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Stress retardation versus stress relaxation in linear viscoelasticity

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This paper is dedicated to the memory of Prof. C.I. Christov.

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ABSTRACT

We present a preliminary examination of a new approach to a long-standing problem in non-Newtonian fluid mechanics. First, we summarize how a general implicit functional relation between stress and rate of strain of a continuum with memory is reduced to the well-known linear differential constitutive relations that account for "relaxation" and "retardation." Then, we show that relaxation and retardation are asymptotically equivalent for small Deborah numbers, whence causal pure relaxation models necessarily correspond to ill-posed pure retardation models. We suggest that this dichotomy could be a possible way to reconcile the discrepancy between the theory of and certain experiments on viscoelastic liquids that are conjectured to exhibit only stress retardation.

1. Introduction

Viscoelastic non-Newtonian fluids continue to be an active area of research not only because of the difficulties in their theoretical modeling [1] and the challenges in their experimental interrogation [2], but also because of their abundance in biophysics [3–5] and their relevance to continua with local thermal non-equilibrium effects [6, §8.4].

Recently, new experimental methods have been proposed for rheological measurements of polymeric solutions [2] and novel calculations have been performed for the locomotion of microorganisms in "weakly viscoelastic" fluids [4]. Yet, the "second-order fluid" model used in the latter works, and also for interpreting previous experiments [7], is unstable (ill-posed in the sense of Hadamard) [8–10] for a first normal stress difference $\Psi_1 > 0$ as measured. Various explanations have been put forth [11], often questioning the experimental setup and data analysis. Others dismiss the difficulty as not important for "small" departures from Newtonian behavior. Similar ill-posed models arise from the Chapman–Enskog expansion of the Boltzmann–Bhatnagar–Gross–Krook equation when keeping only leading-order non-Newtonian terms [12,13].

In the face of such extensive evidence that, in the real world, the first normal stress difference $\Psi_1 > 0$ for a second-order fluid, it appears to us that it is neither satisfactory to claim that the instability is not manifested for "slow flows" or "small departures from Newtonian behavior" nor is it satisfactory to repeat the mantra that all experimental results are inconclusive or wrong. New insights are needed to understand such a non-trivial discrepancy in the foundations of viscoelasticity, given the resurgence of the "second-order fluid" model [2,4,12,13]. In this preliminary research report, we propose another approach. Specifically, we show how the ill-posed second-order (retardational) fluid model may arise as an improper interpretation of a fluid that is actually exhibiting stress relaxation of the Maxwell type [14],[2] since the latter would be indistinguishable from the former for small departures from Newtonian rheology.

2. Background on memory effects and nonlocal rheology

In this section, in order to make this preliminary research report self-contained and accessible to a wider audience, we summarize

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[2] Maxwell-type relaxation is also common in nonclassical theories of heat conduction [15–17] and thermoelasticity [18].

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the standard background on constitutive modeling for viscoelastic fluids.

As usual, we decompose the stress tensor $\mathbf{T}$ into an indeterminate part (the spherical pressure p) and a constitutive part $\mathbf{S}$ as $\mathbf{T} = -p\mathbf{I} + \mathbf{S}$. We consider only isochoric motions (or incompressible fluids) so that $\mathrm{tr}(\nabla \boldsymbol{u}) = \nabla \cdot \boldsymbol{u} = 0$, where $\boldsymbol{u}$ is the velocity field. The fluid is assumed homogeneous and isotropic so that it has constant density ϱ_0, and its rheological parameters (e.g., the viscosity) are constant scalars.

The most general implicit relationship between the stress tensor $\mathbf{T}(\boldsymbol{x}, t)$ and the rate-of-strain tensor $\mathbf{E}(\boldsymbol{x}, t)$ that includes the effect of *memory* is a *functional* that depends on the independent variables. The relationship is further assumed to be local in the spatial variable (i.e., the functional's value at a given point $\boldsymbol{x}$ is a point function of these tensors at $\boldsymbol{x}$) to preclude "action at a distance" effects. Hence,

$$\mathfrak{F}[\mathbf{S}(\boldsymbol{x}, \cdot), \mathbf{E}(\boldsymbol{x}, \cdot)](\boldsymbol{x}, t) = const., \tag{1}$$

where $\mathfrak{F}$ is a continuous functional, and the "dummy" variable of integration is substituted in place of the dots.

Eq. (1) can be developed into a *Volterra functional series* (see, e.g., Walters [19] and Bird et al. [20, §9.6]):

$$\begin{aligned} &M^{(0)}(t) + \int_{-\infty}^{t} M^{(1)}(t-s;t)\mathbf{S}(\boldsymbol{x}, s)\,\mathrm{d}s + \cdots \\ &+ \sum_{j=2}^{\infty} \int_{-\infty}^{t} \cdots \int \frac{1}{j!} M^{(j)}(t-s_1, \ldots, t-s_j; t) \prod_{l=1}^{j} \mathbf{S}(\boldsymbol{x}, s_l)\,\mathrm{d}s_l \\ &= K^{(0)}(t) + \int_{-\infty}^{t} K^{(1)}(t-s;t)\mathbf{E}(\boldsymbol{x}, s)\,\mathrm{d}s + \cdots \\ &+ \sum_{j=2}^{\infty} \int_{-\infty}^{t} \cdots \int \frac{1}{j!} K^{(j)}(t-s_1, \ldots, t-s_j; t) \prod_{l=1}^{j} \mathbf{E}(\boldsymbol{x}, s_l)\,\mathrm{d}s_l. \end{aligned} \tag{2}$$

Let us further assume that the constitutive relation (1) does not depend explicitly on time, i.e., the functional $\mathfrak{F}$ is *stationary*, or *time invariant* [21], so that the kernels $M^{(0)}$, $K^{(0)} = const.$, and the kernels $M^{(j)}$, $K^{(j)}$ are functions of the "dummy" variable only. Since the fluid is isotropic, the kernels are scalar functions of their argument.[3] Also, requiring that zero stress produces zero strain (i.e., we do not consider plasticity), together with the time-invariance of the constitutive relation, implies that $M^{(0)} = K^{(0)} = 0$.

Eq. (2) is the most general nonlocal (functional) dependence of the stress on the rate of strain as first proposed by Green and Rivlin [22] from a different perspective. The memory effects are modeled for all time, i.e., from $t = -\infty$, without loss of generality, since a cutoff from fading (or somehow limited) memory can be introduced through the kernels. The upper limit of integration is t so that the relation is causal, i.e., $\mathbf{S}$ (and therefore $\mathbf{T}$) depends only on the values of $\mathbf{E}$ for the instants of time prior to the current one.

2.1. Linearized memory relations

When the functional $\mathfrak{F}$ in (1) is *linear* in its two arguments, (2) reduces to

$$\int_0^\infty M(\zeta)\mathbf{S}(t-\zeta)\,\mathrm{d}\zeta = \int_0^\infty K(\zeta)\mathbf{E}(t-\zeta)\,\mathrm{d}\zeta \tag{3}$$

after the change of variables $\zeta = t - s$. The superscript "(1)" on the kernels is omitted for the sake of simplicity of notation. Furthermore, for consistency with Navier–Stokes theory, we assume that $\int_0^\infty M(\zeta)\,\mathrm{d}\zeta = 1$ and $\int_0^\infty K(\zeta)\,\mathrm{d}\zeta \neq 0$. Under mild restrictions on the kernels, one can resolve (3), using the Laplace transform and the convolution theorem, into $\mathbf{S} = \int_0^\infty \mathcal{K}(\zeta)\mathbf{E}(t-\zeta)\,\mathrm{d}\zeta$ (strain memory only) or $\mathbf{E} = \int_0^\infty \mathcal{M}(\zeta)\mathbf{S}(t-\zeta)\,\mathrm{d}\zeta$ (stress memory only). The former case is related to the classic memory assumption of Coleman and Noll [23,24], which is recovered if a Dirac delta is stipulated to be part of the resolved kernel. The latter case gives the implicit "twin" of the Coleman–Noll theory. Though the kernels M and K in (3) may be well-behaved for fast fading memory, after the resolution with respect to either $\mathbf{S}$ or $\mathbf{E}$, the effective kernels $\mathcal{M}$ and $\mathcal{K}$ do not necessarily have the same smoothness properties. In other words, *it may not always be desirable to separate relaxation from retardation in the general linear constitutive relation* (3).

2.2. Differential constitutive relations

Constitutive relations involving derivatives of $\mathbf{S}$ and $\mathbf{E}$ have been used extensively in the last couple of decades [25]. To motivate such differential approximations of the rheology with memory, we expand the tensors $\mathbf{S}(t-\zeta)$ and $\mathbf{E}(t-\zeta)$ into Taylor series about $t = 0$ (see also [26] for a related derivation in the hyperbolic heat conduction context):

$$\mathbf{S}(t-\zeta) = \sum_{j=0}^{\infty} \frac{(-\zeta)^j}{j!}\mathbf{S}^{(j)}(t), \quad \mathbf{E}(t-\zeta) = \sum_{j=0}^{\infty} \frac{(-\zeta)^j}{j!}\mathbf{E}^{(j)}(t). \tag{4}$$

Substituting the latter expressions into (3), we obtain

$$\mathbf{S} + \tau_1\dot{\mathbf{S}} + \tau_2\ddot{\mathbf{S}} + \cdots = \mu_0(\mathbf{E} + \mu_1\dot{\mathbf{E}} + \mu_2\ddot{\mathbf{E}} + \cdots), \tag{5}$$

where $\tau_0 = 1$, $\tau_j := \frac{(-1)^j}{j!}\int_0^\infty \zeta^j M(\zeta)\,\mathrm{d}\zeta$ $(j \geq 1)$, $\mu_0 = \int_0^\infty K(\zeta)\,\mathrm{d}\zeta$ and $\mu_0\mu_j := \frac{(-1)^j}{j!}\int_0^\infty \zeta^j K(\zeta)\,\mathrm{d}\zeta$ $(j \geq 1)$; τ_j, μ_j $(j \geq 1)$ carry units of time^j, while $\mu_0(>0)$ is the viscosity understood in the sense of Navier–Stokes theory. The general differential constitutive relation (5) was anticipated by Burgers [27].

The terms with derivatives on the left-hand side of (5) are called ("generalized") *relaxations*, while the respective terms on the right-hand side of (5) are termed ("generalized") *retardations*.[4] Respectively, the coefficients τ_j are the "generalized relaxation times," while the μ_j are the "generalized retardation times." Note that we have changed the primes to dots in order to emphasize the fact that these are derivatives with respect to t. For the present purposes, it suffices to identify these with ordinary time derivatives, and henceforth $\dot{(\,)} \equiv \partial_t(\cdot) \equiv \partial(\cdot)/\partial t$. However, going beyond unidirectional flows in stationary media, one has to replace them with properly invariant convected time rates [28–31].

Finally, it is important to note that a nonlocal rheology of differential type may only be used when all the integrals defining each τ_j and μ_j exist. The issue was brought up by Coleman and Markovitz [32, §2] and elucidated further by Joseph [10]. This means that the decay of the kernel at infinity must be super-algebraic (unless the expansion is truncated at some finite j); the simplest case is that of exponential decay [33–36].[5] In this case, the differential approximation can be especially good quantitatively since only the first few

[3] The kernels $M^{(j)}$ and $K^{(j)}$ are related to the Fréchet derivatives of the functional $\mathfrak{F}$ in (1) [21], which makes the Volterra expansion analogous to a Taylor series. Its convergence is beyond the scope of the present work, however.

[4] Another name for the physical effect described by the word 'retardation' is *elastic hysteresis* due to internal friction [27, p. 19].

[5] If the fading memory follows a power law $\zeta^{-\beta}$, $\beta \in (0, 1)$, then even the integral defining τ_1 and/or μ_1 can diverge, and the differential constitutive relation will feature a fractional-order derivative, if it exists at all. In heat conduction through a polydisperse suspension (see, e.g., [37]), one has $\frac{1}{\Gamma(\beta)}\int_0^t (t-s)^{\beta-1}\mathbf{E}(s)\,\mathrm{d}s \equiv {}_0D_t^{-\beta}\mathbf{E}$, i.e., the Riemann–Liouville fractional integral [38, §1.1], as the right-hand side of (3).