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Resolvent operator method for solving rheological equations of state

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Fano's tetradic representation of Liouville–Neumann operators is used as the basis of a systematic procedure for solving an important class of rheological equations of state. This procedure is complementary to the techniques which Bird and his co-workers have used in their studies of complicated rheological models. To illustrate how our method works it is applied to a rheological model of the Maxwell–Oldroyd type. Explicit formulas are derived for the stress tensors associated with a number of time-independent, homogeneous flows.

I. INTRODUCTION

It is shown in this paper that Fano's[1] tetradic representation of Liouville–Neumann operators can be adapted to a number of rheological equations of state containing corotational and codeformational derivatives.[2,3] Consequently, these equations can be recast as inhomogeneous Liouville equations (with tetradic operators), the solutions of which can then be constructed using well-established methods.

We begin the following section with an examination of the kinetic theory of the elastic dumbbell model for flexible polymers. The purpose of this is threefold: (1) to identify the rheological equation of state arising from this model; (2) to show that this equation can be written in the form of a Liouville equation; and (3) to illustrate the tetradic representation of the associated Liouville operator. The remainder of Sec. II is devoted to presenting several more general rheological equations of state involving corotational and codeformational derivatives and to illustrating that these equations can be recast in Liouville form.

In the third section the conventional resolvent operator formalism is used to solve the rheological equation of state specific to the elastic dumbbell model for a polymer solution. This produces a formula for the fluid stress tensor expressed in terms of the velocity gradient tensor $\boldsymbol{\kappa} = (\nabla \mathbf{v})^T$. In addition to a general perturbation series solution which is applicable for arbitrary $\boldsymbol{\kappa}$, two special cases (potential flow and simple shear) are examined for which the stress tensor can be expressed in closed, analytic form. In the final section a perturbative solution is constructed as a power series in the vorticity tensor $\boldsymbol{\omega} = \boldsymbol{\kappa}^T - \boldsymbol{\kappa}$.

The present investigation is limited to homogeneous, time-independent flows. Less restrictive conditions will be treated in subsequent communications.

II. TETRADIC REPRESENTATION OF LIOUVILLE OPERATORS ASSOCIATED WITH RHEOLOGICAL EQUATIONS OF STATE

A. Elastic dumbbell model (Ref. 4)

This model for a polymer solution consists of n identical dumbbells per unit volume suspended in a Newtonian solvent. The symbol $\mathbf{R}$ is used to denote the vector extending from one end to the other of a representative dumbbell. H is the force constant of the Hookean spring which connects the two. The polymer contribution to the fluid stress tensor is given by the expression

$$\tau_p = -nH\Xi + nk_BT\delta, \tag{1}$$

where k_B is the Boltzmann constant and T the absolute temperature. δ is the unit tensor and $\Xi = \langle \mathbf{RR} \rangle$ the ensemble, orientation average of the dyad $\mathbf{RR}$. The trace of Ξ is the mean-square end-to-end length of these polymer molecules.

When the solvent velocity field is homogeneous [$\boldsymbol{\kappa} = (\nabla \mathbf{v})^T$ independent of position] Ξ depends only on time and is governed by the equation of motion

$$\frac{d}{dt}\Xi - (\boldsymbol{\kappa}\cdot\Xi + \Xi\cdot\boldsymbol{\kappa}^T) + \frac{4H}{\zeta}\Xi = \frac{4k_BT}{\zeta}\delta, \tag{2}$$

the Cartesian components of which are

$$\frac{d}{dt}\Xi_{ij} - (\kappa_{im}\Xi_{mj} + \Xi_{im}\kappa_{jm}) + \frac{4H}{\zeta}\Xi_{ij} = \frac{4k_BT}{\zeta}\delta_{ij}. \tag{3}$$

Here ζ is the friction coefficient characteristic of the Stokesean, hydrodynamic drag which acts on the dumbbells.

Equations (2) and (3) can be written in the alternative forms

$$\frac{d}{dt}\Xi - \mathcal{L}\Xi = \frac{4k_BT}{\zeta}\delta \tag{4}$$

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and

$$\frac{d}{dt}\Xi_{ij} - \mathcal{L}_{ij,mn}\Xi_{mn} = (4k_BT/\zeta)\delta_{ij}, \tag{5}$$

respectively, with $\mathcal{L} = \mathcal{L}_0 + \delta\mathcal{L}$ denoting a "Liouville operator" with the tetradic representation

$$(\mathcal{L}_0)_{ij,mn} = -\lambda_H^{-1}(\delta \otimes \delta)_{ij,mn} = -\lambda_H^{-1}\delta_{im}\delta_{jn}, \tag{6}$$

$$(\delta\mathcal{L})_{ij,mn} = (\kappa \otimes \delta + \delta \otimes \kappa)_{ij,mn} = \kappa_{im}\delta_{jn} + \delta_{im}\kappa_{jn}. \tag{7}$$

The quantity $\lambda_H = \zeta/4H$ is a time constant characteristic of the Hookean dumbbell.

B. Rheological equations of state

By invoking the connection [Eq. (1)] between Ξ and τ_p, we can rewrite the inhomogeneous Liouville equation (4) as a "rheological equation of state,"

$$\frac{d}{dt}\tau_p - \mathcal{L}\tau_p = -nk_BT\dot{\gamma} \tag{8}$$

relating the stress tensor of the Hookean dumbbell model to the (symmetric) rate-of-strain tensor $\dot{\gamma} = \kappa + \kappa^T$. Rheological model building[4] often involves equations which can be cast into the same general form as Eq. (8). We consider three examples of this, limiting our attention to homogeneous, time-independent flow fields.

1. Corotational equations

This category consists of rheological equations of state having the structure

$$\tau + \lambda\frac{\mathcal{D}}{\mathcal{D}t}\tau = \mathcal{F}(\dot{\gamma}). \tag{9}$$

Here $\mathcal{F}(\dot{\gamma})$ is some tensor valued function of $\dot{\gamma}$ (equal to $-\eta\dot{\gamma}$ in the Fromm model[5]) and

$$\frac{\mathcal{D}}{\mathcal{D}t}\tau \equiv \frac{D}{Dt}\tau + \tfrac{1}{2}(\omega\cdot\tau - \tau\cdot\omega) \tag{10}$$

is the so-called corotational or Jaumann derivative. $D/Dt = \partial/\partial t + \mathbf{v}\cdot\nabla$ is the substantial or material derivative and $\omega = \kappa^T - \kappa$ is the (antisymmetric) vorticity tensor.

2. Codeformational equations (Ref. 6)

There are two types of equations included in this category, the first of which is

$$\tau + \lambda\tau^{(1)} = \mathcal{F}(\dot{\gamma}) \quad : \quad \text{CONTRAVARIANT}. \tag{11}$$

The symbol $\tau^{(1)}$, defined by

$$\tau^{(1)} \equiv \frac{\mathcal{D}}{\mathcal{D}t}\tau + \tfrac{1}{2}(\dot{\gamma}\cdot\tau + \tau\cdot\dot{\gamma}) = \frac{D}{Dt}\tau + (\kappa^T\cdot\tau + \tau\cdot\kappa) \tag{12}$$

is called the contravariant form of the (Oldroyd) codeformational derivative, sometimes written $\delta\tau/\delta t$. The second type is

$$\tau + \lambda\tau_{(1)} = \mathcal{F}(\dot{\gamma}) \quad : \quad \text{COVARIANT} \tag{13}$$

and involves the covariant form of the codeformational derivative defined by

$$\tau_{(1)} \equiv \frac{\mathcal{D}}{\mathcal{D}t}\tau - \tfrac{1}{2}(\dot{\gamma}\cdot\tau + \tau\cdot\dot{\gamma}) = \frac{D}{Dt}\tau - (\kappa\cdot\tau + \tau\cdot\kappa^T). \tag{14}$$

It is easily demonstrated that (for homogeneous, time-independent flows) the three rheological equations of state (9), (11), and (13) can be written in a common form,

$$\frac{d}{dt}\tau - \mathcal{L}\tau = \lambda^{-1}\mathcal{F}(\dot{\gamma}), \tag{15}$$

analogous to that [Eq. (8)] for elastic dumbbells. The Liouville operator $\mathcal{L} = \mathcal{L}_0 + \delta\mathcal{L}$ appearing in this equation is the sum of a part, $\mathcal{L}_0 = -\lambda^{-1}(\delta\otimes\delta)$, which is identical to that [cf. (Eq. (6)] which occurred in the theory of elastic dumbbells, and a second part $\delta\mathcal{L}$ which differs from case to case. The tetradic representatives of these three operators are the following:

$$\delta\mathcal{L} = \begin{cases} -\tfrac{1}{2}(\omega\otimes\delta + \delta\otimes\omega)\text{:} & \text{corotational [Eq. (9)]} \\ -(\kappa^T\otimes\delta + \delta\otimes\kappa^T)\text{:} & \text{contravariant, codeformational [Eq. (11)]} \\ +(\kappa\otimes\delta + \delta\otimes\kappa)\text{:} & \text{covariant, codeformational [Eq. (13)].} \end{cases} \tag{16}$$

The third of these is the same as the operator which we encountered [cf. Eq. (7)] in the theory of elastic dumbbells. Consequently, the rheological equation of state for this model falls into the covariant–codeformational category with the choice $\mathcal{F}(\dot{\gamma}) = -nk_BT\dot{\gamma}$ and with λ equal to the time constant λ_H.

III. SOLUTIONS OF THE RHEOLOGICAL EQUATIONS OF STATE

It has been demonstrated in the preceding section that many rheological equations of state can be recast as inhomogeneous Liouville equations. This observation opens the way to solving these equations of state by using well-established methods of solution for the Liouville equation. Here we illustrate the use of one of the simplest of these methods, based on the resolvent operator formalism.

Because of the close structural similarities among the various corotational and codeformational equations of state, we confine our attention to a single example, namely, the specific case of the covariant–codeformational equation associated with the elastic-dumbbell model for a polymer solution. The analysis of this model will be based on the equation of motion (4) for the tensor $\Xi(t)$ rather than on the equivalent rheological equation of state (8).

We begin by introducing the Laplace transform

$$\Xi(s) = \int_0^\infty dt e^{-st}\Xi(t), \text{ Re } s > 0 \tag{17}$$

and replacing Eq. (4) with the transformed equation

$$\Xi(s) = (s - \mathcal{L})^{-1}B(s)\delta, \tag{18}$$

where

$$B(s) = (k_BT/H)\left(1 + \frac{1}{\lambda_H s}\right). \tag{19}$$

To obtain this result use has been made of the initial condition $\Xi(t = 0) = \langle \mathbf{RR} \rangle_{eq} = (k_BT/H)\delta$. A formally exact solution of Eq. (18) is given by the expression

$$\begin{aligned}\Xi(s) &= (s - \mathcal{L}_0 - \delta\mathcal{L})^{-1}B(s)\delta \\ &= \{(s - \mathcal{L}_0)^{-1} + (s - \mathcal{L}_0)^{-1}\delta\mathcal{L}(s - \mathcal{L}_0)^{-1} \\ &\quad + \cdots\}B(s)\delta\end{aligned} \tag{20}$$

or

$$\Xi_{ij}(s) = \frac{B(s)}{s + \lambda_H^{-1}} \sum_{n=0}^{\infty} \left\{\left(\frac{\delta\mathcal{L}}{s + \lambda_H^{-1}}\right)^n\right\}_{ij,kk}, \tag{21}$$

wherein $(\delta\mathcal{L}^n)_{ij,kk} = (\delta\mathcal{L}^n{:}\delta)_{ij}$.

The solution appropriate to the time domain is the inverse Laplace transform of Eq. (21), namely,

$$\Xi(t) = (k_BT/H) \sum_{n=0}^{\infty} \{1 - e^{-t/\lambda_H}\Phi_{n-1}(t/\lambda_H)\} \times \{(\lambda_H\delta\mathcal{L})^n{:}\delta\}, \tag{22}$$

with

$$\Phi_n(z) = 1 + \frac{z}{1!} + \cdots + \frac{z^n}{n!}; \quad n \geq 0,$$

$$\Phi_{-1}(z) = 0. \tag{23}$$

Finally, the steady-state (SS) solution, gotten by either of the two limiting operations $\Xi^{SS} = \lim_{t\to\infty} \Xi(t)$ or $\Xi^{SS} = \lim_{s\to 0} \times s\Xi(s)$, is given by the formula

$$\Xi^{SS} = (k_BT/H) \sum_{n=0}^{\infty} \{(\lambda_H\delta\mathcal{L})^n{:}\delta\}. \tag{24}$$

A. Arbitrary κ; power series for Ξ

To proceed beyond the formally exact results [Eqs. (21), (22), and (24)] one must evaluate the tensors $\delta\mathcal{L}^n{:}\delta$, the first three of which are

$$\delta\mathcal{L}{:}\delta = \kappa + \kappa^T = \dot{\gamma}, \tag{25}$$

$$\begin{aligned}\delta\mathcal{L}^2{:}\delta &= (\kappa\cdot\kappa) + (\kappa\cdot\kappa)^T + 2(\kappa\cdot\kappa^T) \\ &= \dot{\gamma}\cdot\dot{\gamma} + \tfrac{1}{2}(\omega\cdot\dot{\gamma} - \dot{\gamma}\cdot\omega),\end{aligned} \tag{26}$$

$$\begin{aligned}\delta\mathcal{L}^3{:}\delta &= \kappa\cdot\kappa\cdot\kappa + (\kappa\cdot\kappa\cdot\kappa)^T + 3(\kappa\cdot\kappa\cdot\kappa^T) + 3(\kappa\cdot\kappa\cdot\kappa^T)^T \\ &= \dot{\gamma}\cdot\dot{\gamma}\cdot\dot{\gamma} + \tfrac{3}{4}(\dot{\gamma}\cdot\dot{\gamma}\cdot\omega - \omega\cdot\dot{\gamma}\cdot\dot{\gamma}) \\ &\quad + \tfrac{1}{4}(\dot{\gamma}\cdot\omega\cdot\omega - 2\omega\cdot\dot{\gamma}\cdot\omega + \omega\cdot\omega\cdot\dot{\gamma}).\end{aligned} \tag{27}$$

By substituting these expressions into Eq. (22) we obtain the result

$$\begin{aligned}\Xi(t) = (k_BT/H)[\delta &+ \{1 - e^{-t/\lambda_H}\}\lambda_H\dot{\gamma} \\ &+ \{1 - [1 + t/\lambda_H]e^{-t/\lambda_H}\}\lambda_H^2\{\dot{\gamma}\cdot\dot{\gamma} + \tfrac{1}{2}(\omega\cdot\dot{\gamma} - \dot{\gamma}\cdot\omega)\} \\ &+ \{1 - [1 + t/\lambda_H + \tfrac{1}{2}(t/\lambda_H)^2]e^{-t/\lambda_H}\}\lambda_H^3 \\ &\times \{\dot{\gamma}\cdot\dot{\gamma}\cdot\dot{\gamma} + \tfrac{3}{4}(\dot{\gamma}\cdot\dot{\gamma}\cdot\omega - \omega\cdot\dot{\gamma}\cdot\dot{\gamma}) \\ &+ \tfrac{1}{4}(\dot{\gamma}\cdot\omega\cdot\omega - 2\omega\cdot\dot{\gamma}\cdot\omega + \omega\cdot\omega\cdot\dot{\gamma})\} + \cdots],\end{aligned} \tag{28}$$

the steady-state limit of which differs from a formula reported by Armstrong[7] where $\dot{\gamma}\cdot\dot{\gamma}\cdot\dot{\gamma}$ appears multiplied by a factor of $\frac{1}{2}$ instead of 1.

There are two special cases for which either or both of $\Xi(s)$ and $\Xi(t)$ can be obtained in closed, analytic form.

B. κ is symmetric, that is, $\kappa^T = \kappa$

In this case $(\delta\mathcal{L})^n{:}\delta = (2\kappa)^n$ and so, according to Eq. (21):

$$\Xi(s) = \frac{B(s)}{s + \lambda_H^{-1}} \sum_{n=0}^{\infty} \left\{\left(\frac{2\kappa}{s + \lambda_H^{-1}}\right)^n\right\}$$

or

$$\Xi(s) = [(s + \lambda_H^{-1})\delta - 2\kappa]^{-1}B(s). \tag{29}$$

The corresponding solution for the time domain is

$$\Xi(t) = (k_BT/H) \sum_{n=0}^{\infty} \{1 - e^{-t/\lambda_H}\Phi_{n-1}(t/\lambda_H)\}(2\lambda_H\kappa)^n. \tag{30}$$

C. κ is a sparse matrix such that $\delta\mathcal{L}^n{:}\delta = 0$ for $n \geq m$

The simplifying feature in this case is that the infinite series appearing in Eqs. (21), (22), and (24) terminate. As an illustration we consider the example of *simple shear flow* for which

$$\kappa = \begin{pmatrix} 0 & 0 & 0 \\ \dot{\gamma} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{31}$$

From this it follows immediately that $\kappa\cdot\kappa = 0$ and [cf. Eqs. (25)–(27)] $\delta\mathcal{L}^n{:}\delta = 0$ for $n \geq 3$. Consequently, the steady-state value of $\Xi(t)$ is given by

$$\begin{aligned}\Xi^{SS} &= (k_BT/H) \sum_{n=0}^{2} (\lambda_H\delta\mathcal{L})^n{:}\delta \\ &= (k_BT/H)\{\delta + (\lambda_H\delta\mathcal{L}) + (\lambda_H\delta\mathcal{L})^2\}{:}\delta \\ &= \frac{k_BT}{H}\begin{pmatrix} 1 & \lambda_H\dot{\gamma} & 0 \\ \lambda_H\dot{\gamma} & 1 + 2(\lambda_H\dot{\gamma})^2 & 0 \\ 0 & 0 & 1 \end{pmatrix}.\end{aligned} \tag{32}$$

An identical result attributed to H. A. Kramers has been cited by Hermans.[8]

From Eq. (21) we obtain the corresponding time-dependent solution

$$\Xi(t) = (k_BT/H) \sum_{n=0}^{2} \{1 - e^{-t/\lambda_H}\Phi_{n-1}(t/\lambda_H)\}\{(\lambda_H\delta\mathcal{L})^n{:}\delta\}$$

$$= \begin{pmatrix} \Xi_{11}(t) & \Xi_{12}(t) & 0 \\ \Xi_{21}(t) & \Xi_{22}(t) & 0 \\ 0 & 0 & \Xi_{33}(t) \end{pmatrix}, \quad (33)$$

with

$$\Xi_{11}(t) = \Xi_{33}(t) = k_BT/H,$$
$$\Xi_{12}(t) = \Xi_{21}(t) = (k_BT/H)\{(1 - e^{-t/\lambda_H})(\lambda_H\dot{\gamma})\},$$
$$\Xi_{22}(t) = (k_BT/H)\{1 + 2(\lambda_H\dot{\gamma})^2 \times (1 - [1 + t/\lambda_H]e^{-t/\lambda_H})\}. \quad (34)$$

IV. PERTURBATIVE SERIES IN THE VORTICITY

The resolvent operator method presented in the preceding section produces a useful perturbative solution when the modulus of the velocity gradient is small and, as illustrated in Secs. III B and III C, it also works very well when this tensor κ either is symmetric or sparse (in the sense defined in Sec. III C). However, the solutions generated by this standard procedure are not appropriate to the important class of flows for which $\dot{\gamma}(\equiv\kappa^T + \kappa)$ is large and $\omega(\equiv\kappa^T - \kappa)$ is small but significantly different from zero. In this final section we present a method of solution which is specially tailored to flows of this type.

It has been demonstrated in Sec. III B that $\Xi(s)$ can be obtained in closed form when the velocity gradient tensor is symmetric. Here we use this fact to express $\Xi(s)$ as a power series in the vorticity tensor ω. The first term in this series is the solution (29) appropriate to the symmetric part $[\kappa^s = \frac{1}{2}(\kappa + \kappa^T) = \frac{1}{2}\dot{\gamma}]$ of the velocity gradient tensor. Successive terms are of ascending order in ω, each exact to *all* orders in $\dot{\gamma}$.

We begin by separating the Liouville operator $\mathcal{L} = \mathcal{L}_0 + \delta\mathcal{L}$, given by Eqs. (6) and (7), into the sum of two operators

$$\mathcal{L}_1 = \mathcal{L}_0 + \kappa^s \otimes \delta + \delta \otimes \kappa^s \quad (35.1)$$

and

$$\mathcal{L}_2 = \kappa^a \otimes \delta + \delta \otimes \kappa^a. \quad (35.2)$$

The first of these is specific to the potential flow associated with the symmetric part of κ. The second, which will be treated as a perturbation, is proportional to the antisymmetric part of κ, namely, $\kappa^a = \frac{1}{2}(\kappa - \kappa^T) = -\frac{1}{2}\omega$. The resolvent operator $(s - \mathcal{L})^{-1}$ appearing in Eq. (18) is now expanded in powers of $\mathcal{L}_2$ to yield the formula

$$\Xi(s) = \{(s - \mathcal{L}_1)^{-1} \sum_{l=0}^{\infty} [\mathcal{L}_2(s - \mathcal{L}_1)^{-1}]^l\delta\}B(s). \quad (36)$$

The next step is to introduce a unitary operator $\mathcal{U}$ (and its inverse $\mathcal{U}^{-1} = \mathcal{U}^T$) which has the following action on a tetradic $\mathbf{a} \otimes \mathbf{b}$:

$$\mathcal{U}(\mathbf{a} \otimes \mathbf{b})\mathcal{U}^T = u\mathbf{a}u^T \otimes u\mathbf{b}u^T. \quad (37)$$

Here u is the orthogonal matrix which diagonalizes κ^s, viz. $u\kappa^s u^T = \mathbf{D}$, $D_{ij} = D_i\delta_{ij}$. It then follows that

$$\mathcal{D}_1 \equiv \mathcal{U}\mathcal{L}_1\mathcal{U}^T = -\lambda_H^{-1}\delta \otimes \delta + \mathbf{D} \otimes \delta + \delta \otimes \mathbf{D} \quad (38.1)$$

and

$$\mathcal{D}_2 \equiv \mathcal{U}\mathcal{L}_2\mathcal{U}^T = \mathbf{w} \otimes \delta + \delta \otimes \mathbf{w}, \quad (38.2)$$

with $\mathbf{w} = u\kappa^a u^T = -\frac{1}{2}u\omega u^T$. The Cartesian components of these two tetradics are given by the expressions

$$(\mathcal{D}_1)_{ij,mn} = d_{ij}\delta_{im}\delta_{jn}, \quad (39.1)$$

$$(\mathcal{D}_2)_{ij,mn} = -\tfrac{1}{2}\omega_{pq}[u_{ip}u_{mq}\delta_{jn} + u_{jp}u_{nq}\delta_{im}], \quad (39.2)$$

where $d_{ij} = -\lambda_H^{-1} + D_i + D_j$.

From Eq. (36) and the definitions (37) and (38) it then can be seen that

$$\Xi(s) = \mathcal{U}^T\{\mathcal{U}\Xi(s)\}$$
$$= \mathcal{U}^T\{(s - \mathcal{D}_1)^{-1} \sum_{l=0}^{\infty} [\mathcal{D}_2(s - \mathcal{D}_1)^{-1}]^l\delta\}B(s) \quad (40)$$

or

$$\Xi(s)_{ij} = u_{mi}u_{nj}\{(s - \mathcal{D}_1)^{-1}_{mn,pq} \sum_{l=0}^{\infty} [\mathcal{D}_2(s - \mathcal{D}_1)^{-1}]^l_{pq,kk}\}B(s)$$
$$= u_{mi}u_{nj}\{(s - d_{mn})^{-1} \sum_{l=0}^{\infty} ([\mathcal{D}_2(s - \mathcal{D}_1)^{-1}]^{l-1}\mathcal{D}_2)_{mn,kk} \times (s - d_{kk})^{-1}\}B(s). \quad (41)$$

The leading term of the series (41) can be evaluated by the following sequence of manipulations,

$$\Xi^{(0)}(s)_{ij} = u_{mi}u_{nj}(s - \mathcal{D}_1)^{-1}_{mn,kk}B(s)$$
$$= u_{mi}u_{mj}(s - d_{mm})^{-1}B(s)$$
$$= (s + \lambda_H^{-1})^{-1}B(s)u_{mi}u_{mj}\left[1 - \frac{2D_m}{s + \lambda_H^{-1}}\right]^{-1}$$
$$= (s + \lambda_H^{-1})^{-1}B(s) \sum_{k=0}^{\infty} (s + \lambda_H^{-1})^k[u_{mi}(2D_m)^k u_{mj}]$$
$$= \frac{B(s)}{s + \lambda_H^{-1}} \sum_{k=0}^{\infty} \left\{\left(\frac{2\kappa^s}{s + \lambda_H^{-1}}\right)^k\right\}_{ij}, \quad (42)$$

the last of which involves the identification of $u_{mi}(2D_m)^k u_{mj}$ with $\{(2\kappa^s)^k\}_{ij}$. $\Xi^{(0)}(s)$ given by Eq. (42) is clearly the same as Eq. (29).

The second term of the series (41) is

$$\Xi^{(1)}(s)_{ij} = u_{mi}u_{nj}(s - d_{mn})^{-1}(\mathcal{D}_2)_{mn,kk}(s - d_{kk})^{-1}B(s)$$
$$= \tfrac{1}{2}B(s)\omega_{pq}(u_{mi}u_{mp})(u_{nj}u_{nq})(s - d_{mn})^{-1} \times [(s - d_{mm})^{-1} - (s - d_{nn})^{-1}]. \quad (43)$$

By writing $(s - d_{mn})^{-1}$ in the form

$$(s - d_{mn})^{-1} = (s + \lambda_H^{-1}) \sum_{p=0}^{\infty} \sum_{q=0}^{p} \binom{p}{q} 2^{-p}\left(\frac{2D_m}{s + \lambda_H^{-1}}\right)^q\left(\frac{2D_n}{s + \lambda_H^{-1}}\right)^{p-q},$$

wherein $\binom{p}{q} = p!/q!(p-q)!$, Eq. (43) can be transformed into the expression

$$\Xi^{(1)}(s)_{ij} = \tfrac{1}{2}B(s)(s+\lambda_H^{-1})^{-2}\sum_{k=0}^{\infty}\sum_{p,q}\binom{p}{q}2^{-p} \times\left\{\left(\frac{2\kappa^s}{s+\lambda_H^{-1}}\right)^{q+k}_{im}\omega_{mn}\left(\frac{2\kappa^s}{s+\lambda_H^{-1}}\right)^{p-q}_{nj} - \left(\frac{2\kappa^s}{s+\lambda_H^{-1}}\right)^{p-q}_{im}\omega_{mn}\left(\frac{2\kappa^s}{s+\lambda_H^{-1}}\right)^{q+k}_{nj}\right\}. \tag{44}$$

A more compact formula for $\Xi^{(1)}(s)$ is

$$\Xi^{(1)}(s) = \tfrac{1}{2}B(s)(s+\lambda_H^{-1})^{-2}[(\delta-\dot{\boldsymbol{\Gamma}})^{-1}\cdot\boldsymbol{\Omega} - \boldsymbol{\Omega}\cdot(\delta-\dot{\boldsymbol{\Gamma}})^{-1}], \tag{45}$$

with $\dot{\boldsymbol{\Gamma}} = \dot{\gamma}(s+\lambda_H^{-1})^{-1}$ and where

$$\boldsymbol{\Omega} = \sum_{p=0}^{\infty}\sum_{q=0}^{p}\binom{p}{q}2^{-p}(\dot{\boldsymbol{\Gamma}})^q\cdot\omega\cdot(\dot{\boldsymbol{\Gamma}})^{p-q} = \omega + \tfrac{1}{2}(\omega\cdot\dot{\boldsymbol{\Gamma}} - \dot{\boldsymbol{\Gamma}}\cdot\omega) + (1/2^2) \times(\omega\cdot\dot{\boldsymbol{\Gamma}}^2 - 2\dot{\boldsymbol{\Gamma}}\cdot\omega\cdot\dot{\boldsymbol{\Gamma}} + \dot{\boldsymbol{\Gamma}}^2\cdot\omega) + \cdots. \tag{46}$$

$\Xi^{(1)}(s)$ includes all contributions to $\Xi(s)$ which are first order in the vorticity tensor ω.

Manipulations similar to those used in the computation of $\Xi^{(1)}$ lead to the result

$$\Xi^{(2)}(s) = -\tfrac{1}{4}B(s)(s+\lambda_H^{-1})^{-3}\sum_{p,q}\sum_{p',q'}\binom{p}{q} \times\binom{p'}{q'}2^{-p-p'}(\mathbf{A}\cdot\mathbf{B} - \mathbf{B}^T\cdot\mathbf{A}^T), \tag{47}$$

wherein

$$\mathbf{A} = (\dot{\boldsymbol{\Gamma}})^q\cdot\omega, \tag{48.1}$$

$$\mathbf{B} = (\dot{\boldsymbol{\Gamma}})^{p'-q'}\cdot(\delta-\dot{\boldsymbol{\Gamma}})^{-1}\cdot\omega\cdot(\dot{\boldsymbol{\Gamma}})^{p-q+q'} + (\dot{\boldsymbol{\Gamma}})^{p-q+q'}\cdot\omega\cdot(\delta-\dot{\boldsymbol{\Gamma}})^{-1}\cdot(\dot{\boldsymbol{\Gamma}})^{p'-q'}. \tag{48.2}$$

Although the results reported in these last two sections are specific to the rheological equation of state for the Hookean dumbbell model, the methods of solution obviously can be applied to all of the corotational and codeformational equations presented in Sec. II and cast there into the common form of Eq. (15).

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