}\tau\right] = c_1,$$

where c_1 is a constant; the first and third parts yield the solution

$$u_2(\sigma,f) = \frac{1}{f\sin(2\sigma)} = c_2$$

involving the constant c_2. The solution to the Fokker–Planck equation is $u_2 = \Phi(u_1)$, where the function Φ is determined from (non-degenerate) initial conditions. For microstructure in an isotropic initial state, we obtain the solution

$$f(\sigma,t) = \frac{\tan(\sigma)\exp\left[2G\int_0^t e(\tau)\,\mathrm{d}\tau\right]}{\pi\sin(2\sigma)\left(1+\tan^2(\sigma)\exp\left[4G\int_0^t e(\tau)\,\mathrm{d}\tau\right]\right)}. \tag{A 11}$$

Note that when $t = 0$, (A 11) gives the (isotropic) initial condition $f(\sigma, t=0) = 1/2\pi$. Moreover, one can see that if $e(t)$ is periodic with period T and zero mean, then the ODF returns to an isotropic state at $t = nT$, for positive integers n. This completes the solution of the classical Fokker–Planck equation for the ODF.

Now we solve the same problem, but we use the Lagrangian representation of the distribution function. To solve the specific example problem, we begin by calculating the integral curves $\sigma(t;\sigma_0)$ from the evolution equation for σ, (A 2*a*). This yields the result

$$\tan(\sigma(t;\sigma_0)) = \tan(\sigma_0)\exp\left[-2G\int_0^t e(\tau)\,\mathrm{d}\tau\right].$$

Because we know the time traces $\sigma(t;\sigma_0)$, we can compute the nearby contraction exponent from (A 4). This calculation is

$$\begin{aligned}
\mathrm{nCE}[\sigma_0;T] &= \int_0^T \left[2Ge(t)\frac{1-\tan^2(\sigma(t;\sigma_0))}{1+\tan^2(\sigma(t;\sigma_0))}\right]\mathrm{d}t \\
&= \int_0^T \left[2Ge(t)\frac{1-\tan^2(\sigma_0)\exp\left[-4G\int_0^t e(\tau)\,\mathrm{d}\tau\right]}{1+\tan^2(\sigma_0)\exp\left[-4G\int_0^t e(\tau)\,\mathrm{d}\tau\right]}\right]\mathrm{d}t \\
&= \log\left[\exp\left[-2G\int_0^T e(\tau)\,\mathrm{d}\tau\right]\frac{\exp\left[4G\int_0^T e(\tau)\,\mathrm{d}\tau\right]+\tan^2(\sigma_0)}{1+\tan^2(\sigma_0)}\right].
\end{aligned}$$

The Lagrangian representation for the (initially isotropic) ODF is given by equation (A 8), which in this case reads

$$f^*(t;\sigma_0) = \frac{1}{2\pi}\exp\left[-2G\int_0^t e(\tau)\,\mathrm{d}\tau\right]\frac{\exp\left[4G\int_0^t e(\tau)\,\mathrm{d}\tau\right]+\tan^2(\sigma_0)}{1+\tan^2(\sigma_0)}. \qquad \text{(A 12)}$$

To check the Lagrangian representation against the Eulerian representation for the ODF, we simply substitute $\sigma(t;\sigma_0)$ for σ in (A 11). One can easily show that the Lagrangian representation, (A 12), is recovered.

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