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A note about convected time derivatives for flows of complex fluids†

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We present a direct derivation of the typical time derivatives used in a continuum description of complex fluid flows, harnessing the principles of the kinematics of line elements. The evolution of the microstructural conformation tensor in a flow and the physical interpretation of different derivatives then follow naturally.

1 Introduction

In the study of the flow of viscoelastic or complex fluids it is necessary to utilize constitutive descriptions that capture the local deformations, *e.g.*, reorientations, stretching, compression, that give rise to the stress in a material element following the flow.^{1–3} Although perhaps simple to state in words, as in the previous sentence, the mathematical description is complex as the stress and material deformation, the latter expressed *via* the velocity gradients at the position of the fluid element, are second-rank tensorial quantities.

Most presentations of the mathematical descriptions begin by describing the need for material objectivity of a tensorial property when translating, rotating, and deforming with the local flow, then give (messy) details of covariant and contravariant characterizations of vectors and tensors. Thus, different constitutive descriptions, utilizing different time derivatives of tensorial quantities, *e.g.*, known as upper- or lower-convected derivatives, are obtained. These steps lead to the forms of the stress *versus* rate-of-strain relation known as the Oldroyd-B or Oldroyd-A constitutive description.^{1,3,4} It is reasonable to assume that most readers do not find such mathematical, Cartesian tensor presentations helpful for forming a physical picture of the underlying dynamics.

Alternatively, a physical picture of a dilute polymer solution can be gained by considering a dumbbell model.^{2,3,5,6} It is well known that a dumbbell model, written moving with flow, provides a description of the stress *versus* rate-of-strain relation, expressed in terms of the conformation tensor, that yields the upper-convected time derivative and the Oldroyd-B constitutive description.^{2,7–12} The dumbbell model also allows various physically plausible modifications.^{13–23}

Here, we provide a direct derivation of the typical time derivatives using the idea of kinematics of line elements. The physical interpretation, and even some of the steps, have overlap with the common derivation of the dumbbell description of the state of stress, but we are not otherwise aware of the style of presentation we provide here (see also ref. 24 Section 12.4 and ref. 8 Section 1.1). We believe that the physical picture associated with this derivation, focusing as it does on the material line kinematics, which should be familiar from a standard graduate mechanics course, is more transparent than existing alternatives as to the origin of terms in the time derivative associated with the material deformation. It is then straightforward to discuss other uses of the time derivatives in the development of constitutive descriptions.

II. Kinematics of a line element

A. The material derivative of a line element

Consider an Eulerian velocity field $\mathbf{u}(\mathbf{x}, t)$ at position $\mathbf{x}$ and time t . Recall the definition of the material derivative,

$$\frac{D}{Dt} = \frac{\partial}{\partial t} + \mathbf{u} \cdot \nabla. \quad (1)$$

It is shown in fluid dynamics textbooks that discuss kinematics (see, *e.g.*, ref. 25 pp. 131–132) that an infinitesimal line element

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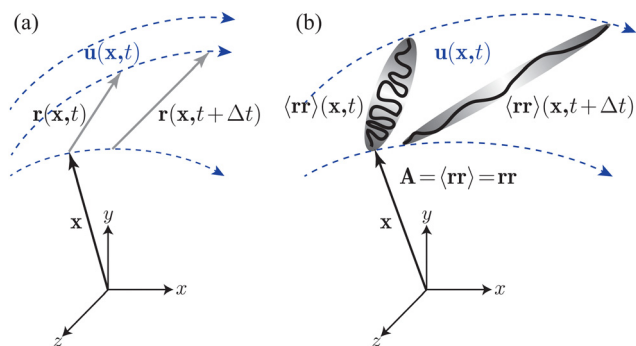


Fig. 1 Kinematics of (a) a material line element $\mathbf{r}$ and (b) conformation tensor $\mathbf{A} = \langle \mathbf{r}\mathbf{r} \rangle$.

$\mathbf{r}(\mathbf{x},t)$ moving with the local fluid velocity is reoriented and changed in length according to

$$\frac{D\mathbf{r}}{Dt} = \mathbf{r} \cdot \nabla \mathbf{u}, \quad (2)$$

as illustrated in Fig. 1(a). Next, we introduce the vectorial upper-convected time derivative representing the motion of a line element in the flow,

$$\overset{\nabla}{\mathbf{r}} \stackrel{\text{def}}{=} \frac{D\mathbf{r}}{Dt} - \mathbf{r} \cdot \nabla \mathbf{u}. \quad (3)$$

It follows that $\overset{\nabla}{\mathbf{r}} = \mathbf{0}$ means that a line element translates with the local fluid velocity, and rotates and stretches (or compresses) according to the local velocity gradient.⁹ In Section III, we apply this idea to second-rank tensors.

B. Lagrangian trajectories of a line element and a theorem of Cauchy on invariance

Before proceeding, we link the above ideas to a Lagrangian description, which makes clear that time derivatives are really expressing invariances associated with following a vectorial or tensorial quantity along the trajectory of a fluid particle. These features have a direct connection to a theorem of Cauchy, which is most simply understood using a Lagrangian description. In this subsection, we discuss the ideas for a vector field, and in Section IIIC we show how the same ideas apply to the conformation tensor (see also ref. 26).

To describe motion, we follow Lagrangian trajectories, $\mathbf{x} = \mathbf{x}(t;\mathbf{X})$, where $\mathbf{x}(0;\mathbf{X}) = \mathbf{X}$, with the label $\mathbf{X}$ indicating the initial position. A differential displacement $d\mathbf{X}$ is mapped to a differential displacement $d\mathbf{x}$ according to

$$d\mathbf{x} = d\mathbf{X} \cdot \mathbf{F}, \quad (4)$$

where $\mathbf{F}(t;\mathbf{X}) = \nabla_{\mathbf{x}} \mathbf{x}$ is the deformation gradient tensor; in index notation we write, $F_{ij} = \frac{\partial x_j}{\partial X_i}$. Taking the material time derivative, we write in a Lagrangian description $\frac{D\mathbf{F}}{Dt} = \nabla_{\mathbf{x}} \frac{\partial \mathbf{x}}{\partial t} = \nabla_{\mathbf{x}} \mathbf{u}^L$, where the Lagrangian velocity is $\mathbf{u}^L(t;\mathbf{X}) = \mathbf{u}(\mathbf{x},t)$. Using the chain rule, we can involve the Eulerian representation as $\nabla_{\mathbf{x}} \mathbf{u} = \mathbf{F} \cdot \nabla \mathbf{u}$. Therefore, the material derivative of $\mathbf{F}$ at fixed $\mathbf{X}$ (as in eqn (1)) is

$$\left. \frac{\partial \mathbf{F}}{\partial t} \right|_{\mathbf{X}} = \frac{D\mathbf{F}}{Dt} = \mathbf{F} \cdot \nabla \mathbf{u}. \quad (5)$$

Since by definition $\mathbf{F}^{-1} \cdot \mathbf{F} = \mathbf{I}$, where $\mathbf{I}$ is the identity tensor, then differentiating with respect to time means $\mathbf{F}^{-1} \cdot \frac{\partial \mathbf{F}}{\partial t} = -\frac{\partial \mathbf{F}^{-1}}{\partial t} \cdot \mathbf{F}$, which are two identities (at fixed $\mathbf{X}$) that will prove useful. The reader should note that in some presentations, eqn (5), because of a notational choice, appears with the transpose (see, e.g., ref. 6).

Given these geometric preliminaries, for the (infinitesimal) line element transport eqn (2), we use the identity tensor in the form $\mathbf{F}^{-1} \cdot \mathbf{F} = \mathbf{I}$ and write $(\mathbf{r}(\mathbf{x},t) = \mathbf{r}_L(t;\mathbf{X}))$,

$$\frac{D\mathbf{r}}{Dt} = \left. \frac{\partial \mathbf{r}_L}{\partial t} \right|_{\mathbf{X}} = \mathbf{r} \cdot \mathbf{F}^{-1} \cdot \underbrace{\mathbf{F} \cdot \nabla \mathbf{u}}_{\frac{\partial \mathbf{F}}{\partial t}} \quad (6)$$

$$= \mathbf{r}_L \cdot \mathbf{F}^{-1} \cdot \frac{\partial \mathbf{F}}{\partial t} = -\mathbf{r}_L \cdot \frac{\partial \mathbf{F}^{-1}}{\partial t} \cdot \mathbf{F}.$$

Taking the inner product on the right with $\mathbf{F}^{-1}$, we then have for fixed $\mathbf{X}$,

$$\frac{\partial \mathbf{r}_L}{\partial t} \cdot \mathbf{F}^{-1} + \mathbf{r}_L \cdot \frac{\partial \mathbf{F}^{-1}}{\partial t} = \mathbf{0} \quad \text{or} \quad \frac{\partial}{\partial t} (\mathbf{r}_L \cdot \mathbf{F}^{-1}) = \mathbf{0}. \quad (7)$$

We conclude that $\mathbf{r}_L \cdot \mathbf{F}^{-1} = \text{constant}$ following the fluid motion, i.e., it is an invariant. Since initially $\mathbf{F}(0) = \mathbf{I}$, then $\mathbf{r}_L \cdot \mathbf{F}^{-1} = \mathbf{r}_L(0)$, the initial vectorial line element. As $\mathbf{F}$ is a function of time that is in principle known, once a velocity field is determined, we conclude that the line element, as a Lagrangian object, evolves in time according to

$$\mathbf{r}_L(t;\mathbf{X}) = \mathbf{r}_L(0) \cdot \mathbf{F}(t;\mathbf{X}). \quad (8)$$

This result is known as Cauchy's theorem and yields how any initial line element is changed in magnitude and orientation by the time-dependent deformation tensor $\mathbf{F}$. The short derivation presented here may be helpful to readers to see the origin of this idea. Below we indicate the extension of these ideas to second-rank tensors, such as the conformation tensor and the convected time derivatives.

III. The material derivative of a second-rank tensor

A. Conformation tensor

Consider an end-to-end vector $\mathbf{R}$, whose magnitude at equilibrium is R_{eq} . We define the dimensionless conformation tensor $\mathbf{A} \stackrel{\text{def}}{=} \langle \mathbf{R}\mathbf{R} \rangle / R_{\text{eq}}^2 \stackrel{\text{def}}{=} \langle \mathbf{r}\mathbf{r} \rangle$, where $\langle \cdot \rangle$ denotes the ensemble average and the dyadic product $\mathbf{r}\mathbf{r}$ characterizes the state of deformation of a given microstructure, as sketched in Fig. 1(b); for example, $\mathbf{A} = \mathbf{I}$ represents an undeformed isotropic (equilibrium) state. For simplicity of the notation, we drop $\langle \cdot \rangle$ in the calculation that follows.

Proceeding formally to take the material derivative, and applying the product rule for differentiation, we obtain

$$\frac{D}{Dt} \mathbf{A} = \frac{D}{Dt} (\mathbf{r}\mathbf{r}) = \frac{D\mathbf{r}}{Dt} \mathbf{r} + \mathbf{r} \frac{D\mathbf{r}}{Dt}. \quad (9)$$

Using eqn (2) and the properties of the dyadic product, we find

$$\begin{aligned}\frac{D}{Dt}\mathbf{A} &= \frac{D}{Dt}(\mathbf{r}\mathbf{r}) = \mathbf{r} \cdot (\nabla\mathbf{u})\mathbf{r} + \mathbf{r}\mathbf{r} \cdot (\nabla\mathbf{u}) \\ &= (\nabla\mathbf{u})^T \cdot \mathbf{A} + \mathbf{A} \cdot \nabla\mathbf{u}.\end{aligned}\quad (10)$$

This identity is also recognized in ref. 24 Section 12.4. Therefore, we introduce a time derivative for the change of the tensor (dyadic product) deforming in a fluid flow:

$$\overset{\nabla}{\mathbf{A}} \stackrel{\text{def}}{=} \frac{D}{Dt}\mathbf{A} - (\nabla\mathbf{u})^T \cdot \mathbf{A} - \mathbf{A} \cdot \nabla\mathbf{u}.\quad (11)$$

The derivative $\overset{\nabla}{\mathbf{A}}$ is known as the upper-convected or contra-variant time derivative.¹ We conclude that $\overset{\nabla}{\mathbf{A}} = \mathbf{0}$ means that the conformation tensor deforms exactly as does the corresponding line elements in a flow. In contrast, $\overset{\nabla}{\mathbf{A}} \neq \mathbf{0}$ means that $\mathbf{A}$ does not deform comparable to the line elements in the flow.

To the best of our knowledge, Giesekus²⁷ was the first to use the idea of vectorial line element kinematics when modeling the stress response of dilute suspensions of chain-like elastic particles. The stress response, which is a second-rank tensor, then led to the upper-convected derivative for the conformation tensor.²⁸ We also note that Graham, in an excellent book, provides a derivation similar in spirit to what we have presented (see ref. 8 Section 1.1), though introduces basis vectors that are attached to the material line elements, so requires many more steps to arrive at the upper-convected derivative, eqn (11). Also, the physical interpretation of $\overset{\nabla}{\mathbf{A}} = \mathbf{0}$ is only given much later in the book.

B. Arbitrary dyadic products and second-rank tensors

As a further remark, we note that the kinematic approach here is valid for any dyadic product. Since an arbitrary second-rank tensor, such as the stress tensor, can be written as a linear combination of three dyadic products (see ref. 29 Sections 61–63), then it follows that the derivation of the time derivatives discussed above also applies to an arbitrary second-rank tensor. For example, if we define the dyadic product $\mathbf{B} = \mathbf{a}\mathbf{b}$, where $\mathbf{a}$ and $\mathbf{b}$ are vectors, then taking the material derivative and treating $\mathbf{a}$ and $\mathbf{b}$ as line elements (see eqn (2)), we obtain

$$\begin{aligned}\frac{D}{Dt}\mathbf{B} &= \frac{D}{Dt}(\mathbf{a}\mathbf{b}) = \frac{D\mathbf{a}}{Dt}\mathbf{b} + \mathbf{a}\frac{D\mathbf{b}}{Dt} = \mathbf{a} \cdot (\nabla\mathbf{u})\mathbf{b} \\ &+ \mathbf{a}\mathbf{b} \cdot (\nabla\mathbf{u}) = (\nabla\mathbf{u})^T \cdot \mathbf{B} + \mathbf{B} \cdot \nabla\mathbf{u},\end{aligned}\quad (12)$$

which is the form of the upper-convected derivative.

C. Invariance associated with convected time derivatives of the conformation tensor

Recall Section II on the kinematics of line elements, where the upper-convected time derivative $\overset{\nabla}{\mathbf{r}} = \mathbf{0}$ following the flow (eqn (3)) implies the invariant solution $\mathbf{r}_L \cdot \mathbf{F}^{-1} = \text{constant}$, from which the time variation of $\mathbf{r}_L(t; \mathbf{X})$ can be determined for a given flow or $\mathbf{F}$ (see eqn (8)). Following similar steps, one can take a Lagrangian view and integrate the equation $\overset{\nabla}{\mathbf{A}} = \frac{D\mathbf{A}}{Dt} - (\nabla\mathbf{u})^T \cdot \mathbf{A} - \mathbf{A} \cdot \nabla\mathbf{u} = \mathbf{0}$.

These ideas are known in the literature and the notation from Section II provides a compact way to proceed (see ESI†). In particular, using a Lagrangian notation with $\mathbf{A}(\mathbf{x}, t) = \mathbf{A}_L(t; \mathbf{X})$ one can show that $\overset{\nabla}{\mathbf{A}} = \mathbf{0}$ leads to

$$\frac{\partial}{\partial t}(\mathbf{F}^{-T} \cdot \mathbf{A}_L \cdot \mathbf{F}^{-1}) = \mathbf{0}.\quad (13)$$

It follows that $\mathbf{F}^{-T} \cdot \mathbf{A}_L \cdot \mathbf{F}^{-1} = \text{constant}$, following the fluid motion, *i.e.*, it is an invariant. Using the initial data, we obtain $\mathbf{F}^{-T} \cdot \mathbf{A}_L \cdot \mathbf{F}^{-1} = \mathbf{A}_L(0; \mathbf{X})$ since $\mathbf{F}(0) = \mathbf{I}$, which means $\mathbf{A}_L(t; \mathbf{X}) = \mathbf{F}^T \cdot \mathbf{A}_L(0; \mathbf{X}) \cdot \mathbf{F}$. In other words, the initial data and $\mathbf{F}$ determine the evolution of the conformation tensor in this case.

IV. The Jeffery equation for a particle in a linear flow

A. A line element that lags the flow

A slender extensible filament or rod with orientation and length $\mathbf{r}$, not necessarily infinitesimal, in a flow with vorticity $\boldsymbol{\omega} = \nabla \wedge \mathbf{u}$ and rate-of-strain tensor $\mathbf{E} = (1/2)(\nabla\mathbf{u} + (\nabla\mathbf{u})^T)$ is assumed to reorient approximately according to (see, *e.g.*, ref. 30–32)

$$\frac{D\mathbf{r}}{Dt} = \frac{1}{2}\boldsymbol{\omega} \wedge \mathbf{r} + \beta\mathbf{r} \cdot \mathbf{E},\quad (14)$$

where β is a constant. A rigid rod is unable to change length so eqn (14) is then modified to preserve length by adding an extra term $-\beta\mathbf{r} \cdot \mathbf{E} \cdot \mathbf{r}$ to the right-hand side, resulting in the Jeffery equation³³ (see Section V(vii)). Recalling the vorticity tensor $\boldsymbol{\Omega} = (1/2)(\nabla\mathbf{u} - (\nabla\mathbf{u})^T)$, where $\nabla\mathbf{u} = \mathbf{E} + \boldsymbol{\Omega}$ and $(1/2)\boldsymbol{\omega} \wedge \mathbf{r} = \mathbf{r} \cdot \boldsymbol{\Omega}$, eqn (14) can be written as

$$\frac{D\mathbf{r}}{Dt} = \mathbf{r} \cdot \nabla\mathbf{u} + (\beta - 1)\mathbf{r} \cdot \mathbf{E},\quad (15)$$

where the last term indicates that the straining motion produces non-affine motion of the filament, relative to following the motion and deformation of the fluid. Therefore, a filament behaves like a line element and follows the flow for $\beta = 1$, and lags the deformation for $\beta < 1$, as arises below.

B. A surface element

In the same way that the divergence of the velocity vector expresses the fractional rate of change of volume (V) of an infinitesimal fluid element in a flow (so is zero for an incompressible flow), and a line element $\mathbf{r}$ changes its length and orientation according to eqn (2), one can consider an infinitesimal (vectorial) surface element $\mathbf{S}$, *i.e.*, $V = \mathbf{r} \cdot \mathbf{S}$. It is an exercise to show that for an incompressible flow, $\nabla \cdot \mathbf{u} = 0$, then (see, *e.g.*, ref. 25 pp. 131–132 and ref. 34 and 35)

$$\frac{D\mathbf{S}}{Dt} = -(\nabla\mathbf{u}) \cdot \mathbf{S} = \frac{1}{2}\boldsymbol{\omega} \wedge \mathbf{S} - \mathbf{S} \cdot \mathbf{E}.\quad (16)$$

Hence, we see that an infinitesimal surface element satisfies eqn (14) with $\beta = -1$. We also note that, similar to eqn (3), we can introduce the vectorial lower-convected time derivative

$\overset{\Delta}{\mathbf{S}} \stackrel{\text{def}}{=} \frac{D\mathbf{S}}{Dt} + (\nabla\mathbf{u}) \cdot \mathbf{S}$. Thus, from eqn (16) it follows that an infinitesimal surface element satisfies $\overset{\Delta}{\mathbf{S}} = \mathbf{0}$.

V Non-affine motion of the conformation tensor

The derivation of eqn (11), and the steps that now follow, are the main contributions of this note focusing on the meaning of time derivatives for the kinematics of fluids with deformable microstructure. Substituting eqn (15) into eqn (9), we obtain

$$\frac{D}{Dt}\mathbf{A} = \frac{D}{Dt}(\mathbf{r}\mathbf{r}) = (\nabla\mathbf{u})^T \cdot \mathbf{A} + \mathbf{A} \cdot \nabla\mathbf{u} + (\beta - 1)(\mathbf{A} \cdot \mathbf{E} + \mathbf{E} \cdot \mathbf{A}), \quad (17)$$

or, using the definition of $\overset{\nabla}{\mathbf{A}}$,

$$\overset{\nabla}{\mathbf{A}} = \frac{D}{Dt}\mathbf{A} - (\nabla\mathbf{u})^T \cdot \mathbf{A} - \mathbf{A} \cdot \nabla\mathbf{u} = (\beta - 1)(\mathbf{A} \cdot \mathbf{E} + \mathbf{E} \cdot \mathbf{A}). \quad (18)$$

The right-hand side of eqn (18) yields non-affine motion that changes the response from purely following the deformation of the fluid, *i.e.*, we no longer have $\overset{\nabla}{\mathbf{A}} = \mathbf{0}$. We next make several observations.

(i) $\beta = 1$: we observe that $\beta = 1$ gives back the case of a line element following the flow, *i.e.*, the upper-convected derivative vanishes, *i.e.*, $\overset{\nabla}{\mathbf{A}} = \mathbf{0}$.

(ii) $\beta = 0$: using the relation $\mathbf{\Omega} = \nabla\mathbf{u} - \mathbf{E}$, we obtain

$$\overset{\circ}{\mathbf{A}} \stackrel{\text{def}}{=} \frac{D}{Dt}\mathbf{A} - \mathbf{\Omega}^T \cdot \mathbf{A} - \mathbf{A} \cdot \mathbf{\Omega} = \mathbf{0}. \quad (19)$$

$\overset{\circ}{\mathbf{A}}$ is referred to as the (Jaumann) co-rotational derivative,^{36,37} representing the rate of change of a fluid element advected with the flow and rotating with the local vorticity, *e.g.*, see eqn (14).

(iii) $\beta = -1$: in this case, using the definition for the rate-of-strain tensor $\mathbf{E} = (1/2)(\nabla\mathbf{u} + (\nabla\mathbf{u})^T)$, eqn (17) becomes

$$\begin{aligned} \frac{D}{Dt}\mathbf{A} &= (\nabla\mathbf{u})^T \cdot \mathbf{A} + \mathbf{A} \cdot \nabla\mathbf{u} - 2(\mathbf{A} \cdot \mathbf{E} + \mathbf{E} \cdot \mathbf{A}) \\ &= -(\nabla\mathbf{u}) \cdot \mathbf{A} - \mathbf{A} \cdot (\nabla\mathbf{u})^T. \end{aligned} \quad (20)$$

Similar to eqn (11), we can introduce the lower-convected or covariant time derivative (see, *e.g.*, ref. 1, 3 and 9):

$$\overset{\Delta}{\mathbf{A}} \stackrel{\text{def}}{=} \frac{D}{Dt}\mathbf{A} + (\nabla\mathbf{u}) \cdot \mathbf{A} + \mathbf{A} \cdot (\nabla\mathbf{u})^T. \quad (21)$$

(iv) $\beta = \text{const}$: for a constant value of β , using eqn (17), we can introduce the Gordon-Schowalter convected time derivative $\overset{\square}{\mathbf{A}}$ (see, *e.g.*, ref. 3, 9 and 32),

$$\overset{\square}{\mathbf{A}} \stackrel{\text{def}}{=} \frac{D}{Dt}\mathbf{A} - (\nabla\mathbf{u})^T \cdot \mathbf{A} - \mathbf{A} \cdot \nabla\mathbf{u} - (\beta - 1)(\mathbf{A} \cdot \mathbf{E} + \mathbf{E} \cdot \mathbf{A}). \quad (22)$$

(v) We note that, in practice, β , appearing in eqn (22), can be a function of the conformation tensor $\mathbf{A}$. In fact, for a dumbbell

model, Hinch¹⁴ and Phan-Thien *et al.*¹⁹ pointed out that β depends on the trace of $\mathbf{A}$ via

$$\beta = \frac{\text{tr } \mathbf{A} - a}{\text{tr } \mathbf{A} + b}, \quad (23)$$

where $\text{tr } \mathbf{A}$ represents the square of the dumbbell extension and a and b are constants (see also ref. 9 and 38).

(vi) An interesting property of the conformation tensor $\mathbf{A}$ that satisfies $\overset{\nabla}{\mathbf{A}} = \mathbf{0}$ (upper-convected derivative) is that its inverse $\mathbf{A}^{-1}$ satisfies $(\mathbf{A}^{-1})^\Delta = \mathbf{0}$ (lower-convected derivative). This result can be readily shown by left and right multiplying $\overset{\nabla}{\mathbf{A}} = \mathbf{0}$ by $\mathbf{A}^{-1}$ and using the identity $\frac{\partial \mathbf{A}^{-1}}{\partial t} = -\mathbf{A}^{-1} \cdot \frac{\partial \mathbf{A}}{\partial t} \cdot \mathbf{A}^{-1}$. Similarly, $\overset{\circ}{\mathbf{A}} = \mathbf{0}$ (co-rotational derivative) implies that $(\mathbf{A}^{-1})^\circ = \mathbf{0}$. In fact, this symmetric property is more general and can be extended to the Gordon-Schowalter convected time derivative $\overset{\square}{\mathbf{A}}$, that if $\overset{\square}{\mathbf{A}}_\beta = \mathbf{0}$ for some β , then $(\mathbf{A}^{-1})_{-\beta}^\square = \mathbf{0}$ with $-\beta$.

(vii) For a rigid inextensible rod, we modify eqn (14) to preserve the length of the rod by including an additional term $-\beta\mathbf{r} \cdot \mathbf{E} \cdot \mathbf{r}$ on the right-hand side. Thus, we have the Jeffrey equation for the motion of a rigid ellipsoid,³³

$$\frac{D\mathbf{r}}{Dt} = \frac{1}{2}\boldsymbol{\omega} \wedge \mathbf{r} + \beta(\mathbf{r} \cdot \mathbf{E} - \mathbf{r} \cdot \mathbf{E} \cdot \mathbf{r}), \quad (24)$$

which also can be written as

$$\frac{D\mathbf{r}}{Dt} = \mathbf{r} \cdot \nabla\mathbf{u} + (\beta - 1)\mathbf{r} \cdot \mathbf{E} - \beta\mathbf{r} \cdot \mathbf{E} \cdot \mathbf{r}. \quad (25)$$

Here, the parameter β is $\beta = (L^2 - 1)/(L^2 + 1)$, where L is the length-to-radius aspect ratio of an axisymmetric ellipsoid.³³ Substituting eqn (24) into eqn (9), we obtain the time-rate-of-change of the corresponding conformation tensor,

$$\begin{aligned} \frac{D}{Dt}\mathbf{A} &= \frac{D}{Dt}(\mathbf{r}\mathbf{r}) = (\nabla\mathbf{u})^T \cdot \mathbf{A} + \mathbf{A} \cdot \nabla\mathbf{u} \\ &\quad + (\beta - 1)(\mathbf{A} \cdot \mathbf{E} + \mathbf{E} \cdot \mathbf{A}) - 2\beta\mathbf{A} \cdot \mathbf{E} \cdot \mathbf{A}. \end{aligned} \quad (26)$$

Using the definition of $\overset{\nabla}{\mathbf{A}}$ yields,

$$\begin{aligned} \overset{\nabla}{\mathbf{A}} &= \frac{D}{Dt}\mathbf{A} - (\nabla\mathbf{u})^T \cdot \mathbf{A} - \mathbf{A} \cdot \nabla\mathbf{u} \\ &= (\beta - 1)(\mathbf{A} \cdot \mathbf{E} + \mathbf{E} \cdot \mathbf{A}) - 2\beta\mathbf{A} \cdot \mathbf{E} \cdot \mathbf{A}. \end{aligned} \quad (27)$$

Again, we see how β causes the conformation tensor to evolve differently than a “passive” structure simply following the fluid flow, $\overset{\nabla}{\mathbf{A}} \neq \mathbf{0}$. As noted by Hinch and Harlen,⁹ fibers, long thin rods, and elongated particles with $L \gg 1$ behave like line elements (of zero thickness) for which $\beta \approx 1$, so that the upper-convected derivative is appropriate. On the other hand, flattened particles with $L \ll 1$ are assumed to behave like area elements, so that $\beta \approx -1$, and thus, the lower-convected derivative is appropriate; however, see the recent analysis of Eggers *et al.*³⁵ showing that the dynamics in this limit is more complex.

(viii) As a particular case of the time derivative in eqn (27), consider the case with $\beta = 1$. We refer to this time derivative as the constrained upper-convected time derivative, given as

$$\overset{\nabla}{\mathbf{A}} + 2\mathbf{A} \cdot \mathbf{E} \cdot \mathbf{A} = \frac{D}{Dt}\mathbf{A} - (\nabla\mathbf{u})^T \cdot \mathbf{A} - \mathbf{A} \cdot \nabla\mathbf{u} + 2\mathbf{A} \cdot \mathbf{E} \cdot \mathbf{A} = \mathbf{0}. \quad (28)$$

This time derivative arises, for example, in the so-called quadratic closure for the Doi-Onsager rod theory as shown in Weady *et al.*³⁹ and in the sharply aligned case of the Doi-Onsager rod theory.⁴⁰ It is possible to extend the ideas introduced above (Sections II and IIIC) for identifying invariances following the fluid motion to the special case of $\overset{\nabla}{\mathbf{A}} \neq \mathbf{0}$ when $\overset{\nabla}{\mathbf{A}} = -2\mathbf{A} \cdot \mathbf{E} \cdot \mathbf{A}$.

For example, using a Lagrangian notation with $\mathbf{A}(\mathbf{x}, t) = \mathbf{A}_L(t; \mathbf{X})$ one can show (see ESI†) that eqn (28) leads to

$$\frac{\partial}{\partial t}(\mathbf{F}^{-T} \cdot \mathbf{A}_L \cdot \mathbf{F}^{-1}) = -(\mathbf{F}^{-T} \cdot \mathbf{A}_L \cdot \mathbf{F}^{-1}) \cdot \frac{\partial}{\partial t}(\mathbf{F} \cdot \mathbf{F}^T) \cdot (\mathbf{F}^{-T} \cdot \mathbf{A}_L \cdot \mathbf{F}^{-1}), \quad (29)$$

or

$$\frac{\partial \mathbf{G}_L}{\partial t} = -\mathbf{G}_L \cdot \frac{\partial}{\partial t}(\mathbf{F} \cdot \mathbf{F}^T) \cdot \mathbf{G}_L \quad \text{with} \quad \mathbf{G}_L = \mathbf{F}^{-T} \cdot \mathbf{A}_L \cdot \mathbf{F}^{-1}. \quad (30)$$

Under the assumption that $\mathbf{G}_L$ (in fact, $\mathbf{A}_L$) is invertible and using the identities $\mathbf{G}_L^{-1} = \mathbf{F} \cdot \mathbf{A}_L^{-1} \cdot \mathbf{F}^T$ and $\frac{\partial \mathbf{G}_L}{\partial t} = -\mathbf{G}_L \cdot \frac{\partial \mathbf{G}_L^{-1}}{\partial t}$, from eqn (30) it follows that

$$\frac{\partial \mathbf{G}_L^{-1}}{\partial t} = \frac{\partial}{\partial t}(\mathbf{F} \cdot \mathbf{F}^T) \quad \text{or} \quad \frac{\partial}{\partial t}(\mathbf{G}_L^{-1} - \mathbf{F} \cdot \mathbf{F}^T) = \frac{\partial}{\partial t}(\mathbf{F} \cdot \mathbf{A}_L^{-1} \cdot \mathbf{F}^T - \mathbf{F} \cdot \mathbf{F}^T) = \mathbf{0}. \quad (31)$$

It follows that $\mathbf{F} \cdot \mathbf{A}_L^{-1} \cdot \mathbf{F}^T - \mathbf{F} \cdot \mathbf{F}^T = \text{constant}$, following the fluid motion, *i.e.*, it is an invariant. Using the initial data, we obtain $\mathbf{F} \cdot \mathbf{A}_L^{-1} \cdot \mathbf{F}^T - \mathbf{F} \cdot \mathbf{F}^T = \mathbf{A}_L^{-1}(0; \mathbf{X}) - \mathbf{I}$ since $\mathbf{F}(0) = \mathbf{I}$, which means $\mathbf{A}_L^{-1}(t; \mathbf{X}) = \mathbf{I} - \mathbf{F}^{-1} \cdot \mathbf{F}^{-T} + \mathbf{F}^{-1} \cdot \mathbf{A}_L^{-1}(0; \mathbf{X}) \cdot \mathbf{F}^{-T}$. Note that if $\mathbf{A}_L(0; \mathbf{X}) = \mathbf{I}$, then $\mathbf{A}_L = \mathbf{I}$, which is a steady-state solution of eqn (28).

By analogy to eqn (30) for the constrained upper-convected time derivative, one can show (see ESI†) for the constrained lower-convected time derivative, $\overset{\Delta}{\mathbf{A}} = 2\mathbf{A} \cdot \mathbf{E} \cdot \mathbf{A}$, that

$$\frac{\partial}{\partial t}(\mathbf{H}_L^{-1} - \mathbf{F}^{-T} \cdot \mathbf{F}^{-1}) = \frac{\partial}{\partial t}(\mathbf{F}^{-T} \cdot \mathbf{A}_L^{-1} \cdot \mathbf{F}^{-1} - \mathbf{F}^{-T} \cdot \mathbf{F}^{-1}) = \mathbf{0} \quad \text{with} \quad \mathbf{H}_L = \mathbf{F} \cdot \mathbf{A}_L \cdot \mathbf{F}^T.$$

Thus, $\mathbf{F}^{-T} \cdot \mathbf{A}_L^{-1} \cdot \mathbf{F}^{-1} - \mathbf{F}^{-T} \cdot \mathbf{F}^{-1}$ is an invariant following the motion.

For the constrained upper-/lower-convected derivatives, we have conserved quantities $\mathbf{G}_L = \mathbf{F}^{-T} \cdot \mathbf{A}_L \cdot \mathbf{F}^{-1}$ and $\mathbf{H}_L = \mathbf{F} \cdot \mathbf{A}_L \cdot \mathbf{F}^T$, respectively, whose invariant structure is essentially through

inverses of each other. The co-rotational derivative arises from adding these two transport derivatives together, thus suggesting that finding an invariance for the co-rotational time derivative is challenging, if possible at all.

VI. Connecting conformation and its change to stress

A. The continuity and Cauchy momentum equations with additional microstructural stresses

Although the focus of this note is on the time derivatives, it is natural to now ask about the physical effects that exert forces on suspended filaments, which cause additional changes to these material derivatives and contribute to stress in the fluid. Describing the incompressible hydrodynamics of a complex fluid generally requires incorporating two new equations for the conformation tensor in addition to the continuity and Cauchy momentum equations. The first equation indicates how changes in the conformation give rise to stress in the fluid, whereas the second equation connects the evolution of the conformation to the gradients in the velocity field. As a specific example, consider the case of a viscoelastic dilute polymer solution, whose fluid motion is governed by the continuity equation and Cauchy momentum equations,

$$\nabla \cdot \mathbf{u} = 0 \quad \text{and} \quad \rho \frac{D\mathbf{u}}{Dt} = \nabla \cdot \boldsymbol{\sigma}, \quad (33)$$

where ρ is fluid density and $\boldsymbol{\sigma}$ is the stress tensor given by

$$\boldsymbol{\sigma} = -p\mathbf{I} + 2\eta_s\mathbf{E} + \boldsymbol{\tau}_p. \quad (34)$$

The first term on the right-hand side of eqn (34) is the pressure contribution, the second term is the viscous stress contribution of Newtonian solvent with a constant viscosity η_s , and the last term, $\boldsymbol{\tau}_p$, is the polymer contribution to the stress tensor. The latter contribution arises due to the response of polymer chains to fluid flow so that their microstructure deviates from the equilibrium state and deforms, $\mathbf{A} \neq \mathbf{I}$. This deformation results in a polymer contribution to the stress tensor, which, in the simplest Hookean case and affine motion, can be expressed *via* $\mathbf{A}$ as (see, *e.g.*, ref. 9–11 and 26)

$$\boldsymbol{\tau}_p = G(\mathbf{A})(\mathbf{A} - \mathbf{I}), \quad (35)$$

where $G(\mathbf{A})$ is the elastic modulus, which may be a function of $\mathbf{A}$. When non-affine effects are present, one should be cautious while identifying the proper form for the relation between the polymer contribution to the stress tensor and microstructure. We refer the reader to ref. 41 Ch. 8 for further discussion and the connection to irreversible dynamics associated with non-affine motion in contrast to the reversible, Hamiltonian dynamics corresponding to the affine one.

Equations (33)–(35) show the coupling between the fluid velocity and deformation of microstructure. Understanding this two-way coupling also requires determining the evolution equation for the conformation changes that describes how polymer chains deform in the flow.

If we treat polymer chains like line elements transported and freely deformed by the flow, it implies $\overset{\nabla}{\mathbf{A}} = \mathbf{0}$, according to eqn (11). However, polymer chains have elastic features with an inherent restoring mechanism opposing stretching/compression in the flow and causing relaxation to an equilibrium (coiled) state in the absence of flow. In general, this relaxation process may be characterized by several time scales; we denote by λ the longest relaxation time of the polymers. Thus, for affine motion ($\beta = 1$), accounting for the relaxation to an undeformed state on a time scale λ , the evolution equation for $\mathbf{A}$ can be written as

$$\overset{\nabla}{\mathbf{A}} = -\frac{1}{\lambda(\mathbf{A})}(\mathbf{A} - \mathbf{I}), \quad (36)$$

which, for a constant λ , is known as the Oldroyd-B constitutive equation.^{9,10} We note that $\lambda(\mathbf{A})$ may be a function of $\mathbf{A}$, as was first elucidated by de Gennes¹⁵ and Hinch.¹³

From eqn (36), it follows that when the observation time t_{obs} is much shorter than the relaxation time λ , $t_{\text{obs}} \ll \lambda$, $\overset{\nabla}{\mathbf{A}} \approx \mathbf{0}$ and thus, polymer chains behave like line elements that are transported and deformed by the flow without relaxing. The ratio λ/t_{obs} is known as the Deborah number, $\text{De} \stackrel{\text{def}}{=} \lambda/t_{\text{obs}}$,^{1,2} which follows from eqn (36). In many cases, the observation time t_{obs} is based on the convective time scale L_c/u_c , where L_c is the characteristic length scale and u_c is the characteristic velocity, so that the Deborah number is $\text{De} = \lambda u_c/L_c$. Furthermore, as many flows are characterized by a shear rate, $\dot{\gamma}$, Eqn (36) also naturally introduces the Weissenberg number, $\text{Wi} = \lambda\dot{\gamma}$. While, for brevity, we have considered only the Oldroyd-B model, the approach illustrated here allows various physically plausible modifications similar to the modeling of dumbbells.^{9,13–23} Finally, we refer the reader to the recent work by Eggers *et al.*³⁵ on flat elastic particles in a Newtonian fluid that, by analogy with dumbbell models, were modeled as three beads connected by nonlinear springs. In this work, a lower-convected time derivative naturally arises as part of the constitutive equation.

B. Lagrangian integration of the Oldroyd-B constitutive equation

The ideas introduced above (Sections II and IIIC) for identifying invariances following the fluid motion can also be applied to the evolution equation for the conformation tensor when elastic stresses are included. In this case, the conformation tensor evolves according to the deformation tensor $\mathbf{F}$ and the time constant λ_0 . Again, the detailed steps can be found in the literature (see, *e.g.*, ref. 1 pp. 431–432),^{26,42} and (ESI[†]).

For example, we can consider the Oldroyd-B constitutive equation (eqn (36)), $\overset{\nabla}{\mathbf{A}} = -\lambda_0^{-1}(\mathbf{A} - \mathbf{I})$, where λ_0 is a constant relaxation time. Again, we use the notation $\mathbf{A}(\mathbf{x}, t) = \mathbf{A}_L(t; \mathbf{X})$. The equation for $\overset{\nabla}{\mathbf{A}}$ can be rearranged to find an ordinary differential equation for $\mathbf{F}^{-T} \cdot \mathbf{A}_L \cdot \mathbf{F}^{-1}$ with a time-dependent forcing,

$$\frac{\partial}{\partial t}(\mathbf{F}^{-T} \cdot \mathbf{A}_L \cdot \mathbf{F}^{-1}) + \frac{1}{\lambda_0} \mathbf{F}^{-T} \cdot \mathbf{A}_L \cdot \mathbf{F}^{-1} = \frac{1}{\lambda_0} \mathbf{F}^{-T} \cdot \mathbf{F}^{-1}. \quad (37)$$

This equation is solved with initial data (suppressing dependence on $\mathbf{X}$) $\mathbf{F}^{-T} \cdot \mathbf{A}_L \cdot \mathbf{F}^{-1} = \mathbf{A}_L(0)$ since $\mathbf{F}(0) = \mathbf{I}$. Therefore, integrating, we obtain (see also *e.g.* ref. 26 and 42),

$$\begin{aligned} \mathbf{A}_L(t) &= e^{-t/\lambda_0} \mathbf{F}^T(t) \cdot \mathbf{A}_L(0) \cdot \mathbf{F}(t) \\ &+ \frac{1}{\lambda_0} \mathbf{F}^T(t) \cdot \int_0^t \mathbf{F}^{-T}(t') \cdot \mathbf{F}^{-1}(t') e^{-(t-t')/\lambda_0} dt' \cdot \mathbf{F}(t). \end{aligned} \quad (38)$$

Thus, we observe that the flow history matters and the material is always relaxing, relative to the local flow features, according to an exponential in time. The reader is referred to ref. 26 for more discussion and the connection of these ideas to the theory of elastic solids.

VII. Concluding remarks

The approach illustrated here for the derivation of the typical time derivatives of the conformation tensor, eqn (11) and (18), commonly used in the study of the flow of complex fluids, is the main contribution of this note. Our derivation utilizes the idea of the kinematics of line elements, and we believe that the physical picture associated with this derivation may be useful in providing insight and developing intuition when using these time derivatives and the corresponding constitutive equations. For completeness, in Section VI, we briefly discussed the connection of the flow kinematics to the conformation changes that produce stresses. In several places in the discussion, we highlighted how a Lagrangian view of the kinematics allows one to identify invariances following the flow, or, in the case of elastic stresses, how the conformation tensor relaxes in a time-varying way relative to the stretch expected due to the flow alone.

Conflicts of interest

There are no conflicts to declare.

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