

## Motions with superposed proportional stretch histories as applied to combined steady and oscillatory flows of simple fluids

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VARIOUS problems connected with viscoelastic fluid behaviour under combined steady and oscillatory shearing flows are very important from the theoretical as well as the experimental point of view (cf. [1–11]). In the present paper, we apply the theory of motions with superposed proportional stretch histories (cf. [12]) to the case of two motions, one of which is always steady, while the other is any small unsteady motion. In particular, shearing oscillations superposed on steady shear, and extensional oscillations superposed on steady extension are discussed in greater detail. Certain rheological relations and their implications for incompressible simple fluids are also presented.

Różne zagadnienia związane z zachowaniem się cieczy lepkosprężystych w przepływach złożonych z ustalonego i oscylacyjnego ścinania są bardzo ważne zarówno z teoretycznego jak i doświadczalnego punktu widzenia (por. [1–11]). W niniejszej pracy zastosowano teorię ruchów z nałożonymi proporcjonalnymi historiami deformacji (por. [12]) do przypadku dwóch ruchów, z których jeden jest ustalony, a drugi jest dowolnym małym ruchem nieustalonym. W szczególności rozważono dokładniej oscylacje ścinające, nałożone na ustalone ścinanie oraz oscylacje rozciągające nałożone na ustalone rozciąganie. Przedstawiono także pewne zależności reologiczne i ich implikacje dla nieściśliwych cieczy prostych.

Различные задачи, связанные с поведением вязкоупругих жидкостей в течениях, состоящих из наложения установившегося и осциллирующего сдвиговых движений, имеют весьма большое значение, как с теоретической, так и экспериментальной точек зрения ([1–11]). В данной работе теория течений с наложенными пропорциональными историями деформаций ([12]) применяется к двум типам движений, одно из которых установившееся, а второе соответствует произвольно малому неустановившемуся течению. В частности, более подробно исследованы сдвиговые осцилляции, наложенные на установившуюся деформацию сдвига, а также растягивающие осцилляции, наложенные на установившееся растяжение. Выведены также реологические зависимости и изучены вытекающие из них следствия для несжимаемых простых жидкостей.

### 1. Introduction

VARIOUS problems connected with viscoelastic fluid behaviour under combined oscillatory and steady shearing flows have recently attracted the attention of many theoreticians and experimentalists (cf. [1–11]). A serious need for such investigations results from increasing interest in polymer rheology as well from more developed experimental techniques. Arbitrary superposed flows on one hand give more information on material properties and, on the other, they better resemble many practical situations in polymer processing when steady flows are disturbed by unsteady flows, usually of oscillatory type.

The most general and comprehensive theoretical approach to the problem has been presented by Pipkin and his collaborators [2, 3] in connection with flows which may be considered as “nearly viscometric flows” of a simple fluid. Many other authors, including

WALTERS and JONES [7, 8], TANNER and SIMMONS [4, 5], were involved in similar problems but for different models of fluids.

In the present paper, we reconsider the whole problem, on the basis of the previously formulated theory of "motions with superposed proportional stretch histories" [12]. Some results implied by this approach are well known, while others are quite new. In the first three sections, and partly in Sec. 5, we develop a theory of two superposed motions for the case of small disturbances imposed on steady viscometric flows and steady extensional flows, respectively. Section 4 is devoted to the more specialized case of small shearing motions of oscillatory type superposed on steady shear. There are considered also the cases of both parallel and transverse oscillations. Certain relations satisfied by the material functionals introduced are briefly discussed. In Sec. 6, we present some conditions which are implied by "the rheological relations" derived for BKZ fluids (cf. [6, 9, 13, 14]). In the last Sec. 7, we briefly repeat the most important conclusions formulated on the basis of the present considerations.

## 2. Motions with superposed proportional stretch histories

In our previous paper [12] on the theory of flows with proportional stretch histories, we introduced the class of "motions with proportional stretch histories" — i.e., motions in which the tensor exponent describing the stretch of a fluid element is proportional to a single, smooth function of time. We also introduced the following definition of "motions with superposed proportional stretch histories" (hereafter called briefly MSPSH):

DEFINITION. A motion is called a MSPSH, if and only if, relative to a fixed reference configuration at time 0, the deformation gradient at time  $\tau$  is given by

$$(2.1) \quad \mathbf{F}_0(\tau) = \mathbf{Q}(\tau) \exp \left( \sum_{i=1}^n \mathbf{M}_i k_i(\tau) \right), \quad \mathbf{Q}(0) = \mathbf{1},$$

where  $\mathbf{Q}(\tau)$  is an orthogonal tensor,  $\mathbf{M}_i$  are mutually commuting constant tensors,  $k_i(\tau)$  are arbitrary smooth functions of time such that  $k_i(0) = 0$  for all subscripts  $i$ .

Thus we can also write

$$(2.2) \quad \mathbf{F}_0(\tau) = \mathbf{Q}(\tau) \prod_{i=1}^n \exp(\mathbf{M}_i k_i(\tau)), \quad \text{if } \mathbf{M}_i \mathbf{M}_j = \mathbf{M}_j \mathbf{M}_i \quad \text{for } i \neq j.$$

Introducing the auxiliary history functions

$$(2.3) \quad g_i(s) \stackrel{\text{def}}{=} k_i(t-s) - k_i(t), \quad s \in [0, \infty),$$

where  $t$  denotes actual instant of time, we arrive at the history of the relative right Cauchy-Green deformation tensors (cf. [15]) in the form:

$$(2.4) \quad \mathbf{C}(s) = \mathbf{F}^T(s) \mathbf{F}(s) = \prod_{i=1}^n \exp(\mathbf{N}_i^T g_i(s)) \prod_{i=1}^n \exp(\mathbf{N}_i g_i(s)),$$

where  $\mathbf{F}(s)$  is the history of the relative deformation gradient, and

$$(2.5) \quad \mathbf{N}_i \equiv \mathbf{N}_i(t) = \mathbf{Q}(t) \mathbf{M}_i \mathbf{Q}^T(t) = \mathbf{L}_i(t) / \dot{k}_i(t).$$

The quantities  $\mathbf{L}_i = \mathbf{N}_i k_i(t)$  — called rotated parametric tensors (cf. [12]) — may be interpreted as velocity gradients at time  $t$ , measured in a rotating reference frame, the rotation of which is determined by the time-dependent tensor  $\mathbf{Q}(t)$ .

If for a given motion,  $\mathbf{Q}$  is identically equal to unity, the tensors  $\mathbf{L}_i(t)$  are nothing but the velocity gradients for component motions. This is the case of spatially homogeneous motions — i.e., motions which are homogeneous with respect to all spatial coordinates. Other interesting properties of MSPSH are discussed elsewhere [12].

For the present purposes, we briefly discuss the case of MSPSH composed from two simpler motions. Bearing in mind that  $\mathbf{M}_i$  or  $\mathbf{N}_i$  are mutually commuting tensors, we obtain for  $i = 1, 2$  either

$$(2.6) \quad \mathbf{C}(s) = \exp(\mathbf{N}_1^T g_1(s)) \mathbf{C}_2(s) \exp(\mathbf{N}_1 g_1(s)),$$

or

$$(2.7) \quad \mathbf{C}(s) = \exp(\mathbf{N}_2^T g_2(s)) \mathbf{C}_1(s) \exp(\mathbf{N}_2 g_2(s)),$$

where we have denoted

$$(2.8) \quad \mathbf{C}_i(s) = \exp(\mathbf{N}_i^T g_i(s)) \exp(\mathbf{N}_i g_i(s)), \quad i = 1, 2.$$

In what follows, we shall understand  $\mathbf{C}_1(s)$  as the deformation tensor for a fundamental (primary) motion, while  $\mathbf{C}_2(s)$  denotes the deformation tensor for an additional (secondary) motion which is superposed on the fundamental one.

If the resulting motion is isochoric, it results from (2.6) or (2.7) that

$$(2.9) \quad \det \mathbf{C}(s) = \det \mathbf{C}_1(s) \det \mathbf{C}_2(s) = 1.$$

If, moreover, the fundamental motion is of the same type, then (2.9) implies that also  $\det \mathbf{C}_2(s) = 1$ . We should bear in mind, however, that for isochoric motions also

$$(2.10) \quad \text{tr} \mathbf{L}_1 = \text{tr} \mathbf{L}_2 = \text{tr} \mathbf{N}_1 = \text{tr} \mathbf{N}_2 = 0.$$

The constitutive equation for incompressible simple fluids can be written in the form (cf. [12, 15]):

$$(2.11) \quad \mathbf{T}_E(t) = \mathbf{T}(t) + p \mathbf{1} = \mathcal{F}_{s=0}^{\infty} (\mathbf{C}(s) - \mathbf{1}), \quad \text{tr} \mathbf{T}_E = 0,$$

where  $\mathbf{T}_E$  is the extra stress tensor,  $\mathbf{T}$  is the stress tensor,  $p$  — the hydrostatic pressure, and  $\mathcal{F}$  denotes the isotropic functional, the domain of which is the space of all symmetric deformation histories.

According to the notations

$$(2.12) \quad \mathcal{F}_{s=0}^{\infty} \left( \exp(\mathbf{N}_1^T g_1(s)) (\mathbf{G}(s) + \mathbf{1}) \exp(\mathbf{N}_1 g_1(s)) - \mathbf{1} \right) = \mathcal{G}_{s=0}^{\infty} (g_1(s), \mathbf{G}(s); \mathbf{N}_1),$$

$$(2.13) \quad \mathbf{G}(s) \stackrel{\text{def}}{=} \mathbf{C}_2(s) - \mathbf{1},$$

where  $\mathcal{G}$  is a functional of the scalar function  $g_1(s)$  and the tensor function  $\mathbf{G}(s)$ , and a function of the tensor argument  $\mathbf{N}_1$ , it is easily shown, in a manner similar to [12], that

$$(2.14) \quad \mathbf{Q} \mathcal{G}_{s=0}^{\infty} (g_1(s), \mathbf{G}(s); \mathbf{N}_1) \mathbf{Q}^T = \mathcal{G}_{s=0}^{\infty} \left( \alpha g_1(s), \mathbf{Q} \mathbf{G}(s) \mathbf{Q}^T; \frac{1}{\alpha} \mathbf{Q} \mathbf{N}_1 \mathbf{Q}^T \right),$$

for all orthogonal tensors  $\mathbf{Q}$  and all real non-vanishing  $\alpha$ . Therefore, the extra stress tensor for two superposed motions is determined by

$$(2.15) \quad \mathbf{T}_E(t) = \int_{s=0}^{\infty} \mathcal{G}(g_1(s), \mathbf{G}(s); \mathbf{N}_1).$$

We can also use the following alternative form:

$$(2.16) \quad \mathbf{T}_E(t) = \int_{s=0}^{\infty} \mathcal{K}(g_1(s), g_2(s); \mathbf{N}_1, \mathbf{N}_2),$$

where  $\mathcal{K}$  is a functional in  $g_1(s)$ ,  $g_2(s)$ , and a function in  $\mathbf{N}_1, \mathbf{N}_2$ .

For certain classes of additional motions — e.g., for motions which are small in the sense of history, we can achieve more explicit forms of (2.15), (2.16), taking into account asymptotic expansions of the functionals. In accordance with the principle of fading memory (cf. [15]), if  $\mathcal{G}$  is partially Fréchet differentiable of the class  $\mathcal{C}^1$  in  $\mathbf{G}(s)$  at the neighbourhood of the zero history, simultaneously being continuous in  $g_1(s)$  and  $\mathbf{N}_1$ , we arrive at

$$(2.17) \quad \int_{s=0}^{\infty} \mathcal{G}(g_1(s), \mathbf{G}(s); \mathbf{N}_1) = \int_{s=0}^{\infty} \mathcal{G}(g_1(s), \mathbf{0}; \mathbf{N}_1) + \delta_{\mathbf{G}} \int_{s=0}^{\infty} \mathcal{G}(g_1(s), |\mathbf{G}(s); \mathbf{N}_1) + o(\|\mathbf{G}(s)\|^2),$$

where  $\delta_{\mathbf{G}} \mathcal{G}$  is a linear functional in  $\mathbf{G}(s)$ , and

$$(2.18) \quad \lim_{\|\mathbf{G}\| \rightarrow 0} \frac{o(\|\mathbf{G}\|^2)}{\|\mathbf{G}\|^2} = 0, \quad \text{tr} \int_{s=0}^{\infty} \mathcal{G}(g_1(s), \mathbf{G}(s); \mathbf{N}_1) = 0.$$

It should be emphasized that further simplifications in the representation form (2.17) may be achieved on the basis of more specialized forms for  $g_1(s)$  and  $\mathbf{G}(s)$ . In the next two sections, we discuss in greater detail the cases of small time-dependent motions superposed on viscometric flows and extensional flows, respectively. For such motions, the terms of order  $o(\|\mathbf{G}\|^2)$  may be disregarded.

### 3. Small motions superposed on viscometric flows

Now, let us assume that the fundamental motion is represented by a viscometric flow (cf. [15]), while the additional motion is any small unsteady non-viscometric flow, the description of which through the constitutive equations (2.17) is admissible.

This assumption means that

$$(3.1) \quad \text{tr} \mathbf{N}_1 = 0, \quad \text{tr} \mathbf{N}_1^2 = 0, \quad \text{tr} \mathbf{N}_1 \mathbf{N}_1^T = \text{tr} \mathbf{N}_1^T \mathbf{N}_1 = \kappa^2,$$

where  $\kappa$  denotes amount of shear in a viscometric flow, and

$$(3.2) \quad \int_{s=0}^{\infty} \mathcal{G}(g_1(s), \mathbf{0}; \mathbf{N}_1) = \int_{s=0}^{\infty} \mathcal{t}(g_1(s); \kappa^2)(\mathbf{N}_1 + \mathbf{N}_1^T) \\ + \int_{s=0}^{\infty} \mathcal{s}_1(g_1(s); \kappa^2) \mathbf{N}_1 \mathbf{N}_1^T + \int_{s=0}^{\infty} \mathcal{s}_2(g_1(s); \kappa^2) \mathbf{N}_1^T \mathbf{N}_1.$$

The functionals  $\mathcal{t}$ ,  $\mathcal{s}_1$  and  $\mathcal{s}_2$  are functions of the basic invariants which are expressible by  $\kappa^2$ .

In the case of steady fundamental viscometric flows, since  $g_1(s) = -s$ , we shall use the notations:

$$(3.3) \quad \begin{aligned} \tau(\kappa) &= \kappa \eta(\kappa) = \kappa \int_{s=0}^{\infty} (-s; \kappa^2), \\ \sigma_1(\kappa) &= \kappa^2 \int_{s=0}^{\infty} S_1(-s; \kappa^2), \quad \sigma_2(\kappa) = \kappa^2 \int_{s=0}^{\infty} S_2(-s; \kappa^2). \end{aligned}$$

It is well known that the shear stress function  $\tau(\kappa)$  is an odd function, while the normal stress functions are even functions of the argument  $\kappa$  (cf. [15]).

Since the second term on the right-hand side of (2.17) can always be represented by an integral, we obtain

$$(3.4) \quad \delta_{\mathbf{G}} \int_{s=0}^{\infty} \mathfrak{F}(g_1(s), |\mathbf{G}(s); \mathbf{N}_1) = \int_0^{\infty} \Phi(g_1(s); \mathbf{N}_1) [\mathbf{G}(s)] ds,$$

where  $\Phi[ ]$  denotes the linear transformation of the tensor indicated in brackets. An explicit representation of the integrand may be taken in the form:

$$(3.5) \quad \begin{aligned} \Phi(g_1(s); \mathbf{N}_1) [\mathbf{G}(s)] &= \varphi_0 \mathbf{G} + \varphi_1 (\mathbf{G}(\mathbf{N}_1 + \mathbf{N}_1^T) + (\mathbf{N}_1 + \mathbf{N}_1^T) \mathbf{G}) + \varphi_2 (\mathbf{G} \mathbf{N}_1^T \mathbf{N}_1 \\ &+ \mathbf{N}_1^T \mathbf{N}_1 \mathbf{G}) + \varphi_3 (\mathbf{G} \mathbf{N}_1 \mathbf{N}_1^T + \mathbf{N}_1 \mathbf{N}_1^T \mathbf{G}) + \varphi_4 (\mathbf{N}_1 \mathbf{G} + \mathbf{G} \mathbf{N}_1^T) + \varphi_5 (\mathbf{G} \mathbf{N}_1 + \mathbf{N}_1^T \mathbf{G}) + \varphi_6 \mathbf{N}_1 \mathbf{G} \mathbf{N}_1^T \\ &+ \varphi_7 \mathbf{N}_1^T \mathbf{G} \mathbf{N}_1 + \varphi_8 (\mathbf{N}_1 \mathbf{G} \mathbf{N}_1 + \mathbf{N}_1^T \mathbf{G} \mathbf{N}_1^T) + \varphi_9 (\mathbf{N}_1 + \mathbf{N}_1^T) \text{tr}(\mathbf{N}_1 \mathbf{G}) + \varphi_{10} \mathbf{N}_1^T \mathbf{N}_1 \text{tr}(\mathbf{N}_1 \mathbf{G}) \\ &+ \varphi_{11} \mathbf{N}_1 \mathbf{N}_1^T \text{tr}(\mathbf{N}_1 \mathbf{G}) + \varphi_{12} (\mathbf{N}_1 + \mathbf{N}_1^T) \text{tr}(\mathbf{N}_1 \mathbf{G} \mathbf{N}_1^T) + \varphi_{13} \mathbf{N}_1^T \mathbf{N}_1 \text{tr}(\mathbf{N}_1 \mathbf{G} \mathbf{N}_1^T) \\ &+ \varphi_{14} \mathbf{N}_1 \mathbf{N}_1^T \text{tr}(\mathbf{N}_1 \mathbf{G} \mathbf{N}_1^T) + \varphi_{15} (\mathbf{N}_1 + \mathbf{N}_1^T) \text{tr}(\mathbf{N}_1^T \mathbf{G} \mathbf{N}_1) + \varphi_{16} \mathbf{N}_1^T \mathbf{N}_1 \text{tr}(\mathbf{N}_1^T \mathbf{G} \mathbf{N}_1) \\ &+ \varphi_{17} \mathbf{N}_1 \mathbf{N}_1^T \text{tr}(\mathbf{N}_1^T \mathbf{G} \mathbf{N}_1), \end{aligned}$$

where, for simplicity, we have denoted:  $\mathbf{G} \equiv \mathbf{G}(s)$ , and where  $\varphi_i$  ( $i = 0, \dots, 17$ ) must be considered as functions in  $g_1(s)$  and functions in  $\kappa^2$  — i.e.,  $\varphi_i = \varphi_i(g_1(s); \kappa^2)$ .

The terms involving  $\mathbf{P}_1$  as well as those involving  $\text{tr} \mathbf{G}(s)$  have been omitted in (3.5). Following the arguments of PIPKIN [1, 2], the terms  $\mathbf{P}_1$ , as being proportional to  $\mathbf{C}_2^{-1}(s)$ , may be expressed by  $\mathbf{G}$ ,  $\mathbf{G}^2$ ,  $\mathbf{G}^3$ , etc. — i.e., higher order terms in sense of the norm  $\|\mathbf{G}(s)\|$ . Similarly, from the Cayley-Hamilton theorem (cf. [16]) it results that

$$(3.6) \quad \text{tr} \mathbf{G} - \frac{1}{2} \text{tr} \mathbf{G} \text{tr} \mathbf{G}^2 + \frac{1}{6} (\text{tr} \mathbf{G})^3 + \frac{1}{3} \text{tr} \mathbf{G}^3 + \frac{1}{2} [(\text{tr} \mathbf{G})^2 - \text{tr} \mathbf{G}^2] = 0;$$

which means that the terms involving  $\text{tr} \mathbf{G}$  are also redundant in (3.5).

Further reductions in (3.5) may be justified as follows. For viscometric flows characterized by  $\mathbf{N}_1$

$$(3.7) \quad \begin{aligned} \mathbf{C}(s) &= (\mathbf{1} + \mathbf{N}_1^T g_1(s)) \mathbf{C}_2(s) (\mathbf{1} + \mathbf{N}_1 g_1(s)) \\ &= \mathbf{C}_2 (\mathbf{1} + \mathbf{N}_1 g_1(s) + \mathbf{C}_2^{-1} \mathbf{N}_1^T \mathbf{C}_2 g_1(s) + \mathbf{C}_2^{-1} \mathbf{N}_1^T \mathbf{C}_2 \mathbf{N}_1 g_1^2(s)), \end{aligned}$$

and since  $\mathbf{C}_2^{-1} = \mathbf{1} - \mathbf{G} + o(\|\mathbf{G}\|^2)$ ,

$$(3.8) \quad \begin{aligned} \det \mathbf{C}(s) &= \det \mathbf{C}_2 \det (\mathbf{1} + \mathbf{N}_1^T g_1(s) + \mathbf{N}_1 g_1(s) - \mathbf{G} \mathbf{N}_1^T g_1(s) \\ &+ \mathbf{N}_1^T \mathbf{G} g_1(s) + \mathbf{N}_1^T \mathbf{N}_1 g_1^2(s) - \mathbf{G} \mathbf{N}_1^T \mathbf{N}_1 g_1^2(s) + o(\|\mathbf{G}\|^2)). \end{aligned}$$

Thus, for incompressible flows,

$$(3.9) \quad \det \mathbf{C}_1(s) = 1 + \text{tr}(\mathbf{N}_1^T \mathbf{N}_1) g_1^2(s) - \text{tr}(\mathbf{N}_1 \mathbf{G} \mathbf{N}_1^T) g_1^2(s) + o(\|\mathbf{G}\|^2),$$

and, if the terms of order  $o(\|\mathbf{G}\|^2)$  are disregarded,

$$(3.10) \quad \text{tr} \mathbf{N}_1^T \mathbf{G} \mathbf{N}_1 = \text{tr} \mathbf{N}_1 \mathbf{G} \mathbf{N}_1^T - \text{tr} \mathbf{N}_1^T \mathbf{N}_1.$$

This means that  $\varphi_{15}$ ,  $\varphi_{16}$  and  $\varphi_{17}$  may be rejected from (3.5).

Let us observe, moreover, that the normalization condition:  $\text{tr} \mathbf{T}_E = 0$  leads to the relation (cf. (2.18)<sub>2</sub>):

$$(3.11) \quad \begin{aligned} \varphi_0 \text{tr} \mathbf{G} + 4\varphi_1 \text{tr} \mathbf{G} \mathbf{N}_1 + 2\varphi_2 \text{tr} \mathbf{N}_1 \mathbf{G} \mathbf{N}_1^T + 2\varphi_3 \text{tr} \mathbf{N}_1^T \mathbf{G} \mathbf{N}_1 + 2\varphi_4 \text{tr} \mathbf{G} \mathbf{N}_1 + 2\varphi_5 \text{tr} \mathbf{G} \mathbf{N}_1 \\ + \varphi_6 \text{tr} \mathbf{N}_1 \mathbf{G} \mathbf{N}_1^T + \varphi_7 \text{tr} \mathbf{N}_1^T \mathbf{G} \mathbf{N}_1 + \varphi_{10} \text{tr}(\mathbf{G} \mathbf{N}_1) \text{tr} \mathbf{N}_1^T \mathbf{N}_1 + \varphi_{11} \text{tr}(\mathbf{G} \mathbf{N}_1) \text{tr} \mathbf{N}_1^T \mathbf{N}_1 \\ + \varphi_{13} \text{tr}(\mathbf{N}_1 \mathbf{G} \mathbf{N}_1^T) \text{tr} \mathbf{N}_1^T \mathbf{N}_1 + \varphi_{14} \text{tr}(\mathbf{N}_1 \mathbf{G} \mathbf{N}_1^T) \text{tr} \mathbf{N}_1^T \mathbf{N}_1 = 0. \end{aligned}$$

Disregarding the term  $\varphi_0 \text{tr} \mathbf{G}$  and taking into account (3.10), we arrive at  $\varphi_7 = -\varphi_3$ , and

$$(3.12) \quad \begin{aligned} \text{tr} \mathbf{G} \mathbf{N}_1 (4\varphi_1 + 2\varphi_4 + 2\varphi_5 + \kappa^2 \varphi_{10} + \kappa^2 \varphi_{11}) \\ + \text{tr}(\mathbf{N}_1 \mathbf{G} \mathbf{N}_1^T) ((2\varphi_2 + 2\varphi_3 + \varphi_6 + \varphi_7 + \kappa^2 \varphi_{13} + \kappa^2 \varphi_{14}) = 0, \end{aligned}$$

Since (3.12) is to be satisfied for any positive or negative tensor  $\mathbf{N}_1$  (the change of shear direction), the both expressions in parantheses in (3.12) must be equal to zero. Therefore, any two of the functions occurring in these parantheses can be expressed by the remaining terms. If we choose  $\varphi_{11}$  and  $\varphi_{14}$  as redundant terms, we finally obtain only 12 independent material functions. Let us denote them as follows:

$$(3.13) \quad \begin{aligned} \psi_i(g_1(s); \kappa^2) &= \varphi_i, & \text{for } i &= 0, 1, \dots, 6, \\ \psi_{i-1}(g_1(s); \kappa^2) &= \varphi_i & \text{for } i &= 8, \dots, 10, \\ \psi_{10}(g_1(s); \kappa^2) &= \varphi_{12}, & \psi_{11}(g_1(s); \kappa^2) &= \varphi_{13}. \end{aligned}$$

The number of independent material functions is therefore less by one than that obtained by PIPKIN [1, 2], for the case of nearly viscometric flows of incompressible fluids.

Similarly, the condition (2.9), with  $\det \mathbf{C}_1(s) = 1$ , is equivalent to that derived by PIPKIN and OWEN [2]. Introducing the new strain tensor

$$(3.14) \quad \mathbf{E}(s) = \mathbf{C}(s) - \mathbf{C}_1(s),$$

we see that

$$(3.15) \quad \det \mathbf{C}(s) = \det(\mathbf{C}_1 + \mathbf{E}) = \det \mathbf{C}_1 \det(\mathbf{1} + \mathbf{C}_1^{-1} \mathbf{E}) = 1 + \text{tr} \mathbf{C}_1^{-1} \mathbf{E} + o(\|\mathbf{E}\|^2) = 1,$$

and, if the corresponding higher order terms may be disregarded,

$$(3.16) \quad \text{tr}(\mathbf{C}_1^{-1}(s) \mathbf{E}(s)) = 0.$$

#### 4. Small shearing oscillations superposed on steady shear

Let us consider the case in which the fundamental motion is steady shearing flow, while the additional motion is unsteady shearing flow. This assumption concerning shearing motions is not necessary; the main results still remain valid if we consider two superposed viscometric flows.

In the case of viscometric or shearing flows, we have

$$(4.1) \quad \mathbf{G}(s) = g_2(s) (\mathbf{N}_2^T + \mathbf{N}_2) + g_2^2(s) \mathbf{N}_2^T \mathbf{N}_2, \quad \mathbf{N}_2^2 = \mathbf{N}_2^T \mathbf{N}_2 = \mathbf{0}.$$

If the additional motion is of oscillatory type, the function  $k_2(\tau)$ , responsible for its time-dependence, may be written in the form:

$$(4.2) \quad k_2(\tau) = -ie^{i\omega\tau}, \quad k_2(0) = 0, \quad k_2(\tau) = \omega e^{i\omega\tau},$$

and

$$(4.3) \quad g_2(s) = ie^{i\omega t}(1 - e^{-i\omega s}), \quad g_2^2(s) = -e^{2i\omega t}(1 - e^{-i\omega s})^2,$$

where only real parts of functions are meaningful.

In considering shearing motions, there exist two possibilities: the additional shear is realized either in the plane parallel to the fundamental motion or in the plane perpendicular (transverse) to the fundamental shear. In the case of oscillatory shearing flows, we can speak about in-line oscillations or about cross oscillations (cf. [3, 5]). We shall discuss these two cases separately.

#### 4.1. The case of parallel flows (in-line oscillations)

For parallel shearing flows, we have in Cartesian coordinates

$$(4.1.1) \quad [N_1] = \begin{bmatrix} 0 & \kappa & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad [N_2] = \begin{bmatrix} 0 & \alpha & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad N_1 N_2 = N_2 N_1,$$

where  $\kappa$  and  $\alpha$  are shear gradients for the fundamental and additional flows, respectively. On the basis of (2.15), (2.17), (3.3), (3.5) and (4.1), we obtain the following extra stress components:

$$(4.1.2) \quad \begin{aligned} T_E^{23} &= T_E^{13} = 0, \\ T_E^{12} &= \tau(\kappa) + \int_0^\infty (\psi_0 + \kappa^2 \psi_2 + \kappa^2 \psi_3 + \kappa^2 \psi_7 + \kappa^2 \psi_8) g_2(s) \alpha ds \\ &\quad + \int_0^\infty (\kappa \psi_1 + \kappa \psi_4 + \kappa^3 \psi_{10}) g_2^2(s) \alpha^2 ds, \\ T_E^{11} &= \sigma_1(\kappa) + \int_0^\infty (2\kappa \psi_1 + 2\kappa \psi_4) g_2(s) \alpha ds + \int_0^\infty \psi_6 \kappa^2 g_2^2(s) \alpha^2 ds, \\ T_E^{22} &= \sigma_2(\kappa) + \int_0^8 (2\kappa \psi_1 + 2\kappa \psi_5 + 2\kappa^3 \psi_9) g_2(s) \alpha ds \\ &\quad + \int_0^\infty (\psi_0 + 2\kappa^2 \psi_2 + \kappa^4 \psi_{11}) g_2^2(s) \alpha^2 ds, \\ T_E^{33} &= 0, \end{aligned}$$

where  $\psi_i$  ( $i = 0, 1, \dots, 11$ ) are the kernels of the form:  $\psi_i = \psi_i(-s; \kappa^2)$ .

The above expressions are valid for small finite deformations superposed on steady shear. In the case of infinitesimal deformations or infinitesimal oscillations characterized

by the amplitude  $\alpha$ , all second integrals on the right-hand side of (4.1.2) may be disregarded as the terms of order  $O(\alpha^2)$ .

The relations (4.3) and (4.1.2) can be used to determine dynamic characteristics of an incompressible simple fluid under combined steady shear and small amplitude in-line oscillations (cf. [3, 5]). In connection with this, there arise two important questions. First, the effect of steady shear on the dynamic characteristics, and second, the effect of small oscillations on the mean characteristics of steady shear (cf. [8]). Unfortunately, these questions can be satisfactorily answered for simple fluids only in the case of very small angular velocities  $\omega$ , or more precisely for  $\omega \rightarrow 0$ .

First, let us proceed to the first question posed above. Defining the complex dynamic viscosity in the usual form as  $\mu_{12}^* = (T_E^{12} - \tau(\kappa))/\alpha\omega e^{i\omega t}$ , we obtain for infinitesimal oscillations

$$(4.1.3) \quad \mu_{12}^*(\kappa, \omega) = \frac{i}{\omega} \int_0^\infty (\psi_0 + \kappa^2\psi_2 + \kappa^2\psi_3 + \kappa^2\psi_7 + \kappa^2\psi_8)(1 - e^{-i\omega s}) ds,$$

and in the limit

$$(4.1.4) \quad \begin{aligned} \lim_{\omega \rightarrow 0} \mu_{12}^*(\kappa, \omega) &= - \int_0^\infty (\psi_0 + \kappa^2\psi_2 + \kappa^2\psi_3 + \kappa^2\psi_7 + \kappa^2\psi_8) s ds, \\ \lim_{\omega \rightarrow 0} \mu'_{12}(\kappa, \omega) &= \lim_{\omega \rightarrow 0} \mu_{12}^*(\kappa, \omega), \quad \lim_{\omega \rightarrow 0} G'_{12}(\kappa, \omega) = 0, \end{aligned}$$

where  $\mu'_{12}$  is the dynamic viscosity (real) and  $G'_{12}$  is the real part of dynamic modulus.

Introducing the notion of complex dynamic modulus  $H^*$  for the normal stress difference, namely

$$(4.1.5) \quad H^* = H' + iH'' \stackrel{\text{def}}{=} (T_E^{11} - T_E^{22} - \sigma_1(\kappa) + \sigma_2(\kappa))/-i\alpha e^{i\omega t},$$

and taking into account the consistency relations derived by PIPKIN [3], we finally obtain

$$(4.1.6) \quad \lim_{\omega \rightarrow 0} \mu'_{12}(\kappa, \omega) = - \int_0^\infty (\psi_0 + \kappa^2\psi_2 + \kappa^2\psi_3 + \kappa^2\psi_7 + \kappa^2\psi_8) s ds = \frac{d\tau(\kappa)}{d\kappa},$$

$$(4.1.7) \quad \lim_{\omega \rightarrow 0} \frac{H''(\kappa, \omega)}{\omega} = - \int_0^\infty (2\kappa\psi_4 - 2\kappa\psi_5 - \kappa^3\psi_9) s ds = \frac{d}{d\kappa} (\sigma_1(\kappa) - \sigma_2(\kappa)),$$

where  $\tau(\kappa)$ ,  $\sigma_1(\kappa)$ ,  $\sigma_2(\kappa)$  are viscometric functions defined in (3.3).

The first of the above relations is in formal consistency with the result obtained elsewhere [3], (cf. also  $R-1$  in [6, 13]). The second one is nothing but the relation  $R-3$  on Bernstein's list [6] of so called "rheological relations" (cf. Sec. 6).

It should be noted, however, that for  $\kappa \rightarrow 0$  one more relation is valid for incompressible simple fluids. On the basis of the theory of second order fluids, COLEMAN and MARKOVITZ [17] have shown that

$$(4.1.8) \quad \lim_{\omega \rightarrow 0} \frac{G'}{\omega^2} = \lim_{\kappa \rightarrow 0} \frac{\sigma_1(\kappa) - \sigma_2(\kappa)}{2\kappa^2}.$$

According to our notation, we have

$$(4.1.9) \quad \lim_{\omega \rightarrow 0} \frac{G'_{12}(\kappa, \omega)}{\omega^2} = -\frac{1}{2} \int_0^{\infty} (\psi_0 + \kappa^2 \psi_2 + \kappa^2 \psi_3 + \kappa^2 \psi_7 + \kappa^2 \psi_8) s^2 ds.$$

Since

$$(4.1.10) \quad \lim_{\kappa \rightarrow 0} \frac{\sigma_1 - \sigma_2}{\kappa^2} = \lim_{\kappa \rightarrow 0} \frac{d}{d\kappa} \left( \frac{\sigma_1 - \sigma_2}{\kappa} \right),$$

we finally arrive at the relation:

$$(4.1.11) \quad \lim_{\kappa \rightarrow 0} - \int_0^{\infty} \psi_0 s^2 ds = \lim_{\kappa \rightarrow 0} \frac{d}{d\kappa} \left( \frac{\sigma_1(\kappa) - \sigma_2(\kappa)}{\kappa} \right).$$

Now, let us proceed to the second question concerning the effect of small finite oscillations on the mean stresses in steady shear. This problem was considered theoretically and experimentally by JONES and WALTERS [8]. To follow their way of thinking, we shall take into account the full expressions for stresses in (4.1.2) — i.e., the terms of order  $O(\alpha^2)$ .

The change of mean shear stress  $\Delta \langle T_E^{12} \rangle$  depends on the following integral:

$$(4.1.12) \quad \int_0^{\infty} (\kappa \psi_1 + \kappa \psi_4 + \kappa^3 \psi_{10}) \alpha^2 \operatorname{Re} g_2(s) \operatorname{Re} g_2(s) ds,$$

where  $\operatorname{Re}$  denotes the real part. Bearing in mind that

$$(4.1.13) \quad \operatorname{Re}(g_2) \operatorname{Re}(g_2) = \frac{1}{2} [\operatorname{Re}(g_2^2) + \operatorname{Re}(g_2 \bar{g}_2)],$$

where the overbar denotes a complex conjugate, we obtain

$$(4.1.14) \quad \Delta \langle T_E^{12} \rangle = \frac{1}{2} \alpha^2 \int_0^{\infty} (\kappa \psi_1 + \kappa \psi_4 + \kappa^3 \psi_{10}) (1 - e^{-i\omega s}) (1 - e^{i\omega s}) ds.$$

Denoting by  $\epsilon = \alpha\omega/\kappa$  the small dimensionless parameter characterizing the amplitude of oscillations, we easily arrive at the following limits:

$$(4.1.15) \quad \lim_{\kappa \rightarrow 0} \Delta \langle T_E^{12} \rangle = 0 \quad \text{with } \kappa = \text{const},$$

$$(4.1.16) \quad \lim_{\omega \rightarrow 0} \Delta \langle T_E^{12} \rangle = \frac{\epsilon^2 \kappa^3}{2} \int_0^{\infty} (\psi_1 + \psi_2 + \kappa^2 \psi_{10}) s^2 ds \quad \text{with } \epsilon = \text{const}.$$

The changes of the mean extra stresses  $\Delta \langle T_E^{11} \rangle$  and  $\Delta \langle T_E^{22} \rangle$  may be achieved in the same manner. This leads to

$$(4.1.17) \quad \lim_{\omega \rightarrow 0} \Delta \langle T_E^{11} \rangle = \lim_{\omega \rightarrow 0} \Delta \langle T_E^{22} \rangle = 0 \quad \text{with } \kappa = \text{const},$$

$$(4.1.18) \quad \lim_{\omega \rightarrow 0} \Delta \langle T_E^{11} \rangle = \frac{\epsilon^2 \kappa^4}{2} \int_0^{\infty} \psi_6 s^2 ds \quad \text{with } \epsilon = \text{const}.$$

$$\lim_{\omega \rightarrow 0} \Delta \langle T_E^{22} \rangle = \frac{\epsilon^2 \kappa^2}{2} \int_0^{\infty} (\psi_0 + 2\kappa^2 \psi_2 + \kappa^4 \psi_{11}) s^2 ds$$

The above relations may be interpreted as follows. If the fundamental steady shear realized with strictly constant shear gradient  $\kappa$  is disturbed by any in-line shearing oscillations, acting with small angular velocity  $\omega$ , the mean shear and normal stresses show no change at all. On the other hand, if the fundamental motion is produced with such small shear rate that the ratio of  $\alpha\omega$  to  $\kappa$  is kept as approximately constant, the mean stresses may undergo essential changes according to (4.1.16) and (4.1.18). It is worthwhile to note that the phenomena of the type considered have been detected experimentally by JONES and WALTERS [8]. Their indirect conclusion was that any small variation from a state of steady shear may have a measurable effect on the mean conditions.

#### 4.2. The case of perpendicular flows (transverse or cross oscillations)

For perpendicular shearing flows, we have in Cartesian coordinates

$$(4.2.1) \quad [N_1] = \begin{bmatrix} 0 & \kappa & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad [N_2] = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & \alpha & 0 \end{bmatrix}, \quad N_1 N_2 = N_2 N_1.$$

Proceeding similarly as in Sec. 4.1, we obtain the following extra stress components:

$$(4.2.2) \quad \begin{aligned} T_E^{23} &= \int_0^\infty (\psi_0 + \kappa^2 \psi_2 + \kappa^2 \psi_3) g_2(s) \alpha ds, \\ T_E^{13} &= \int_0^\infty (\kappa \psi_1 + \kappa \psi_4) g_2(s) \alpha ds, \\ T_E^{12} &= \tau(\kappa) + \int_0^\infty (\kappa \psi_1 + \kappa \psi_4 + \kappa^3 \psi_{10}) g_2^2(s) \alpha^2 ds, \\ T_E^{11} &= \sigma_1(\kappa) + \int_0^\infty \kappa^2 \psi_6 g_2^2(s) \alpha^2 ds, \\ T_E^{22} &= \sigma_2(\kappa) + \int_0^\infty (\psi_0 + 2\kappa^2 \psi_2 + \kappa^4 \psi_{11}) g_2^2(s) \alpha^2 ds, \\ T_E^{33} &= 0, \end{aligned}$$

where, as previously,  $\psi_i = \psi_i(-s; \kappa^2)$ ,  $i = 0, 1, \dots, 11$ .

The latter expressions are quite different from those occurring in (4.1.2), although some integrands are similar or even identical in form. In the case of infinitesimal deformations or oscillations, only the integrals on the right-hand sides of  $T_E^{23}$  and  $T_E^{13}$  must be retained.

Proceeding to determination of dynamic characteristics for the case of combined steady shear and small amplitude cross oscillations (cf. [4, 5]), we shall bear in mind two questions mentioned in Sec. 4.1.

Defining two complex dynamic viscosities in the usual forms, we obtain for infinitesimal oscillations

$$(4.2.3) \quad \eta_{23}^*(\kappa, \omega) = \frac{i}{\omega} \int_0^{\infty} (\psi_0 + \kappa^2 \psi_2 + \kappa^2 \psi_3) (1 - e^{-i\omega s}) ds,$$

$$(4.2.4) \quad \eta_{13}^*(\kappa, \omega) = \frac{i}{\omega} \int_0^{\infty} \kappa(\psi_1 + \psi_4) (1 - e^{-i\omega s}) ds,$$

and in the limits

$$(4.2.5) \quad \lim_{\omega \leftarrow 0} \eta_{23}^*(\kappa, \omega) = - \int_0^{\infty} (\psi_0 + \kappa^2 \psi_2 + \kappa^2 \psi_3) s ds,$$

$$\lim_{\omega \rightarrow 0} \eta'_{23}(\kappa, \omega) = \lim_{\omega \rightarrow 0} \eta_{23}^*(\kappa, \omega), \quad \lim_{\omega \rightarrow 0} G'_{23}(\kappa, \omega) = 0;$$

$$(4.2.6) \quad \lim_{\omega \rightarrow 0} \eta_{13}^*(\kappa, \omega) = - \int_0^{\infty} \kappa(\psi_1 + \psi_4) s ds,$$

$$\lim_{\omega \rightarrow 0} \eta'_{13}(\kappa, \omega) = \lim_{\omega \rightarrow 0} \eta_{13}^*(\kappa, \omega), \quad \lim_{\omega \rightarrow 0} G'_{13}(\kappa, \omega) = 0,$$

where  $\eta'_{23}$ ,  $\eta'_{13}$  are the dynamic viscosities (real) and  $G'_{23}$ ,  $G'_{13}$  — the real parts of dynamic moduli, respectively.

Taking into account the consistency relations derived by Pipkin [3], we finally obtain

$$(4.2.7) \quad \lim_{\omega \rightarrow 0} \eta'_{23}(\kappa, \omega) = - \int_0^{\infty} (\psi_0 + \kappa^2 \psi_2 + \kappa^2 \psi_3) s ds = \frac{\tau(\kappa)}{\kappa} \equiv \eta(\kappa),$$

$$(4.2.8) \quad \lim_{\omega \rightarrow 0} \eta'_{13}(\kappa, \omega) = - \int_0^{\infty} \kappa(\psi_1 + \psi_4) s ds = \frac{\sigma_1(\kappa)}{\kappa},$$

where  $\tau(\kappa)$ ,  $\sigma_1(\kappa)$  and  $\sigma_2(\kappa)$  have been defined in (3.3).

The relation (4.2.7) is consistent with the well known property of dynamic viscosity functions (cf. [3, 5, 6]). The relation (4.2.8) does not occur among those listed by TANNER and WILLIAMS [9] (cf. Sec. 6). It should be emphasized that (4.2.7), (4.2.8) are the only relations resulting from consistency conditions in the case under consideration.

It is possible to obtain one additional relation taking into account (4.1.8) in Sec. 4.1. Since in the case under consideration

$$(4.2.9) \quad \lim_{\omega \leftarrow 0} \frac{G'_{23}(\kappa, \omega)}{\omega^2} = - \frac{1}{2} \int_0^{\infty} (\psi_0 + \kappa^2 \psi_2 + \kappa^2 \psi_3) s ds,$$

in the limit of  $\kappa \rightarrow 0$ , we arrive at the relation

$$(4.2.10) \quad \lim_{\kappa \rightarrow 0} - \int_0^{\infty} \psi_0 s^2 ds = \lim_{\kappa \rightarrow 0} \frac{\sigma_1(\kappa) - \sigma_2(\kappa)}{\kappa^2},$$

which is essentially the same as that in (4.1.11), and results directly from the theory of second-order fluids.

As regards the behaviour of the mean shear stresses under the influence of small finite amplitude oscillations, the results are very similar to those presented in Sec. 4.1. There are no changes in  $\langle T_E^{13} \rangle$  and  $\langle T_E^{23} \rangle$ , while the changes in  $\langle T_E^{12} \rangle$  are described by the formulae identical to (4.1.15), (4.1.16). The changes of the normal extra stresses  $\Delta \langle T_E^{11} \rangle$  and  $\Delta \langle T_E^{22} \rangle$  are determined by the formulae analogous to (4.1.17), (4.1.18).

### 5. Small motions superposed on extensional flows

Let us assume that the fundamental motion is represented by a simple extension, while the additional motion is any small unsteady non-viscometric flow, the description of which through the constitutive equations (2.17) is admissible.

We can write instead of (3.2)

$$(5.1) \quad \mathcal{G}(g_1(s), \mathbf{0}; \mathbf{N}_1) = \overset{\infty}{\underset{s=0}{\int}} (g_1(s); q) \mathbf{N}_1 + \overset{\infty}{\underset{s=0}{\int}} (g_1(s); q) \mathbf{N}_1^2,$$

since  $\mathbf{N}_1$  is symmetric, and

$$(5.2) \quad \text{tr} \mathbf{N}_1 = 0, \quad \text{tr} \mathbf{N}_1^2 = \frac{3}{2} q^2, \quad \text{tr} \mathbf{N}_1^3 = \frac{3}{4} q^3,$$

where  $q$  denotes the amount of extension. In the case of steady fundamental extensions, we shall use the following functions:

$$(5.3) \quad \alpha_1(q) = q \overset{\infty}{\underset{s=0}{\int}} (-s; q), \quad \alpha_2(q) = q^2 \overset{\infty}{\underset{s=0}{\int}} (-s; q).$$

The representation (3.5) is now replaced by

$$(5.4) \quad \Phi(g_1(s); \mathbf{N}_1) [\mathbf{G}(s)] = \beta_0 \mathbf{G} + \beta_1 (\mathbf{G} \mathbf{N}_1 + \mathbf{N}_1 \mathbf{G}) + \beta_2 (\mathbf{G} \mathbf{N}_1^2 + \mathbf{N}_1^2 \mathbf{G}) + \beta_3 \mathbf{N}_1 \text{tr}(\mathbf{N}_1 \mathbf{G}) + \\ + \beta_4 \mathbf{N}_1 \text{tr}(\mathbf{N}_1^2 \mathbf{G}) + \beta_5 \mathbf{N}_1^2 \text{tr}(\mathbf{N}_1 \mathbf{G}) + \beta_6 \mathbf{N}_1^2 \text{tr}(\mathbf{N}_1^2 \mathbf{G}),$$

where the quantities  $\beta_i$  ( $i = 0, 1, \dots, 6$ ) are functions in  $g_1(s)$  and functions in  $q$  - i.e.,  $\beta_i = \overset{\infty}{\underset{s=0}{\int}} \beta_i(g_1(s); q)$ .

The terms involving  $\mathbf{P1}$  as well as  $\text{tr} \mathbf{G}(s)$  have been omitted in (5.4) on the ground of the same arguments as those presented in Sec. 3. Moreover, terms of the type:  $\mathbf{N}_1 \mathbf{G} \mathbf{N}_1$ ,  $\mathbf{N}_1 \mathbf{G} \mathbf{N}_1^2 + \mathbf{N}_1^2 \mathbf{G} \mathbf{N}_1$ ,  $\mathbf{N}_1^2 \mathbf{G} \mathbf{N}_1^2$ , etc. are redundant on the basis of the Cayley-Hamilton theorem (cf. [16]).

Further redundancies in (5.4) result from the normalization condition:  $\text{tr} \mathbf{T}_E = 0$ . This leads to

$$(5.5) \quad \left( \beta_1 + \frac{3}{2} \beta_5 q^2 \right) \text{tr}(\mathbf{N}_1 \mathbf{G}) + \left( \beta_2 + \frac{3}{2} \beta_6 q^2 \right) \text{tr}(\mathbf{N}_1^2 \mathbf{G}) = 0,$$

which means that  $\beta_5$  and  $\beta_6$ , for example, can be replaced in (5.4) by the remaining functions. Therefore, the material properties are entirely described by  $\beta_i$  ( $i = 0, 1, \dots, 4$ ), five in number.

It is worthwhile to note that the representation (3.5), valid for asymmetric tensors  $\mathbf{N}_1$ , is more general than (5.4), valid only for symmetric  $\mathbf{N}_1$ . Although  $\beta_i$  depend on other invariants than  $\psi_i$ , we can, for sufficiently small values of  $q$  and  $\kappa$ , write the following approximate relations:

$$(5.6) \quad \beta_0 \approx \psi_0, \quad \beta_1 \approx 2\psi_1 + \psi_4 + \psi_5, \quad \beta_2 \approx \psi_2 + \psi_3, \\ \beta_3 \approx 2\psi_8, \quad \beta_4 \approx 2\psi_{11}.$$

Since in general for additional flows (for any  $\mathbf{N}_2$ )

$$(5.7) \quad \mathbf{G}(s) = \mathbf{C}_2(s) - \mathbf{1} = \left( \mathbf{1} + g_2(s)\mathbf{N}_2^T + \frac{1}{2}g_2^2(s)\mathbf{N}_2^{T^2} + \dots \right) \times \\ \times \left( \mathbf{1} + g_2(s)\mathbf{N}_2 + \frac{1}{2}g_2^2(s)\mathbf{N}_2^2 + \dots \right),$$

we may always use  $\mathbf{G}(s) \approx g_2(s)(\mathbf{N}_2 + \mathbf{N}_2^T)$ , if we are interested in the linearized infinitesimal theory of superposed disturbances.

By way of illustration, we shall present the case of small additional extensions which are realized either in the direction of fundamental extension or in any perpendicular plane.

### 5.1. The case of parallel extensions (in-line oscillations)

For parallel extensional flows, we have in Cartesian coordinates

$$(5.1.1) \quad [\mathbf{N}_1] = \begin{bmatrix} -\frac{1}{2} & 0 & 0 \\ 0 & -\frac{1}{2} & 0 \\ 0 & 0 & 1 \end{bmatrix} q, \quad [\mathbf{N}_2] = \begin{bmatrix} -\frac{1}{2} & 0 & 0 \\ 0 & -\frac{1}{2} & 0 \\ 0 & 0 & 1 \end{bmatrix} \alpha, \quad \mathbf{N}_1\mathbf{N}_2 = \mathbf{N}_2\mathbf{N}_1.$$

The extra stress components are as follows:

$$(5.1.2) \quad T_E^{11} = T_E^{22} = \frac{1}{2}\alpha_1(q) + \frac{1}{4}\alpha_2(q) \\ + \int_0^\infty \left( -\beta_0 + \beta_1 q - \frac{1}{2}\beta_2 q^2 - \frac{3}{4}\beta_3 q^2 - \frac{3}{4}\beta_4 q^2 \right) \alpha g_2(s) ds, \\ T_E^{33} = \alpha_1(q) + \alpha_2(q) + \int_0^\infty \left( 2\beta_0 + 4\beta_1 q + 4\beta_2 q^2 + \frac{3}{2}\beta_4 q^2 \right) \alpha g_2(s) ds.$$

If we assume that  $T^{22} = T^{11} = 0$ , we finally obtain

$$(5.1.3) \quad T^{33} = \frac{3}{2}\alpha_1(q) + \frac{3}{4}\alpha_2(q) \\ + \int_0^\infty \left( 3\beta_0 + 3\beta_1 q + \frac{9}{2}\beta_2 q^2 + \frac{9}{2}\beta_3 q^2 + \frac{9}{4}\beta_4 q^2 \right) \alpha g_2(s) ds.$$

Introducing the notion of complex dynamic extensional viscosity for infinitesimal oscillations, namely

$$(5.1.4) \quad \mu_{33}^*(q, \omega) = \frac{i}{\omega} \int_0^{\infty} \left( 3\beta_0 + 3\beta_1 q + \frac{9}{2} \beta_2 q^2 + \frac{9}{2} \beta_3 q^2 + \frac{9}{4} \beta_4 q^2 \right) (1 - e^{-i\omega s}) ds,$$

we have in the limit

$$(5.1.5) \quad \lim_{q \rightarrow 0} \mu_{33}^*(q, \omega) = 3 \lim_{\kappa \rightarrow 0} \mu_{12}^*(\kappa, \omega).$$

The above relation illustrates the property of Trouton viscosity for superposed oscillatory flows.

## 5.2. The case of perpendicular extensions (cross oscillations)

For perpendicular extensional flows, we have in Cartesian coordinates

$$(5.2.1) \quad [\mathbf{N}_1] = \begin{bmatrix} -\frac{1}{2} & 0 & 0 \\ 0 & -\frac{1}{2} & 0 \\ 0 & 0 & 1 \end{bmatrix} q, \quad [\mathbf{N}_2] = \begin{bmatrix} 1 & 0 & 0 \\ 0 & -\frac{1}{2} & 0 \\ 0 & 0 & -\frac{1}{2} \end{bmatrix} \alpha, \quad \mathbf{N}_1 \mathbf{N}_2 = \mathbf{N}_2 \mathbf{N}_1.$$

The extra stress components are as follows:

$$(5.2.2) \quad \begin{aligned} T_E^{11} &= -\frac{1}{2} \alpha_1(q) + \frac{1}{4} \alpha_2(q) + \int_0^{\infty} \left( 2\beta_0 - 2\beta_1 q + \beta_2 q^2 + \frac{3}{4} \beta_3 q^2 + \frac{3}{8} \beta_4 q^2 \right) \alpha g_2(s) ds, \\ T_E^{22} &= -\frac{1}{2} \alpha_1(q) + \frac{1}{4} \alpha_2(q) + \int_0^{\infty} \left( -\beta_0 + \beta_1 q - \frac{1}{2} \beta_2 q^2 + \frac{3}{4} \beta_3 q^2 + \frac{3}{8} \beta_4 q^2 \right) \alpha g_2(s) ds, \\ T_E^{33} &= \alpha_1(q) + \alpha_2(q) + \int_0^{\infty} \left( -\beta_0 - 2\beta_1 q - 2\beta_2 q^2 - \frac{3}{2} \beta_3 q^2 - \frac{3}{4} \beta_4 q^2 \right) \alpha g_2(s) ds. \end{aligned}$$

If we assume that additional stresses  $\Delta T^{33} = 0$ , we obtain

$$(5.2.3) \quad \Delta T^{11} = \int_0^{\infty} \left( 3\beta_0 + 3\beta_2 q^2 + \frac{9}{4} \beta_3 q^2 + \frac{9}{8} \beta_4 q^2 \right) \alpha g_2(s) ds.$$

On the other hand, if we assume that  $\Delta T^{22} = 0$ , we have

$$(5.2.4) \quad \begin{aligned} \Delta T^{11} &= \int_0^{\infty} \left( 3\beta_0 - 3\beta_1 q + \frac{3}{2} \beta_2 q^2 \right) \alpha g_2(s) ds, \\ \Delta T^{33} &= \int_0^{\infty} \left( -3\beta_1 q - \frac{3}{2} \beta_2 q^2 - \frac{9}{4} \beta_3 q^2 - \frac{9}{8} \beta_4 q^2 \right) \alpha g_2(s) ds. \end{aligned}$$

Calculating the complex dynamic extensional viscosities  $\eta_{11}^*$  and  $\eta_{33}^*$ , after taking into account (4.2.3), (4.2.4), we obtain

$$(5.2.5) \quad \begin{aligned} \lim_{q \rightarrow 0} \eta_{11}^*(q, \omega) &= 3 \lim_{\kappa \rightarrow 0} \eta_{23}^*(\kappa, \omega), \\ \lim_{q \rightarrow 0} \eta_{33}^*(q, \omega) &= \lim_{\kappa \rightarrow 0} \eta_{13}^*(\kappa, \omega) = 0. \end{aligned}$$

As can be seen from the above examples, information obtained on the basis of superposed shearing flows is, in general, not sufficient to predict the fluid behaviour in superposed extensional flows. We hope that some other flows realizable in new rheometers (cf. [18]) might be more useful in further investigations.

## 6. Certain rheological relations

Recently BERNSTEIN and FOSDICK [13], for the case of in-line shearing oscillations, and TANNER and WILLIAMS [9], for the case of cross shearing oscillations, derived certain rheological relations satisfied for any incompressible BKZ fluid [19]. Some relations which are also valid for an arbitrary incompressible simple fluid have been presented in Sec. 4.1 and 4.2.

Now, let us mention all of them, paying particular attention to the implications which they cause for simple fluids.

BERNSTEIN and FOSDICK proved for in-line oscillations that

$$(6.1) \quad \lim_{\omega \rightarrow 0} \eta'_{12}(\kappa, \omega) = \frac{d}{d\kappa} \tau(\kappa),$$

$$(6.2) \quad \lim_{\omega \rightarrow 0} \frac{2G'_{12}(\kappa, \omega)}{\omega^2} = \frac{d}{d\kappa} \left( \frac{\sigma_1(\kappa) - \sigma_2(\kappa)}{\kappa} \right),$$

$$(6.3) \quad \lim_{\omega \rightarrow 0} \frac{H''(\kappa, \omega)}{\omega} = \frac{d}{d\kappa} (\sigma_1(\kappa) - \sigma_2(\kappa)),$$

$$(6.4) \quad \lim_{\omega \rightarrow \infty} H'(\kappa, \omega) = \frac{d}{d\kappa} (\kappa \tau(\kappa)),$$

where the notations used in Sec. 4.1, 4.2 were applied.

The relations (6.1) and (6.3) have been derived in Sec. 4.1 for an incompressible simple fluid, while (6.2) and (6.4) are valid only for a BKZ fluid. Since any BKZ fluid is a special case of the general simple fluid, the latter relations determine certain additional conditions imposed on the functions  $\psi_i$ . If (6.2) and (6.4) are satisfied, then

$$(6.5) \quad - \int_0^{\infty} (\psi_0 + \kappa^2 \psi_2 + \kappa^2 \psi_3 + \kappa^2 \psi_7 + \kappa^2 \psi_8) s^2 ds = \frac{d}{d\kappa} \left( \frac{\sigma_1(\kappa) - \sigma_2(\kappa)}{\kappa} \right),$$

$$(6.6) \quad - \int_0^{\infty} (2\kappa \psi_4 - 2\kappa \psi_5 - 2\kappa^3 \psi_9) ds = \kappa \frac{d\tau(\kappa)}{d\kappa} + \tau(\kappa).$$

TANNER and WILLIAMS [9] proved for cross oscillations that

$$(6.7) \quad \lim_{\omega \rightarrow 0} \eta'_{23}(\kappa, \omega) = \frac{\tau(\kappa)}{\kappa} \equiv \eta(\kappa),$$

$$(6.8) \quad \lim_{\omega \rightarrow 0} \frac{G'_{23}(\alpha, \omega)}{\omega^2} = \frac{\sigma_1(\kappa) - \sigma_2(\kappa)}{2\kappa^2}.$$

The relation (6.7) has been derived in Sec. 4.2, while (6.8) implies that

$$(6.9) \quad - \int_0^{\infty} (\psi_0 + \kappa^2 \psi_2 + \kappa^2 \psi_3) s^2 ds = \frac{\sigma_1(\kappa) - \sigma_2(\kappa)}{\kappa^2}.$$

As the consequence of (6.5) and (6.9), we also obtain

$$(6.10) \quad - \int_0^{\infty} \kappa^2 (\psi_7 + \psi_8) s^2 ds = \frac{1}{\kappa} \frac{d}{d\kappa} (\sigma_1 \kappa - \sigma_2 \kappa) - 2 \frac{\sigma_2(\kappa) - \sigma_1(\kappa)}{\kappa^2}.$$

More recently, Bernstein [14] proved for superposed shearing oscillations that

$$(6.11) \quad G_{in}^* = G_{tr}^* + \kappa \frac{\partial G_{tr}^*}{\partial \kappa},$$

where the subscripts in and tr mean in-line and transverse oscillations, respectively. The similar relation can be written in terms of complex viscosities, namely

$$(6.12) \quad \mu_{12}^* = \eta_{23}^* + \kappa \frac{\partial}{\partial \kappa} \eta_{23}^*.$$

It results from (4.1.6) and (4.2.7) that always

$$(6.13) \quad \frac{d\tau}{d\kappa} = \frac{\tau}{\kappa} - \int_0^{\infty} \kappa^2 (\psi_7 + \psi_8) s ds;$$

then (6.12) implies that

$$(6.14) \quad \kappa \frac{d}{d\kappa} \left( \frac{\tau}{\kappa} \right) = - \int_0^{\infty} \kappa^2 (\psi_7 + \psi_8) s ds.$$

This condition will be satisfied if

$$(6.15) \quad \frac{\partial}{\partial \kappa} (\psi_0 + \kappa^2 \psi_2 + \kappa^2 \psi_3) = \kappa (\psi_7 + \psi_8).$$

To conclude this section, let us remark that many of the rheological relations might be verified experimentally. Such an attempt has been presented in [13], although the available experimental data do not seem to be sufficient. In any case, future experiments should be planned and conducted in close connection with theoretical predictions.

## 7. Conclusions

Certain conclusions based on the present considerations can be listed as follows:

1. The behaviour of incompressible simple fluids in superposed shearing flows is entirely described by three viscometric functions characteristic for steady flows, and twelve material functions responsible for additional small disturbances. These material functions enter the constitutive equations in certain combinations.

2. The dynamic viscosities for in-line and cross shearing oscillations are, in general, different. In the case of small angular velocities, the dynamic viscosity for in-line oscillations is less than the dynamic viscosity for cross oscillations.

3. The existing differences in material characteristics of incompressible simple fluids under parallel and perpendicular shearings may, in principle, be verified experimentally. In the case of modified Couette apparatus with concentric cylinders, the torsional oscillations correspond to in-line shearings, while the axial oscillations provide example of cross shearings.

4. The constitutive equations derived may be used to describe the fluid behaviour in other flows with superposed proportional stretch histories, in particular, for superposed extensions. In this latter case, the differences in corresponding dynamic extensional viscosities are observed.

5. The material functions valid for superposed shearing flows and extensional flows are, in general, different. There are no general relations between them. Certain particular relations may be established for other superposed proportional flows — e.g., flows realized in new rheometers with eccentric plates or cylinders, tilted plates, etc. (cf. [18, 20]).

6. Because of the consistency relations (cf. [2, 3]), there exist certain conditions connecting the dynamic and steady characteristics of an incompressible simple fluid. These relations are usually established either for small frequencies or slow fundamental flows.

7. We cannot accept the statement that the rheological relations derived for BKZ fluids are not satisfied for general simple fluids (cf. [6, 13]). These rheological equations imply certain conditions imposed on the material functions. Usually quoted examples prove only that the rheological equations cannot be satisfied by particular second-order simple fluids.

8. Various experimental investigations of dynamic properties of viscoelastic fluids, such as polymer melts and solutions, seem to be necessary for verification of the existing formulae as well as for deeper understanding of material properties (cf. [21]).

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