NON-NEWTONIAN CONSTITUTIVE EQUATIONS USING THE ORIENTATIONAL ORDER PARAMETER

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Abstract. Nonlinear hydrodynamic equations for non-Newtonian fluids are discussed. We start from the recently derived hydrodynamic-like nonlinear description of a slowly relaxing orientational order parameter tensor. The reversible quadratic nonlinearities in this tensor's dynamics are material dependent due to the generalized nonlinear flow alignment effect that comes in addition to the material independent corotational convected derivative. In the entropy production these terms are balanced by linear and nonlinear orientational-elastic contributions to the stress tensor. These can be used to get a nonlinear dynamic equation for the stress tensor (sometimes called constitutive equation) in terms of a power series in the variables. A comparison with existing phenomenological models is given. In particular we discuss how these ad-hoc models fit into the hydrodynamic description and where the various non-Newtonian contributions are coming from. We also discuss the connection to the hydrodynamic-like description of non-Newtonian effects that employs a relaxing strain tensor.

Key words. Constitutive equations, orientational order parameter, non-Newtonian effects, hydrodynamics, flow alignment, relaxing strain tensor.

AMS(MOS) subject classifications. Primary 76A05, 74D10, 80A17, 76A15.

1. Introduction. Hydrodynamics is a well established field to describe macroscopically simple fluids by means of the Navier-Stokes, continuity, and heat conduction equations. However, it applies also to more complex fluids that are fully characterized by conservation laws and broken symmetries. It is based on (the Gibbsian formulation of) thermodynamics [1, 2], symmetries and well-founded physical principles [3]. A detailed description of this method can be found in [4,5]. This method can be generalized to include slowly relaxing variables that are relevant on experimental macroscopic time scales albeit being non-hydrodynamic. Examples are the soft mode near phase transitions [6, 7], the magnetic degree of freedom in ferrofluids [8, 9] and the relative velocity in 2-fluid descriptions [10]. The derivation of such macroscopic nonlinear dynamic equations is still based on first principles, making use of thermostatics, linear irreversible thermodynamics, symmetries and broken symmetries, and invariance principles. Only the choice of the slowly varying variable is heuristic and material dependent. In that sense non-Newtonian fluids are non-universal.

On the other hand, a host of different empirical models have been proposed [11–17] to cope with the rheology of such substances. Typically

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these models are formulated as generalizations of the linear, Newtonian relation between stress and deformational flow allowing for additional time derivatives and nonlinearities. They are tailored to accommodate empirical findings or are based on principles [15] that are ad-hoc and generally insufficient.

Quite recently we have derived a nonlinear hydrodynamic description of elastic media [18, 19] that has been confirmed within the GENERIC formalism [20]. Allowing in this hydrodynamic description the strains to relax (and not only to diffuse) a generalized hydrodynamic description of nonlinear viscoelasticity is obtained in terms of a dynamic equation for the (Eulerian) strain tensor [18, 19]. This strain tensor description can be transformed approximately into one that uses a dynamic equation for the stress tensor [21] and can thus be directly compared with many of the empirical models proposed to describe non-Newtonian rheology. The comparison reveals possible inconsistencies and connects the various ad-hoc additions of those models with physical relevant processes, like strain relaxation, elasticity and viscosity [21].

In this communication we use a different approach that relates non-Newtonian behavior to fluctuating, transient, and slowly relaxing orientational order. This has been used e.g. for describing the dynamics of semiflexible polymers, where long-lived polymer alignments and entanglements lead to viscoelastic effects [22]. The relaxational dynamics of the orientational order parameter tensor has been used in the isotropic phase of low molecular weight nematogens [23] describing orientational fluctuations that become important as pre-transitional effects near the phase transition. The relaxational (and non relaxational) dynamics of the orientational tensor has been derived and rederived pretty often [24–29]. Here, we will rely on the hydrodynamic description [30] that e.g. makes the clear distinction between reversible and irreversible processes and avoids any detours via additional auxiliary and unphysical dynamic variables. In Sec.2 orientational elasticity and the phenomenological material tensors describing reversible and irreversible transport (flow alignment, viscosity, and relaxation) that are part of the hydrodynamic description are given as an expansion in powers of the orientational tensor. The back-flow effect in the stress tensor (Sec.3), which is required for thermodynamic reasons, as well as the part of the viscosity that depends on the orientational tensor provide a coupling between the stress and the orientational tensor. This can be used to generate a dynamic equation for the stress tensor from that of the orientational tensor (Sec.4). This translation is achieved by a power series expansion in the variables and can be done only approximately, since, generally, nonlinear equations cannot be inverted analytically. The power series is truncated after the quadratic order, since most of the phenomenological constitutive models, which we compare with in Sec.5, are of that form. A summary (Sec.6) of the main results concludes the paper.

2. Dynamics of the orientational order parameter tensor. The transient orientational order is described by a symmetric, traceles second rank tensor Q_{ij} ($Q_{ij} = Q_{ji}$ and $Q_{ii} = 0$). In contrast to the case of a nematic phase with spontaneous and permanent orientational order, there is no nematic order in equilibrium and a director does not exist. The relaxational dynamics of Q_{ij} can be written as [30]

$$\dot{Q}_{ij} + v_k \nabla_k Q_{ij} + Q_{jk}\Omega_{ki} + Q_{ik}\Omega_{kj} - \lambda_{ijkl}A_{kl} = -\alpha_{ijkl}\psi_{kl} \qquad (2.1)$$

with $2A_{ij} = \nabla_j v_i + \nabla_i v_j$ and $2\Omega_{ij} = \nabla_j v_i - \nabla_i v_j$ the symmetric and antisymmetric velocity gradients characterizing deformational and rotational flow, respectively. The orientational elastic stress tensor ψ_{kl} is defined by the Gibbs relation [4]

$$d\epsilon - Td\sigma = v_i dg_i + \psi_{ij}dQ_{ij} + \mu d\rho. \qquad (2.2)$$

as the conjugate to Q_{ij}. It has to be taken as symmetric and traceless, since only that part enters the Gibbs relation and has a physical meaning. The Gibbs relation contains all the other variables (density ρ, momentum density g_i, energy density ϵ or entropy density σ) and defines their conjugates (temperature T, velocity v_i, and chemical potential μ), where the latter are related to the more familiar (thermodynamic) pressure p by the Gibbs-Duhem equation

$$dp = \sigma dT + g_i dv_i - \psi_{ij}dQ_{ij} + \rho d\mu \qquad (2.3)$$

In Eq.(2.1) the nonlinear reversible coupling terms to flow are a priori of the corotational or Jaumann derivative type (containing only Ω_{ij} the rotational flow – suitably for the orientational order involved), but there is in addition a phenomenological reversible coupling to symmetric velocity gradients that makes the effective convective derivative material dependent [30]. The phenomenological material tensor λ_{ijkl} (a kind of generalized flow alignment tensor) is given as a power series expansion in Q_{ij}

$$\lambda_{ijkl} = \lambda_1 \left(\delta_{ik}\delta_{jl} + \delta_{jk}\delta_{il} - \frac{2}{3}\delta_{ij}\delta_{kl} \right) + \lambda_3 \delta_{kl}Q_{ij} \qquad (2.4)$$

$$+ \lambda_2 \left(\delta_{ik}Q_{jl} + \delta_{jk}Q_{il} + \delta_{jl}Q_{ik} + \delta_{il}Q_{jk} - \frac{4}{3}\delta_{ij}Q_{kl} \right) + O(2)$$

where higher order terms $O(2)$ have been discussed in [30], but are not needed here. It contains one phenomenological, material dependent, reversible reactive coefficient in linear, and two additional ones in quadratic order. If in Eq.(2.1) the Jaumann terms are combined with the quadratic contribution (2.4) for the special value $\lambda_2 = \frac{1}{2}$ ($= -\frac{1}{2}$) one gets something that looks like an upper (lower) convected derivative – with some additional correction terms that ensure $\dot{Q}_{ii} = 0$. However, there is no general reason why such a relation should hold for all different materials nor can

it hold for all temperatures and pressures, since $\lambda_{1,2,3}$ generally depend on all scalar state variables, like ρ, σ (or p, T) and on the invariants $Q_{ij}Q_{ij}$ and $Q_{ij}Q_{jk}Q_{ki}$. Within the quadratic approximation used here, the latter dependencies do not show up.

In [30] the relaxation of Q_{ij} has been given in linear approximation. More generally, the dissipative material tensor α_{ijkl} reads in a power series expansion in Q_{ij} (with $\alpha_{iikl} = 0 = \alpha_{ijkk}$)

$$\alpha_{ijkl} = \alpha_1 \left(\delta_{ik}\delta_{jl} + \delta_{jk}\delta_{il} - \frac{2}{3}\delta_{ij}\delta_{kl}\right) \tag{2.5}$$

$$+ \alpha_2 \left(\delta_{ik}Q_{jl} + \delta_{jk}Q_{il} + \delta_{jl}Q_{ik} + \delta_{il}Q_{jk} - \frac{4}{3}[\delta_{ij}Q_{kl} + \delta_{kl}Q_{ij}]\right) + O(2)$$

with the relaxation parameters $\alpha_{1,2}$ being functions of the scalar state variables. It should be noted that we stay very well inside the framework of "linear irreversible thermodynamics" that has a solid foundation in statistical mechanics, although the expressions (2.4,2.5) and (3.3) below are genuinely nonlinear due to the dependence on state variables.

The orientational elastic stress is derived from an energy functional by the variational derivative $\psi_{ij} = \delta \int \epsilon \, dV / \delta Q_{ij}$, where only the trace free part enters Eqs.(2.1-2.3), which is given in quadratic order by

$$\psi_{ij} = c_1 Q_{ij} + c_2(Q_{ik}Q_{jk} - \frac{1}{3}\delta_{ij}Q_{kl}Q_{kl}) + O(2) \tag{2.6}$$

neglecting gradient terms. Near a phase transition the rotational elastic moduli c_1, c_2 can be interpreted as Landau parameters. Generally they are still functions of all scalar state variables.

Putting together Eqs.(2.1-2.6) the final dynamic orientational order parameter equations, quadratic in the variables, is obtained as

$$\dot{Q}_{ij} + v_k \nabla_k Q_{ij} + Q_{jk}\Omega_{ki} + Q_{ik}\Omega_{kj} - 2\lambda_1(A_{ij} - \frac{1}{3}\delta_{ij}A_{kk})$$

$$-2\lambda_2(A_{il}Q_{jl} + A_{jl}Q_{il} - \frac{2}{3}\delta_{ij}A_{kl}Q_{kl}) - \lambda_3 Q_{ij}A_{kk}$$

$$= -\frac{1}{\tau_1}Q_{ij} - \frac{1}{\tau_2}(Q_{il}Q_{jl} - \frac{1}{3}\delta_{ij}Q_{kl}Q_{kl}) \tag{2.7}$$

where the relaxation times are related to the elastic moduli and the relaxation parameters by $1/\tau_1 = 2c_1\alpha_1$ and $1/\tau_2 = 2c_2\alpha_1 + 4c_1\alpha_2$.

3. Stress Tensor. In the preceding sections we discussed nonlinear reversible terms in the dynamic equation for the orientational order (2.1) that describe couplings to flow. In the Navier-Stokes or momentum conservation equation

$$\dot{g}_i + \nabla_j(v_j g_i + \delta_{ij}p + \sigma_{ij}) = 0, \tag{3.1}$$

on the other hand, there must be appropriate counter terms describing couplings to orientational order, due to the requirement of zero or positive entropy production, in the case of reversible and irreversible terms, respectively [4,5,31]. Their form can equivalently be derived from Onsager relations [32]. For the stress tensor σ_{ij} this leads to the expression

$$\sigma_{ij} = -\lambda_{klij}\psi_{kl} - \nu_{ijkl}A_{kl} \tag{3.2}$$

The counter term to the linear deformational flow term in (2.1), $\sim \lambda_{klij}$, leads to a symmetric part of the stress tensor, while there are no counter terms to the nonlinear Jaumann terms, since the latter do not at all contribute to the entropy production [30]. The viscosity tensor is again expanded in Q_{ij} as

$$\nu_{ijkl} = \frac{\nu_1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \frac{\nu_2}{2}(Q_{ik}\delta_{jl} + Q_{jk}\delta_{il} + Q_{il}\delta_{jk} + Q_{jl}\delta_{ik})$$
$$+ \nu_3\delta_{ij}\delta_{kl} + \nu_4(\delta_{ij}Q_{kl} + \delta_{kl}Q_{ij}) \tag{3.3}$$

with the viscosities generally being functions of the scalar state variables.

Taking together Eqs.(2.5,3.2,3.3) the stress is given by

$$\sigma_{ij} = -\nu_1 A_{ij} - \nu_2(Q_{ik}A_{jk} + Q_{jk}A_{ik}) - \nu_3\delta_{ij}A_{kk} \tag{3.4}$$
$$- \nu_4(\delta_{ij}Q_{kl}A_{kl} + Q_{ij}A_{kk}) - \bar{\lambda}_1 Q_{ij} - \bar{\lambda}_2 Q_{ik}Q_{jk} - \bar{\lambda}_3\delta_{ij}Q_{kl}Q_{kl}$$

where we have used the abbreviations $\bar{\lambda}_1 = 2c_1\lambda_1$, $\bar{\lambda}_2 = 2c_2\lambda_1 + 4c_1\lambda_2$, and $\bar{\lambda}_3 = c_1\lambda_3 - (2/3)c_2\lambda_1$. In the incompressible limit, which we will use below, $A_{kk} = 0$, and the viscosity ν_3 does not appear in the stress tensor, while λ_3 drops out of Eq.(2.7). If we allow for a "redefinition" of the pressure, $p \to p - \nu_4 Q_{kl}A_{kl} - \bar{\lambda}_3 Q_{kl}Q_{kl}$, also ν_4 and $\bar{\lambda}_3$ do not show up explicitly in the final equations. However, in that case p looses its simple physical meaning. For a general discussion of the incompressible limit and its connection to redefining the pressure cf. [33].

4. Dynamic stress tensor equation. Eqs.(2.7,3.1,3.4) constitute an (isothermal) description of viscoelasticty based on a relaxing orientational order parameter tensor. This hydrodynamic-like description contains as special cases [30] some of the well-known model-based descriptions of viscoelasticity that also employ the orientational order parameter tensor, like e.g. the Doi-Edwards model for isotropic semiflexible polymers [22]. However, most of the heuristic constitutive models are written in terms of a dynamic equation for the stress tensor, very often quadratic in the variables and under the assumption of incompressibility. In order to compare with those models we have to translate our $\dot{Q}_{ij}/\dot{g}_i$ into a $\dot{\sigma}_{ij}/\dot{g}_i$ description by replacing the orientational order parameter tensor (and its derivatives) by the stress tensor (and its derivatives). This can only be done in an approximate way, since the equations are nonlinear. We will set up a power series expansion up to second order in the (old and new) variables. Of

course, the resulting equations are less general than the starting ones and only applicable, if quadratic nonlinearities are sufficient for the problem at hand. This procedure is similar in spirit to that in [21], where we used the hydrodynamic-like description of viscoelasticity in terms of a relaxing Eulerian strain tensor [18,19] and translated it into a dynamic stress tensor description, again to facilitate comparison.

Taking the derivative $d/dt = \partial/\partial t + v_i \nabla_i$ of σ_{ij} in (3.4) and replacing dQ_{ij}/dt according to Eq.(2.7) we get

$$\frac{d}{dt}\sigma_{ij} = -f\left(Q_{ij}, A_{ij}, \frac{d}{dt}A_{ij}, \Omega_{ij}\right) \qquad (4.1)$$

in terms of the orientational order parameter tensor and flow. To convert this into the desired dynamic equation for the stress tensor, we have to invert $\sigma_{ij} = \sigma_{ij}(Q_{ij}, A_{ij})$, Eq.(3.4), into $Q_{ij} = Q_{ij}(\sigma_{ij}, A_{ij})$. This is done approximately by the power expansion $Q_{ij} = Q_{ij}^{(lin)} + Q_{ij}^{(quad)} + \ldots$, where $Q_{ij}^{(lin)}$ and $Q_{ij}^{(quad)}$ contain expressions linear and quadratic in the variables, respectively. In particular we find

$$\bar{\lambda}_1 Q_{ij}^{(lin)} = -\sigma_{ij}^0 - \nu_1 A_{ij} \qquad (4.2)$$

$$\bar{\lambda}_1^3 Q_{ij}^{(quad)} = -\bar{\lambda}_2(\sigma_{ik}\sigma_{jk})^0 + (\bar{\lambda}_1\nu_2 - \bar{\lambda}_2\nu_1)(\sigma_{ik}A_{jk} + \sigma_{jk}A_{ik})^0$$
$$+\nu_1(2\bar{\lambda}_1\nu_2 - \bar{\lambda}_2\nu_1)(A_{ik}A_{jk})^0 \qquad (4.3)$$

where the superscript ^{0} denotes the traceless part of the associated tensor. Since we assume incompressibility, A_{ij} is traceless by itself.

Using these expressions the dynamic equation for the stress tensor takes the final form

$$\tau_1 \frac{D_s}{Dt}\sigma_{ij} + \sigma_{ij} = -\nu_\infty A_{ij} - \nu_1\tau_1 \frac{D_q}{Dt}A_{ij} + \frac{r}{2c_1\lambda_1}\sigma_{ik}\sigma_{jk} + \frac{1}{3}\delta_{ij}\Sigma$$
$$+\frac{\tau_1\nu_2}{2c_1\lambda_1}\left([\sigma_{jk} + \nu_1 A_{jk}]\frac{\partial}{\partial t}A_{ik} + [\sigma_{ik} + \nu_1 A_{ik}]\frac{\partial}{\partial t}A_{jk}\right) + O(3) \quad (4.4)$$

where

$$\nu_\infty = \nu_1 + 4c_1\tau_1\lambda_1^2 \qquad \text{and} \qquad r = \frac{\tau_1}{\tau_2} + \frac{c_2}{c_1} + 2\frac{\lambda_2}{\lambda_1} \qquad (4.5)$$

and

$$\frac{D_s}{Dt}T_{ij} \equiv \frac{d}{dt}T_{ij} - s(T_{ik}A_{jk} + T_{jk}A_{ik}) - (T_{ik}\Omega_{jk} + T_{jk}\Omega_{ik}) \qquad (4.6)$$

for any tensor T_{ij} and number s. For $s = -1$ ($s = +1$) D_s/Dt is the lower (upper) convected derivative, for $s = 0$ the Jaumann or corotational derivative, while for a general s a linear combination of those is invoked.

In our case the numbers s and q are

$$s = -2\lambda_1\Big(\frac{c_2}{c_1} + 3\frac{\lambda_2}{\lambda_1}\Big) - \frac{\nu_1 r}{2c_1\lambda_1\tau_1} \tag{4.7}$$

$$q = -2\lambda_1\Big(\frac{c_2}{c_1} + 3\frac{\lambda_2}{\lambda_1} - \frac{\nu_2}{\nu_1}\Big) - \frac{\nu_1 r}{4c_1\lambda_1\tau_1} \tag{4.8}$$

where r is given in Eq.(4.5). The part $\sim \delta_{ij}$ in Eq.(4.4) is due to the fact that Q_{ij} is traceless, while σ_{ij} is not. It can in principle be incorporated into the pressure term by a redefinition $p \rightarrow p + (1/3)\Sigma$, where

$$\Sigma = \sigma_{kk} + x\,A_{kl}A_{kl} + y\,\sigma_{kl}\sigma_{kl} + z\,\sigma_{kl}A_{kl}$$
$$+ \frac{3\nu_4\tau_1}{2c_1\lambda_1}(\nu_1 A_{kl} + \sigma_{kl})\frac{\partial}{\partial t}A_{kl} \tag{4.9}$$

with

$$x = \frac{\nu_1^2}{2c_1\lambda_1}\Big(\frac{\lambda_3}{\lambda_1} - \frac{\tau_1}{\tau_2} + \frac{c_2}{3c_1} + \frac{2\lambda_2}{\lambda_1}\Big) - \frac{\nu_1(2\nu_2 + 3\nu_4)}{2c_1\lambda_1}$$
$$- 2\nu_1\lambda_1\tau_1\Big(\frac{2\lambda_2}{\lambda_1} - \frac{\lambda_3}{\lambda_1} + \frac{2c_2}{3c_1}\Big) - 6\lambda_1\tau_1\nu_4 \tag{4.10}$$

$$y = \frac{1}{2c_1\lambda_1}\Big(\frac{\lambda_3}{\lambda_1} - \frac{\tau_1}{\tau_2} + \frac{c_2}{3c_1} + \frac{2\lambda_2}{\lambda_1}\Big) \tag{4.11}$$

$$z = \frac{\nu_1}{c_1\lambda_1}\Big(\frac{2\lambda_3}{\lambda_1} - \frac{2\tau_1}{\tau_2} - \frac{c_2}{3c_1} + \frac{2\lambda_2}{\lambda_1}\Big) - \frac{\nu_2}{c_1\lambda_1}$$
$$- 2\lambda_1\tau_1\Big(\frac{2\lambda_2}{\lambda_1} - \frac{\lambda_3}{\lambda_1} + \frac{2c_2}{3c_1}\Big) - \frac{3\nu_4}{2c_1\lambda_1} \tag{4.12}$$

This "redefinition" of the pressure, however, is rather dubious, since it renders a "pressure" that depends nonlinearly on flow and its time derivative, and even more disturbing on the stress tensor itself. It is completely different from the appropriate "redefinition" in the $\dot{Q}_{ij}/\dot{g}_i$ description of Sec.3. In a more reasonable description one notices that σ_{kk} and its derivative are at least of quadratic order (for $A_{kk} = 0$) and do not influence the constitutive equation for σ_{ij}^0 in that order. For the latter one then gets finally

$$\tau_1\Big(\frac{D_s}{Dt}\sigma_{ij}\Big)^0 + \sigma_{ij}^0 = -\nu_\infty A_{ij} - \nu_1\tau_1\Big(\frac{D_q}{Dt}A_{ij}\Big)^0 + \frac{r}{2c_1\lambda_1}(\sigma_{ik}\sigma_{jk})^0$$
$$+ \frac{\tau_1\nu_2}{2c_1\lambda_1}\left([\sigma_{jk} + \nu_1 A_{jk}]\frac{\partial}{\partial t}A_{ik} + [\sigma_{ik} + \nu_1 A_{ik}]\frac{\partial}{\partial t}A_{jk}\right)^0 + O(3) \tag{4.13}$$

with the coefficients defined above. The time evolution of the trace

$$\tau_1\frac{\partial}{\partial t}\sigma_{kk} = \Big(\frac{r}{2c_1\lambda_1} + y\Big)\sigma_{kl}^0\sigma_{kl}^0 + x\,A_{kl}A_{kl} + z\,\sigma_{kl}^0 A_{kl}$$
$$+ \frac{\tau_1}{2c_1\lambda_1}(2\nu_2 + 3\nu_4)(\nu_1 A_{kl} + \sigma_{kl}^0)\frac{\partial}{\partial t}A_{kl} + O(3) \tag{4.14}$$

is completely determined by σ_{ij}^0 and A_{ij} in lowest order.

5. Comparison with constitutive models. Eq.(4.4) constitutes the most general form for a constitutive equation (up to quadratic order in the variables) that can be derived from a transient orientational order parameter as source of non-Newtonian behavior. It contains eight material coefficients (four linear and four quadratic ones with subscript 1 and 2, respectively), and two more in the trace part (λ_3, ν_4), characterizing orientational elasticity, relaxation of orientational order, viscosity and flow alignment. These coefficients are still functions of density and temperature. Most of the traditional constitutive models are much simpler than Eq.(4.4). We will now discuss, whether and how these models fit into the frame derived above.

The general case (4.4) contains the relaxation of stresses as well as of flow with relaxation times τ_1 and $\tau_1 \nu_1/\nu_\infty$, respectively. Here the effective viscosity ν_∞ is different from the bare one (ν_1) due to the relaxation of orientational order and its coupling to flow via the flow alignment effect. Thus, the Maxwell [15], Johnson-Segalman [16], and Giesekus [14] models, which neglect flow relaxation, implicitly assume $\nu_1 = 0$ and ν_∞ is completely due to flow alignment. The quadratic stress contribution $\sim r$ in (4.4) is nonzero (as in the Giesekus model) only, if at least one of the second order material parameters, $c_2, \lambda_2, 1/\tau_2$, is nonzero. Vice versa, all the other models (including the Oldroyd [11] and Jeffreys [15] models) that have $r = 0$ also implicitly assume $c_2 = 0 = \lambda_2 = 0 = 1/\tau_2$. In principle, it would be possible to have $r = 0$ for a special set of nonzero values of the second order parameters, but this would be highly incidentally and would work only for one special point in phase space (for one combination of density and temperature), but not in general. As a consequence the nature of the convected derivatives of stress and flow, characterized in (4.4) by s, q, is fixed to be of the corotational or Jaumann type ($s = 0 = q$), since in all the models mentioned above there is either $\nu_1 = 0$ or $c_2 = 0 = \lambda_2 = 0 = 1/\tau_2$ or both. Thus, only the Jeffreys and Johnson-Segalman model (the latter in the version with the corotational convective derivative of the stress tensor) are compatible with viscoelasticity due to transient orientational order. These models also consistently lack the complicated nonlinear term in the second line of (4.4), since they have $\nu_2 = 0$; in addition, they miss the trace part $\sim \Sigma$. That means in these models the pressure has to be interpreted as the redefined pressure discussed above, rather than the thermodynamic hydrostatic pressure.

6. Summary. We have explored the hydrodynamic form of non-Newtonian fluid dynamics, if viscoelasticity is due to transient orientational order. The dynamic equation for the orientational order parameter tensor has been converted approximately into a dynamic equation for the stress tensor, which is then compared with traditional constitutive models. Due to the intricate relations among the coefficients of the nonlinearities in this effective constitutive equation some of the models are incompatible

with this type of visoelasticity, since they lack one type of nonlinearity, but inconsistently not some other one, or they assume a special type of convective derivative incompatible with other choices of the nonlinear terms. Compatible are a generalized Giesekus model (with the convective derivative of the stress tensor being material dependent, in general), the Jeffreys model and the Johnson-Segalman model with the corotational convective derivative for the stress tensor. This is quite complementary to our recent findings [21] that the latter two models are incompatible with viscoelasticity due to transient elasticity characterized by a relaxing strain tensor, while Maxwell and Oldroyd models (incompatible in the present case) have been found to be compatible. The deeper reason for this difference lies in the type of viscoelasticity used, either a transient elasticity leading to a relaxing strain tensor that contains the lower (upper) convected time derivative in the Eulerian (Lagrangian) case [18, 19], or a transient orientational order leading to a relaxing orientational order parameter tensor that contains the corotational convected time derivative modified by second order flow alignment material parameters [30]. Of course, in nature both (and even other) sources of viscoelasticity can be present allowing all these models to exist, but one should bear in mind that the general effective constitutive equation obtained in that way is by far richer and more complicated than any of the traditional models.

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