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# *A Phenomenological Theory of Streaming Birefringence*

BERNARD D. COLEMAN, ELLIS H. DILL, & RICHARD A. TOUPIN

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## 1. Preface

Every student of physics knows that rays of light generally change direction on passing from one transparent substance into another and that the phenomenon is called *refraction*. It is also well known that for water at rest, or glass in an undistorted state, a single incident ray gives rise to only one ray upon refraction. When a ray of light enters a crystal, however, it generally is split into two waves which propagate with different speeds and are polarized at right angles to each other. Substances which split light in this manner are said to be *doubly refracting* or

*birefringent*. Many materials which are not birefringent in undistorted states become birefringent upon deformation; this phenomenon is called *induced birefringence*.\*

Although a fluid which has been at rest for a reasonable length of time can be expected to show no birefringence, many fluids exhibit birefringence when flowing, even at low rates of shear. It appears that such *streaming birefringence* of fluids was first observed by MAXWELL [1873, 1] and MACH [1872, 1]. The former described an experimental method used to this day:

“In 1866 I made some attempts to ascertain whether the state of strain in a viscous fluid in motion could be detected by its action on polarized light. I had a cylindrical box with a glass bottom. Within this box a solid cylinder could be made to rotate. The fluid to be examined was placed in the annular space between this cylinder and the sides of the box. Polarized light was thrown up through the fluid parallel to the axis, and the inner cylinder was then made to rotate. I was unable to obtain any result with a solution of gum or sirup of sugar, though I observed an effect on polarized light when I compressed some Canada balsam which had become very thick and almost solid in a bottle.”

Our goal here is to develop a general phenomenological theory of streaming birefringence. The theory we give has a mathematical structure closely related to that of the dynamical theory of “simple fluids with fading memory”. In that theory, a material is characterized by a functional  $\mathcal{S}$  which gives the stress when the history  $\mathbf{J}^t$  of the relative strain is specified. Here, in addition to  $\mathcal{S}$  we have, for each material, a functional  $\mathcal{N}$  which relates the dielectric properties of the material to  $\mathbf{J}^t$ .

## 2. Fresnel's Theory of Double Refraction

A successful description of the propagation of light in birefringent bodies was achieved by FRESNEL in 1827. \*\* To introduce the reader to the language used by the experimenters who make quantitative studies of birefringence, in this chapter we attempt a brief outline of FRESNEL'S theory. In the theory it is assumed that light propagates as transverse harmonic waves. One associates with each point  $\mathbf{x}$  of the material a symmetric, positive definite, tensor  $\mathbf{L}$ , which we may call the *Fresnel tensor*. If one chooses a Cartesian coordinate system  $(\xi, \eta, \zeta)$  with its origin at  $\mathbf{x}$  and axes along the proper vectors  $\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3$  of  $\mathbf{L}$ , then the equation,

$$v_1^2 \xi^2 + v_2^2 \eta^2 + v_3^2 \zeta^2 = 1, \quad (2.1)$$

with  $v_i^2$  the proper number of  $\mathbf{L}$  corresponding to  $\mathbf{k}_i$ , describes a figure called the *Fresnel ellipsoid*. The positive numbers  $v_1, v_2, v_3$  are called the *principal wave speeds*, and the vectors  $\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3$  are called the *principal axes of refraction*. The tensor  $\mathbf{L}$ , and hence the quadric (2.1), depend, in general, on the material and its

---

\* Early rational theories of induced birefringence in elastic solids are those of NEUMANN [1841, 1] and MAXWELL [1853, 1]; apparently the phenomenon was first observed in solids by BREWSTER [1816, 1].

\*\* [1827, 1]. A history of the subject is given by WHITTAKER [1951, 1].

deformation history. When a harmonic light wave passes through the point  $x$ , the plane normal to the direction of propagation of the wave (*i.e.* the “wave front”) intersects the Fresnel ellipsoid in an ellipse  $E$ . Fresnel’s theory requires that the amplitude vector of the wave be directed along one of the axes of  $E$ . There are thus, in general, two harmonic waves possible for a given direction of propagation; each has its amplitude vector directed along one axis of  $E$ . The lengths of the semi-axes of  $E$  are the reciprocals of the speeds of these two harmonic waves.

Consider now a single, fixed, Cartesian coordinate system  $(x, y, z)$ , and suppose that the body  $\mathcal{B}$  under consideration has the form of a strip bounded by the parallel planes  $z = +l/2$ ,  $z = -l/2$ . We permit the Fresnel tensor  $L$  to vary with  $x$  and  $y$  in  $\mathcal{B}$  but assume that  $L$  is independent of  $z$  and is everywhere such that it has a proper vector,  $k_3$ , parallel to the  $z$ -axis. When a planar harmonic wave propagating along the  $z$ -axis enters  $\mathcal{B}$ , it is, according to Fresnel’s theory, split into two harmonic waves with mutually perpendicular amplitude vectors. These two waves continue to propagate in the  $z$ -direction, but with different speeds  $v_1, v_2$ . Each wave has its amplitude parallel to a proper vector of  $L$ , and the speed of the wave equals the square root of the corresponding proper number of  $L$ . The numbers

$$n_i = \frac{c}{v_i}, \quad i = 1, 2 \quad (2.2)$$

are called *principal indices of refraction*; here  $c$  is the velocity of light *in vacuo*. During its traverse of  $\mathcal{B}$ , each wave experiences an *absolute retardation*  $r_i$ , given by

$$r_i = l(n_i - 1), \quad i = 1, 2. \quad (2.3)$$

The relative retardation of waves when they leave  $\mathcal{B}$  is

$$r = r_1 - r_2 = l(n_1 - n_2). \quad (2.4)$$

We have here assumed that  $\mathcal{B}$  is such that one principal axis of refraction,  $k_3$ , is everywhere parallel to the  $z$ -axis of the system  $(x, y, z)$ . One can determine  $r$  and the direction of the two remaining principal axes,  $k_1, k_2$ , by using a plane polariscope, *i.e.* by placing  $\mathcal{B}$  between two crossed\* polarizing devices which can be rotated in planes parallel to the  $(x, y)$ -plane in such a way that the polarizing axes of the devices remain at right angles; the first polarizing device, *i.e.* that between  $\mathcal{B}$  and the light source, is called the *polarizer*, and the second polarizing device is called the *analyzer*. Whenever the polarizer has its polarizing axis parallel to  $k_1$  or  $k_2$ , each light wave transmitted by the polarizer passes through  $\mathcal{B}$  with no change in the direction of its amplitude vector; such a light wave is blocked by the analyzer. If  $k_1$  and  $k_2$  vary from point to point in the  $(x, y)$ -plane, then the locus of the points in this plane for which either  $k_1$  or  $k_2$  is parallel to an axis of the polarizer appears as a dark figure when viewed through the analyzer. This dark figure, which depends on the orientation of the polarizer, usually takes the form

\* Other arrangements of the polarizing devices can be used; that which we describe appears to be the most popular.

of a pair of curved lines, called *isoclinic lines* or *isoclines*. By rotating the polarizer and analyzer in unison, a family of such lines, called the *isoclinic fringe pattern*, is obtained. Observation of this pattern enables one to determine the axes of refraction for  $\mathcal{B}$  as functions of  $x$  and  $y$ . Whenever the axis of the polarizer is not parallel to a principal axis of refraction, each of the two harmonic waves transmitted by  $\mathcal{B}$  is resolved by the analyzer into two waves: a component parallel and a component perpendicular to the polarizing axis. Of course, only components along the axis of the analyzer pass through it. Thus, the analyzer transmits two waves with colinear, but oppositely directed amplitude vectors of equal magnitude; these two transmitted waves will interfere (*i.e.* cancel) whenever the relative retardation  $r$  obeys the formula

$$r = N\lambda, \quad (2.5)$$

with  $N$  an integer and  $\lambda$  the wavelength of the light *in vacuo*. For each integer  $N$ , the locus of  $(x, y)$ -values with  $r = N\lambda$  forms a figure which appears dark when viewed through the analyzer; this dark figure, which often is a curved line or set of curved lines, is referred to as the  $N^{\text{th}}$  *isochromatic line* or *isochromatic fringe*. The family of all isochromatic fringes (*i.e.*  $N = 0, \pm 1, \dots$ ) is called the *isochromatic fringe pattern*. Through observations of the isochromatic fringe pattern, the relative retardation  $r$  can be determined at several values of  $x$  and  $y$ . It is clear from equation (2.4) that if the thickness  $l$  of  $\mathcal{B}$  is known, then measurement of  $r$  yields the difference  $\Delta$  between the indices of refraction  $n_1, n_2$ :

$$\Delta \stackrel{\text{def}}{=} n_1 - n_2 = \frac{r}{l}. \quad (2.6)$$

The number  $\Delta$  is called the *birefringence* of  $\mathcal{B}$  (at  $x$  and  $y$ ) for propagation in the direction  $\mathbf{k}_3$ .

The above description of the properties of isoclinic and isochromatic lines is summarized and extended in a single formula which, for given values of  $\lambda$  and  $l$ , describes the variation of the intensity  $I$  of the light transmitted by the analyzer as a function of the birefringence  $\Delta$  and the angle  $\phi$  between  $\mathbf{k}_1$  and the axis of the polarizer: \*

$$I = A \sin^2(2\phi) \sin^2\left(\frac{\pi l}{\lambda} \Delta\right). \quad (2.7)$$

Here  $A$  is a constant which depends upon the loss of light through reflection, absorption, and scattering. It is clear from (2.7) that  $I$  vanishes wherever  $\phi$  equals  $0^\circ$  or  $90^\circ$  and wherever  $l\Delta/\lambda$  is an integer; *i.e.* on the isoclinic and the isochromatic lines. Under appropriate circumstances one can use (2.7) to determine the birefringence from intensity measurements at  $(x, y)$ -values which are not on an isochromatic fringe. An often simpler method of obtaining  $\Delta$  away from the isochromatic fringe pattern involves the insertion of a compensator between  $\mathcal{B}$  and the analyzer. The compensator, when properly aligned with respect to  $\mathbf{k}_1$  and  $\mathbf{k}_2$ ,

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\* Cf. *e.g.* BORN & WOLFE [1959, 1] § 14.4.3.

gives an additional and controllable relative retardation  $R$  to the light reaching the analyzer; one adjusts the compensator so that the total relative retardation,  $r + R$ , equals an integral multiple of  $\lambda$ , and then calculates  $\Delta$ , using (2.6).

The indices of refraction  $n_1$  and  $n_2$ , at given values of  $x$  and  $y$ , can be measured with an interferometer. As usually employed, such a device splits the incident wave into two waves; one is polarized along a principal axis of refraction  $k_i$  for  $\mathcal{B}$  and is passed through  $\mathcal{B}$  to incur a retardation  $r_i$ . The second wave is directed through a medium which induces a controllable retardation. The two waves are then recombined and are found to interfere whenever the difference in their retardations is an integral multiple of  $\lambda$ . Thus, if  $\lambda$  is known,  $r_1$  and  $r_2$  can be determined. Once  $r_1, r_2$ , and the thickness  $l$  of  $\mathcal{B}$  are measured, the indices of refraction  $n_1$  and  $n_2$  may be calculated from (2.3).

### 3. The Classical Theory of Streaming Birefringence

It has long been known that viscous fluids can exhibit birefringence when flowing.\* It follows from results we shall present in Section 11 that, for an incompressible simple fluid with fading memory,\*\* in the limit of slow flow, each proper vector  $d_i$  of the rate of deformation tensor  $D$ \*\*\* is also a principal axis of refraction  $k_i$ , and the principal indices of refraction  $n_i$  are determined by the proper numbers  $d_i$  of  $D$  through equations of the form

$$n_i = n^\circ + 2\eta_N d_i, \quad i = 1, 2, 3, \quad (3.1)$$

with  $n^\circ$  and  $\eta_N$  material constants. This yields the relation

$$\Delta = 2\eta_N(d_1 - d_2) \quad (3.2)$$

for the birefringence  $\Delta = n_1 - n_2$  for wave propagation along  $k_3$ .

In quantitative experimental work on streaming birefringence, one usually follows MAXWELL [1873, 1] and uses Couette flow. In this circular flow of a fluid confined between coaxial cylinders, at each point  $x$ , one proper vector of  $D$ , say  $d_3$ , is parallel to the common axis of the cylinders, and the remaining two form angles of  $45^\circ$  with the radial vector to  $x$ . One studies this flow with a polariscope so arranged that the polarized light propagates in the direction  $d_3$ . If the gap between the bounding cylinders is small, then each isocline is a straight line lying along a radius vector.\*\*\*\* The clockwise angle  $\chi$  from the axis of the polarizer to the isoclinic line nearest to it is called the *extinction angle*. If the proper vectors of  $D$  are also axes of refraction, as in the "slow flow" approximation behind equation (3.1), then  $|\chi| = 45^\circ$ . Steady flow experiments on many "non-Newtonian

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\* As we indicated in the Preface, MAXWELL [1873, 1] observed the effect in Canada balsam in 1866. MACH [1872, 1] independently observed it in molten glass.

\*\* The concepts of "fading memory" and "slow flow" employed here are those used by COLEMAN & NOLL [1960, 1] [1962, 2].

\*\*\*  $D$ , also called the *stretching tensor*, is equal to the symmetric part of the velocity gradient.

\*\*\*\* For more details see Section 8, particularly Figure 8.1.

fluids” show that  $|\chi|$  is not, in general, equal to  $45^\circ$ , but approaches this value as the rate of shear tends to zero.\*

In the same limit of slow motion for which (3.1) holds, the stress in a general incompressible simple fluid with fading memory obeys constitutive relations of the type employed to define Navier-Stokes fluids; \*\* *i.e.* the principal axes of stress equal the proper vectors of  $\mathbf{D}$ , and for the principal stresses  $\sigma_i$  there hold the equations

$$\sigma_i = -p + 2\eta_S d_i, \quad i = 1, 2, 3, \quad (3.3)$$

where  $p$  is a hydrostatic pressure, and  $\eta_S$  is a positive material constant called the *viscosity*. It follows that, in the limit of slow motions, each principal axis of stress is also a principal axis of refraction, and the birefringence  $n_1 - n_2$  is given by a relation of the form

$$\Delta = \beta(\sigma_1 - \sigma_2), \quad (3.4)$$

with  $\beta = \eta_N/\eta_S$ .

There is no reason to believe that the “classical” formulae (3.1) and (3.3), or their corollary (3.4), hold for arbitrary fluids at arbitrary rates of shear. Furthermore, although it is true that in the limit of slow flow the principal axes of refraction should lie along the principal axes of stress, there is no theoretical principle or reliable experimental evidence indicating that such an elementary rule holds in general.\*\*\*

We here seek a simple theory of birefringence in fluids consistent with the general principles of mechanics and electromagnetism and broad enough to permit a rational discussion of the streaming birefringence of “non-Newtonian fluids” away from “slow-flow limits”. Hence we must drop the linear approximations made in classical theories of mechanics and must allow the past kinematical history to influence present optical and mechanical behavior. However, any theory which accounts for past deformations of the medium and non-vanishing rates of shear is beset with difficulties that arise from the fact that *motion* must be discussed. Of course, we shall here neglect relativistic effects of order  $u^2/c^2$ , where  $u$  is the speed of the material points and  $c$  the speed of light *in vacuo*. Yet even in this approximation, the Maxwell-Lorentz equations for the electromagnetic field in moving media are more complicated than for the familiar case of stationary media. A simple theory of birefringence emerges only if one can safely neglect the dragging of light by the medium. Fortunately, for the fluid speeds ordinarily encountered in viscoelastic flows, such dragging is truly negligible.

#### 4. Concepts from Electromagnetic Theory

Fresnel’s theory of double refraction in transparent media can be shown to rest upon the foundation of Maxwell’s electromagnetic theory of light.\*\*\*\* As we

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\* It appears that KUNDT [1881, 1] was the first to report departures of  $|\chi|$  from  $45^\circ$ ; solutions of colloid in ether were among the substances for which he observed large departures. It most likely was DE METZ [1888, 1] [1906, 1] who made the first quantitative measurements of the birefringence  $n_1 - n_2$  in Couette flow.

\*\* Cf. [1960, 1].

\*\*\* COLEMAN & TOUPIN [1962, 3]; see Sections 7 and 11 of this essay.

\*\*\*\* MAXWELL [1865, 1] §§ 102–105, [1873, 2]; see also BORN & WOLF [1959, 1] and TRUESDELL & TOUPIN [1960, 2].

are here concerned with non-magnetic, non-conducting, electrically polarizable media, we may write the basic field equations of electromagnetic theory in the form

$$\begin{aligned}\frac{\partial \mathbf{d}}{\partial t} &= \text{curl } \mathbf{h}, & \text{div } \mathbf{d} &= 0, \\ \frac{\partial \mathbf{b}}{\partial t} &= -\text{curl } \mathbf{e}, & \text{div } \mathbf{b} &= 0,\end{aligned}\tag{4.1}$$

with

$$\mathbf{d} = \epsilon_0 \mathbf{e} + \mathbf{p}, \quad \mathbf{h} = \mu_0^{-1} \mathbf{b} + \mathbf{u} \times \mathbf{p}.\tag{4.2}$$

Here  $\mathbf{e}$  is the *electric field*,  $\mathbf{b}$  is the *magnetic flux density*,  $\mathbf{p}$  is the *polarization density*, and  $\mathbf{u}$  is the *velocity* of the medium. The two vectors  $\mathbf{d}$  and  $\mathbf{h}$  are called, respectively, the *electric displacement* and the *magnetic field*. The positive numbers  $\epsilon_0$  and  $\mu_0$  are fundamental constants which depend upon only the choice of units and obey the relation  $\mu_0 \epsilon_0 = c^{-2}$ , with  $c$  the speed of light *in vacuo*. If  $\mathbf{h}$ ,  $\mathbf{d}$ ,  $\mathbf{e}$ , or  $\mathbf{b}$  experiences a saltus,  $[\mathbf{h}]$ ,  $[\mathbf{d}]$ , *etc.*, across a surface with unit normal  $\mathbf{v}$ , then the following *jump conditions* must hold:

$$\begin{aligned}\mathbf{v} \times [\mathbf{h}] &= \mathbf{0}, & \mathbf{v} \cdot [\mathbf{d}] &= 0, \\ \mathbf{v} \times [\mathbf{e}] &= \mathbf{0}, & \mathbf{v} \cdot [\mathbf{b}] &= 0.\end{aligned}\tag{4.3}$$

The simplest electromagnetic theory of light in moving media is based upon the constitutive equation\*

$$\mathbf{p} = \epsilon_0 \mathbf{Z}(\mathbf{e} + \mathbf{u} \times \mathbf{b}), \quad \text{with } \mathbf{Z} = \mathbf{K} - \mathbf{1};\tag{4.4}$$

here  $\mathbf{1}$  is the unit tensor, and  $\mathbf{K}$  is a symmetric, positive definite, linear transformation, called the *dielectric tensor* and assumed to be independent of  $\mathbf{e}$ ,  $\mathbf{b}$ , and  $\mathbf{u}$ . (We shall discuss later the way  $\mathbf{K}$  is related to the history of the deformation.) On substituting (4.4) into (4.2), one finds that

$$\begin{aligned}\mu_0 \mathbf{h} &= \mathbf{b} + \underline{\underline{c^{-2} \mathbf{u} \times \mathbf{Z}(\mathbf{u} \times \mathbf{b})}} + \underline{c^{-2} \mathbf{u} \times \mathbf{Z} \mathbf{e}} \\ \mathbf{d} &= \epsilon_0 \mathbf{K} \mathbf{e} + \underline{\epsilon_0 \mathbf{Z}(\mathbf{u} \times \mathbf{b})}.\end{aligned}\tag{4.5}$$

The doubly underlined term here is  $O(u^2/c^2)$ , and may be safely neglected when the speed of the medium is small compared with the velocity of light. The singly underlined terms give rise to the dragging of light by the medium.\*\* It is hoped that for the analysis of light waves in slowly moving media, not only the doubly, but also the singly underlined terms can be neglected. In the present essay we make such an approximation and take

$$\begin{aligned}\mathbf{h} &= \mu_0^{-1} \mathbf{b}, \\ \mathbf{d} &= \epsilon_0 \mathbf{K} \mathbf{e},\end{aligned}\tag{4.6}$$

as our starting *electromagnetic constitutive equations*.

\* Cf. TRUESDELL & TOUPIN [1960, 2] p. 737.

\*\* Cf. TRUESDELL & TOUPIN [1960, 2] p. 740.



From a more general point of view, our starting assumptions (4.6) involve approximations beyond the neglect of relativistic effects and the dragging of light. Since we have assumed  $\mathbf{K}$  to be independent of  $\mathbf{e}$  and  $\mathbf{b}$  (and their past histories) and have implicitly set the electric current and the magnetization equal to zero, application of our theory should be restricted to weak fields (*i.e.* to light waves of "normal" intensity) and to media which neither absorb strongly nor rotate light. Materials obeying constitutive relations of the form (4.6) are called *perfect dielectrics*.

If the dielectric tensor  $\mathbf{K}$  is constant in space and time, then Fresnel's construction, discussed in Section 2, gives a correct description of a single planar harmonic electromagnetic wave governed by the equations (4.1) and (4.6), provided one identifies the Fresnel tensor  $\mathbf{L}$  with  $c^2 \mathbf{K}^{-1}$ :

$$\mathbf{L} = c^2 \mathbf{K}^{-1}, \quad \mathbf{K} = c^2 \mathbf{L}^{-1}. \quad (4.7)$$

That is, the principal axes of refraction  $\mathbf{k}_i$  are the proper vectors of  $\mathbf{K}$ , and the principal wave speeds  $v_i$  are related as follows to the proper numbers  $\kappa_i$  of  $\mathbf{K}$ :

$$v_i^2 = c^2 \kappa_i^{-1}, \quad i = 1, 2, 3. \quad (4.8)$$

By (4.8) and (2.2), each principal index of refraction  $n_i$  obeys the simple formula

$$n_i^2 = \kappa_i, \quad i = 1, 2, 3. \quad (4.9)$$

The problem of describing a plane electromagnetic wave propagating in the direction  $\mathbf{k}_3$  through the strip  $\mathcal{B}$  of Section 2 may be solved by applying the jump conditions (4.3) to the incident, reflected, and transmitted waves at both surfaces of  $\mathcal{B}$ . The exact solution is complicated.\* If, however, the material is such that there is but a small loss of light by reflection at the entering and exiting surfaces, then the conclusions of Section 2 give a very good approximation to the exact solution. Thus, under the conditions expected in applications, a planar polarized wave incident normal to  $\mathcal{B}$  is resolved into two waves which have mutually perpendicular amplitude vectors along  $\mathbf{k}_1$  and  $\mathbf{k}_2$  and which, during their traverse of  $\mathcal{B}$ , experience a relative retardation  $r$  given by (2.4).

The optical properties of a perfect dielectric are completely determined when its dielectric tensor  $\mathbf{K}$  is specified. However, any invertible function of  $\mathbf{K}$ , such as the Fresnel tensor  $\mathbf{L}$ , will do equally well. It appears to us that the theory of induced birefringence takes its simplest form if one works with the *refraction tensor*  $\mathbf{N}$ , defined as the positive definite square root of  $\mathbf{K}$ :

$$\mathbf{N} \stackrel{\text{def}}{=} \mathbf{K}^{\frac{1}{2}}. \quad (4.10)$$

By (4.7) and (4.9), the proper vectors of  $\mathbf{N}$  are the principal axes of refraction  $\mathbf{k}_i$ , while the proper numbers of  $\mathbf{N}$  are the principal indices of refraction  $n_i$ . Of course,  $\mathbf{N}$  determines  $\mathbf{L}$  and  $\mathbf{K}$  through the relations

$$\mathbf{L} = c^2 \mathbf{N}^{-2}, \quad \mathbf{K} = \mathbf{N}^2. \quad (4.11)$$

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\* See BORN & WOLF [1959, 1] § 7.6.1, and DILL & FOWLKES [1966, 3].

## 5. Incompressible Fluids

Consistent with our linear treatment of the electromagnetic field, we suppose that the field is so weak that it has no effect on the motion of the material. We assume that Cauchy's stress principle is valid and that the equation of motion has the classical form

$$\operatorname{div} \mathbf{S} + \rho \mathbf{f} = \rho \ddot{\mathbf{x}}, \quad (5.1)$$

with  $\rho$  the density of mass,  $\mathbf{f}$  the body force per unit mass, and  $\mathbf{S}$  the stress tensor. When it is assumed that the electromagnetic field does not influence the motion, the principle of angular momentum, together with (5.1) and the usual assumptions regarding the absence of distributed moments and couples, implies that Cauchy's stress tensor is symmetric:

$$\mathbf{S} = \mathbf{S}^T. \quad (5.2)$$

We here take (5.2) as a postulate.

Consider now a material point  $P$  of a body  $\mathcal{B}$  that is in motion. Let  $\mathbf{x}$  be the spatial position of  $P$  at time  $t$ , and let  $\xi_t(\mathbf{x}, \tau)$  be the spatial position of  $P$  at time  $\tau$ . The function  $\xi_t$  is called the *relative deformation function*. The tensor  $F_t(\tau)$ , defined by

$$F_t(\tau) = \operatorname{grad}_{\mathbf{x}} \xi_t(\mathbf{x}, \tau), \quad (5.3)$$

is the deformation gradient at  $P$  at time  $\tau$  computed employing the configuration at time  $t$  as reference. Thus  $F_t(\tau)$  describes the change of local configuration at  $P$  between times  $t$  and  $\tau$ . Since  $\mathbf{x} = \xi_t(\mathbf{x}, t)$ ,

$$F_t(t) = \mathbf{1}. \quad (5.4)$$

The tensor function  $F_t^s$  defined by

$$F_t^s(s) = F_t(t-s), \quad s \in [0, \infty), \quad (5.5)$$

is called the *history up to  $t$  of the relative deformation gradient at  $P$* . It follows from (5.4) that

$$F_t^s(0) = \mathbf{1}. \quad (5.6)$$

A motion is isochoric if and only if for each material point and time

$$|\det F_t^s(s)| = 1, \quad (5.7)$$

for all  $s \geq 0$ .

In our present theory an *incompressible simple fluid* is a material which obeys three constitutive equations whose content may be stated in words as follows:

- (I) Only isochoric motions are permitted.
- (II) The refraction tensor  $N$  in (4.10) is determined by the history  $F_t^s$  of the relative deformation gradient.
- (III)  $F_t^s$  determines the stress tensor  $\mathbf{S}$  to within an arbitrary hydrostatic pressure.

The first of these constitutive equations is (5.7); the second and third are

$$N(t) = \mathfrak{N}(F_t^i), \quad (5.8)$$

$$S_D(t) = \mathfrak{S}(F_t^i). \quad (5.9)^*$$

Here  $S_D$  is the deviator of the stress tensor  $S$ , *i.e.*

$$S_D \stackrel{\text{def}}{=} S - \frac{1}{3} \mathbf{1} \operatorname{tr} S, \quad (5.10)$$

while  $\mathfrak{N}$  and  $\mathfrak{S}$  are constitutive functionals which characterize the fluid under consideration.

It follows from (5.8) and (4.10) that the electromagnetic constitutive equation (4.6)<sub>2</sub> can be written

$$d(t) = \varepsilon_0 \mathfrak{N}(F_t^i)^2 e(t). \quad (5.11)$$

Throughout this essay we shall assume that the fluid body  $\mathcal{B}$  under consideration is *materially homogeneous* in the sense that the functionals  $\mathfrak{N}$  and  $\mathfrak{S}$  do not vary from point to point in  $\mathcal{B}$ ; of course, in a given motion the arguments  $F_t^i$  and the values  $N(t)$  and  $S_D(t)$  of  $\mathfrak{N}$  and  $\mathfrak{S}$  will depend not only on time but also on location.

## 6. Frame-Indifference

In classical mechanics a *change of frame* is a one-parameter family of transformations of space onto itself, with time the parameter, such that at each instant the mapping  $x \rightarrow x^*$  preserves distances. \*\* It can be proved that each change of frame has the form

$$x^* = c(t) + Q(t)[x - q] \quad (6.1)$$

where, for each time  $t$ ,  $c(t)$  is a point in space and  $Q(t)$  is an orthogonal tensor;  $q$  is a point in space which can be taken independent of  $t$ . It follows that the unit normal to a surface is transformed into

$$n^* = Q(t)n, \quad (6.2)$$

and  $F_t(\tau)$  in (5.3) is carried into \*\*\*

$$F_t(\tau)^* = Q(\tau)F_t(\tau)Q(t)^{-1}. \quad (6.3)$$

Putting

$$R(s) = Q(t-s), \quad s \in [0, \infty), \quad (6.4)$$

we conclude from (6.3) that the history  $F_t^i$  defined in (5.5) is transformed by the change of frame into a new history  $F_t^{i*}$  given by

$$F_t^i(s)^* = R(s)F_t^i(s)R(0)^{-1}, \quad s \in [0, \infty). \quad (6.5)$$

---

\* The constitutive equation (5.9) was proposed and studied by NOLL [1958, 1]; see also COLEMAN & NOLL [1959, 2] [1961, 1] and the expositions of TRUESDELL & NOLL [1965, 1] and COLEMAN, MARKOVITZ, & NOLL [1966, 1]. The equation (5.8) was proposed by COLEMAN & TROUPIN [1962, 3].

\*\* Cf. NOLL [1958, 1].

\*\*\* For details see the elementary discussion of COLEMAN, MARKOVITZ, & NOLL [1966, 1].

It is assumed, in classical mechanics, that contact forces transform as point differences under changes of frame, *i.e.* if  $s$  is the stress vector, then  $s \rightarrow s^*$ , where

$$s^* = Q(t) s. \quad (6.6)$$

Since  $s = S n$ , it follows from (6.2) and (6.6) that the stress tensor  $S$  is transformed into

$$S(t)^* = Q(t) S(t) Q(t)^{-1}, \quad (6.7)$$

or, in the notation of (6.4),

$$S(t)^* = R(0) S(t) R(0)^{-1}. \quad (6.8)$$

We here assume, further, that, under a change of frame,  $e$  and  $d$  transform as point differences, while  $b$  and  $h$  transform as axial vectors (*i.e.* the polar vectors of skew tensors); thus  $e \rightarrow e^*$ ,  $d \rightarrow d^*$ ,  $b \rightarrow b^*$ , and  $h \rightarrow h^*$  with

$$\begin{aligned} e^* &= Q(t) e = R(0) e, \\ d^* &= Q(t) d = R(0) d, \\ b^* &= [\det Q(t)] Q(t) b = [\det R(0)] R(0) b, \\ h^* &= [\det Q(t)] Q(t) h = [\det R(0)] R(0) h. \end{aligned} \quad (6.9)$$

In classical continuum physics, the idea that material properties should be independent of the observer, or frame of reference, is rendered mathematical by assuming the following postulate.

*Principle of Material Frame-Indifference*:\* If a given process is compatible with a constitutive assumption, then all processes obtained from that process by changes of frame must be compatible with the same constitutive assumption.

We here employ this principle. In so doing, we impose on our theory the space-time structure of classical mechanics and, further, make assumptions which imply that the simultaneous spinning of a dielectric and its electromagnetic field has no effect upon the polarization of the dielectric. The use of classical space-time seems appropriate to a theory which is to be applied only in situations involving "small speeds".\*\*

Let us consider first the electromagnetic constitutive equations (4.6). If  $h$ ,  $b$ ,  $d$ , and  $e$  obey these equations, and if  $h^*$ ,  $b^*$ ,  $d^*$ , and  $e^*$  are given by (6.9), then  $b^*$  and  $h^*$  obey (4.6)<sub>1</sub>, *i.e.*  $h^* = \mu_0^{-1} b^*$ , while  $e^*$  and  $d^*$  obey the equation

$$d^* = \varepsilon_0 K^* e^* \quad \text{with} \quad K^* = R(0) K R(0)^{-1}. \quad (6.10)$$

Thus, the defining equations of a perfect dielectric are compatible with the principle of material frame-indifference if and only if we assume that for changes of frame the dielectric tensor obeys the same transformation rule as the stress tensor, *i.e.*  $K \rightarrow K^*$ , where

$$K(t)^* = R(0) K(t) R(0)^{-1}. \quad (6.11)$$

---

\* Also called the "Principle of Objectivity"; see NOLL [1958, 1].

\*\* See the discussion after (4.5).

It follows that the refraction tensor  $N$ , defined in (4.10) also obeys this rule; *i.e.*  $N \rightarrow N^*$ , where

$$N(t)^* = R(0)N(t)R(0)^{-1}. \quad (6.12)$$

Frame-indifference requires that the constitutive functionals  $\mathfrak{N}$  and  $\mathfrak{S}$  in (5.8) and (5.9) be such that these equations remain valid whenever  $N(t)$ ,  $S(t)$ , and  $F_t^i$  are replaced by their transforms  $N(t)^*$ ,  $S(t)^*$ ,  $F_t^{i*}$  under a change of frame, *i.e.*

$$N(t)^* = \mathfrak{N}(F_t^{i*}), \quad S_D^* = \mathfrak{S}(F_t^{i*}), \quad (6.13)$$

or, by (6.12), (6.8), and (6.5),

$$R(0)N(t)R(0)^{-1} = \mathfrak{N}(RF_t^i R(0)^{-1}), \quad R(0)S_D(t)R(0)^{-1} = \mathfrak{S}(RF_t^i R(0)^{-1}). \quad (6.14)$$

Since (6.14) holds under every change of frame, it follows from (6.14), (5.8), and (5.9) that the functionals  $\mathfrak{N}$  and  $\mathfrak{S}$  must obey the relations

$$\mathfrak{N}(RF_t^i R(0)^{-1}) = R(0)\mathfrak{N}(F_t^i)R(0)^{-1}, \quad \mathfrak{S}(RF_t^i R(0)^{-1}) = R(0)\mathfrak{S}(F_t^i)R(0)^{-1} \quad (6.15)$$

for each  $*$  function  $R$  whose values are orthogonal tensors and for every history  $F_t^i$ .

Identities of the form (6.15) occur frequently in continuum physics, and solutions are now well known. If we let  $J^i$  be the function on  $[0, \infty)$  defined by

$$J^i(s) = F_t^i(s)^T F_t^i(s) - \mathbf{1}, \quad (6.16)$$

then it follows from (6.15)  $**$  that there exist two functionals  $\mathcal{M}$  and  $\mathcal{S}$  such that

$$\mathfrak{N}(F_t^i) = \mathcal{M}(J^i), \quad \mathfrak{S}(F_t^i) = \mathcal{S}(J^i), \quad (6.17)$$

and, moreover,  $\mathcal{M}$  and  $\mathcal{S}$  are isotropic functionals in the sense that for each constant orthogonal tensor  $Q$ , the relations,

$$\mathcal{M}(QJ^i Q^{-1}) = Q\mathcal{M}(J^i)Q^{-1}, \quad \mathcal{S}(QJ^i Q^{-1}) = Q\mathcal{S}(J^i)Q^{-1}, \quad (6.18)$$

hold as identities in  $J^i$ . Equation (6.17) permits us to write the following reduced forms of (5.8) and (5.9):

$$N(t) = \mathcal{M}(J^i), \quad (6.19)$$

$$S_D(t) = \mathcal{S}(J^i). \quad (6.20)$$

We call  $J^i$  the *relative strain history up to time  $t$* . By (5.6) and (6.16),

$$J^i(0) = \mathbf{0}, \quad (6.21)$$

where  $\mathbf{0}$  is the zero tensor. The condition (5.7) that the motion be isochoric can be written

$$\det(J^i(s) + \mathbf{1}) \equiv 1, \quad s \in [0, \infty). \quad (6.22)$$

---

\* More precisely, (6.15) holds for every history  $F_t^i$  in the domain  $\mathscr{D}$  of  $\mathfrak{N}$  and  $\mathfrak{S}$  and every orthogonal-tensor-valued function  $R$  on  $[0, \infty)$  such that  $RF_t^i$  is in  $\mathscr{D}$ . In COLEMAN & NOLL's theory of fading memory [1960, 1] [1961, 2], which we employ in Sections 9–12, if  $F_t^i$  is in  $\mathscr{D}$  and if  $R(s)$  is orthogonal for each  $s$ , then for  $RF_t^i$  to be in  $\mathscr{D}$  it suffices that  $R$  be measurable.

\*\* See NOLL [1958, 1]; he employed the right Cauchy–Green tensor,  $C_t^i = F_t^{iT} F_t^i$  instead of  $J^i$ ; the notation we use here is closer to that of COLEMAN & NOLL [1961, 2].

It is easy to show that the equations (6.19) and (6.20), with  $\mathcal{M}$  and  $\mathcal{S}$  obeying (6.18), supply not only a necessary, but also a sufficient condition for our theory to be compatible with the principle of material frame-indifference.

Although the relative strain history  $\mathbf{J}^t$  determines  $N(t)$  completely, in studies of birefringence it is only the deviator of  $N(t)$  that is important. Hence, instead of (6.19), we often employ the equation

$$N_D(t) = \mathcal{N}(\mathbf{J}^t). \quad (6.23)$$

The constitutive functional  $\mathcal{N}$  is determined by  $\mathcal{M}$  through the equation

$$\mathcal{N}(\mathbf{J}^t) = \mathcal{M}(\mathbf{J}^t) - \frac{1}{3} \mathbf{1} \operatorname{tr} \mathcal{M}(\mathbf{J}^t). \quad (6.24)$$

Clearly,

$$\operatorname{tr} \mathcal{N}(\mathbf{J}^t) \equiv \operatorname{tr} \mathcal{S}(\mathbf{J}^t) \equiv 0, \quad (6.25)$$

and, for each orthogonal tensor  $\mathbf{Q}$ ,

$$\mathcal{N}(\mathbf{Q}\mathbf{J}^t\mathbf{Q}^{-1}) = \mathbf{Q}\mathcal{N}(\mathbf{J}^t)\mathbf{Q}^{-1} \quad (6.26)$$

for all  $\mathbf{J}^t$ .

It follows from (5.3) and (5.5) that, in a body which has not experienced any change in configuration other than a rigid rotation,  $F_i^t(s)$  is, for each  $s$ , orthogonal, and hence  $F_i^t(s)^T F_i^t(s) = \mathbf{1}$ , or, by (6.16),  $\mathbf{J}^t = \mathbf{0}^\dagger$ , where  $\mathbf{0}^\dagger$  is the constant function on  $[0, \infty)$  whose value is the zero tensor:

$$\mathbf{0}^\dagger(s) \equiv \mathbf{0}, \quad s \in [0, \infty). \quad (6.27)$$

Furthermore, a familiar argument\* tells us that (6.18) and (6.25) imply that

$$\mathcal{N}(\mathbf{0}^\dagger) = \mathcal{S}(\mathbf{0}^\dagger) = \mathbf{0}, \quad (6.28)$$

or, by (6.19), (6.20), (6.24), and (5.10),

$$\mathbf{J}^t = \mathbf{0}^\dagger \Rightarrow N(t) = n^\circ \mathbf{1}, \quad S(t) = -p\mathbf{1}, \quad (6.29)$$

where  $n^\circ$  is a material constant defined by the relation

$$\mathcal{M}(\mathbf{0}^\dagger) = n^\circ \mathbf{1}, \quad (6.30)$$

and the number  $p$  is indeterminate in the sense that it is not determined by the local motion alone. Thus, *in a simple fluid which has experienced only rigid rotations, the velocity of light is the same for all directions and the stress reduces to a hydrostatic pressure.*

## 7. Shearing Flows

The constitutive equations (6.23) and (6.20) can often be simplified if something is known in advance about the motion the fluid is undergoing. In this section we describe the reduced forms taken by  $\mathcal{N}$  and  $\mathcal{S}$  in a broad class of motions called "shearing flows" or "generalized viscometric flows". The results given here are direct consequences of material symmetry and frame-indifference and thus do not require assumptions of linearity or smoothness for the constitutive functionals.

\* See NOLL [1958, 1] and COLEMAN & NOLL [1961, 1] p. 681.

We shall take up the “principle of fading memory” and its consequences in Sections 9 through 12.

A *shearing flow* is one for which there exists, at each material point  $P$  and time  $t$ , an orthonormal basis,  $\mathbf{h}_1, \mathbf{h}_2, \mathbf{h}_3$  (independent of  $s$ ), such that the components of  $\mathbf{J}^t(s)$  with respect to this basis have the form

$$[\mathbf{h}_i \cdot \mathbf{J}^t(s) \mathbf{h}_j] = \begin{bmatrix} \zeta(t-s)^2 & \zeta(t-s) & 0 \\ \zeta(t-s) & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad (7.1)$$

for all  $s \geq 0$ . The basis  $\mathbf{h}_i$  is called the *canonical basis at  $P$  at time  $t$* , while the number  $\zeta(t-s)$  is called the *amount of relative shear at  $P$  at time  $t-s$*  (relative to the configuration at time  $t$ ). Motions of this type were studied by COLEMAN & NOLL\* and TRUESDELL & NOLL\*\*, and known arguments\*\*\* here yield the following theorem, which rests heavily upon the identities (6.18), (6.26): *In a shearing flow of an incompressible simple fluid, the components, relative to  $\mathbf{h}_i$ , of  $N(t)$  and  $S(t)$  obey the relations*

$$\begin{aligned} N_{12}(t) &= \gamma_N(\zeta^t), \\ N_{11}(t) - N_{22}(t) &= w_N^{(1)}(\zeta^t), \\ N_{22}(t) - N_{33}(t) &= w_N^{(2)}(\zeta^t), \\ N_{13}(t) &= N_{23}(t) = 0, \end{aligned} \quad (7.2)$$

$$\begin{aligned} S_{12}(t) &= \gamma_S(\zeta^t), \\ S_{11}(t) - S_{22}(t) &= w_S^{(1)}(\zeta^t), \\ S_{22}(t) - S_{33}(t) &= w_S^{(2)}(\zeta^t), \\ S_{13}(t) &= S_{23}(t) = 0, \end{aligned} \quad (7.3)$$

where  $\zeta^t$  is the history of the amount of relative shear:

$$\zeta^t(s) = \zeta(t-s), \quad s \in [0, \infty).$$

The functionals  $\gamma_I, w_I^{(1)}, w_I^{(2)}$  ( $I = N, S$ ) are independent of the directions  $\mathbf{h}_i$  and obey the identities

$$\begin{aligned} \gamma_I(-\zeta^t) &= -\gamma_I(\zeta^t), \\ w_I^{(i)}(-\zeta^t) &= w_I^{(i)}(\zeta^t), \quad I = N, S, \quad i = 1, 2. \end{aligned} \quad (7.4)$$

It follows from (6.28) that

$$\gamma_I(0^\dagger) = w_I^{(1)}(0^\dagger) = w_I^{(2)}(0^\dagger) = 0, \quad I = N, S, \quad (7.5)$$

where  $0^\dagger$  is the constant function on  $[0, \infty)$  with value 0.

The proper vectors of  $N_D$  equal the proper vectors  $\mathbf{k}_i$  of  $N$ , i.e. the principal axes of refraction, while the proper vectors of  $S_D$  are the same as the proper

\* [1961, 1].

\*\* [1965, 1] § 106.

\*\*\* See in particular [1961, 1] § 5.

vectors  $s_i$  of  $S$  and are called *principal axes of stress*. It follows from (7.2) and (7.3) that one vector of the set  $(k_1, k_2, k_3)$ , say  $k_3$ , and one vector of the set  $(s_1, s_2, s_3)$ , say  $s_3$ , lie along the direction  $h_3$ , and hence  $k_1, k_2, s_1$ , and  $s_2$  all lie in the plane which contains  $h_1$  and  $h_2$ . Out of equilibrium, *i.e.* when  $\zeta^t$  does not equal  $0^t$ , the relations (7.2)–(7.4) give no reason to suppose that  $k_i = s_i$ , for  $i = 1, 2$ .

Let us index the proper vectors of  $N$  so that  $k_3 = h_3$ , as above, and so that the counterclockwise angle  $\chi$  from  $h_1$  to  $k_1$  satisfies the relation  $0 \leq \chi < \frac{\pi}{2}$ .<sup>\*</sup> Then an elementary calculation, starting with (7.2), shows that  $\chi$  is determined by the relation

$$\cot 2\chi = \frac{N_{11} - N_{22}}{2N_{12}}, \quad (7.6)$$

and the birefringence for light traveling in the direction  $k_3$  is given by

$$\Delta = n_1 - n_2 = \pm \sqrt{(N_{11} - N_{22})^2 + 4N_{12}^2} = (N_{11} - N_{22}) \cos 2\chi + 2N_{12} \sin 2\chi. \quad (7.7)$$

The last expression in (7.7) determines the sign of  $\Delta$ . In view of (7.2) we have

$$\cot 2\chi = \frac{w_N^{(1)}(\zeta^t)}{2y_N(\zeta^t)}, \quad (7.8)$$

and

$$\Delta = \pm \sqrt{w_N^{(1)}(\zeta^t)^2 + 4y_N(\zeta^t)^2} = w_N^{(1)}(\zeta^t) \cos 2\chi + 2y_N(\zeta^t) \sin 2\chi. \quad (7.9)$$

It follows from (7.4), (7.8), and (7.9) that  $\Delta$  and  $\chi - \frac{\pi}{4}$  are odd functions of  $\zeta^t$ ; *i.e.* the transformation  $\zeta^t \rightarrow -\zeta^t$  induces the transformations  $\Delta(t) \rightarrow -\Delta(t)$  and  $\chi - \frac{\pi}{4} \rightarrow -\left(\chi - \frac{\pi}{4}\right)$  [or  $\chi \rightarrow \frac{\pi}{2} - \chi$ ].

We see no reason to believe that the ratio  $w_N^{(1)}(\zeta^t)/y_N(\zeta^t)$  equals the ratio  $w_S^{(1)}(\zeta^t)/y_S(\zeta^t)$  for each history  $\zeta^t$  of an arbitrary incompressible fluid. Thus we repeat, with different wording and emphasis, an assertion made above: In spite of occasional claims to the contrary, there does not appear to be a general symmetry argument indicating that the principal axes of stress  $s_1, s_2$  are parallel to the principal axes of refraction  $k_1, k_2$  in all shearing flows of all incompressible fluids.

A *viscometric flow*<sup>\*\*</sup> is a shearing flow for which the function  $\zeta$  in (7.1) has the simple form

$$\zeta(t-s) = -\kappa s, \quad s \in [0, \infty), \quad (7.10)$$

<sup>\*</sup> Throughout this essay, when we say that an angle in the  $(k_1, k_2)$ -plane is measured “counterclockwise”, or “clockwise”, it is to be understood that the plane is viewed from the side toward which  $k_3$  points.

<sup>\*\*</sup> The definition of a “viscometric flow” employed here is that of COLEMAN [1962, 1], TRUESDELL & NOLL [1965, 1] call such motions “steady viscometric flows”. COLEMAN, MARKOVITZ, & NOLL [1966, 1] give an elementary exposition of the theory of these flows and discuss modern experiments in viscometry. See also [1959, 2 & 3].



with  $\kappa$  a number called the *rate of shear*. Let us define six functions, of the rate of shearing,  $\tau_N$ ,  $\sigma_N^{(1)}$ ,  $\sigma_N^{(2)}$  and  $\tau_S$ ,  $\sigma_S^{(1)}$ ,  $\sigma_S^{(2)}$ , by the relations

$$\tau_I(\kappa) = \mathcal{Y}_I \left( -\kappa s \right)_{s=0}^{\infty}, \quad \sigma_I^{(i)}(\kappa) = \mathcal{W}_I^{(i)} \left( -\kappa s \right)_{s=0}^{\infty}, \quad I = N, S, \quad i = 1, 2. \quad (7.11)$$

By  $\mathcal{Y}_I \left( -\kappa s \right)_{s=0}^{\infty}$  and  $\mathcal{W}_I^{(i)} \left( -\kappa s \right)_{s=0}^{\infty}$  is meant the values of the functionals  $\mathcal{Y}_I$  and  $\mathcal{W}_I^{(i)}$  at the function  $\zeta^t$  defined by  $\zeta^t(s) \equiv -\kappa s$ . The functions  $\tau_I$  and  $\sigma_I^{(i)}$ , mapping the real numbers into the real numbers, are material functions, determined by the fluid under consideration.\* It is clear from (7.8) and (7.9) that in a viscometric flow

$$\chi = \chi(\kappa) = \frac{1}{2} \cot^{-1} \frac{\sigma_N^{(1)}(\kappa)}{2\tau_N(\kappa)}, \quad (7.12)$$

and

$$\Delta = \Delta(\kappa) = \pm \sqrt{\sigma_N^{(1)}(\kappa)^2 + 4\tau_N(\kappa)^2} = \sigma_N^{(1)}(\kappa) \cos 2\chi + 2\tau_N(\kappa) \sin 2\chi. \quad (7.13)$$

As in the case of general shearing flows, there is no argument of symmetry implying coincidence of  $s_i$  and  $k_i$ , for  $i=1$  and  $2$ , in viscometric flows of arbitrary incompressible fluids.

The relations (7.4) and (7.5) yield \*\*

$$\tau_I(-\kappa) = -\tau_I(\kappa), \quad \sigma_I^{(i)}(-\kappa) = \sigma_I^{(i)}(\kappa), \quad (7.14)$$

$$\tau_I(0) = \sigma_I^{(i)}(0) = 0, \quad (7.15)$$

for  $I=N, S$  and  $i=1, 2$ . The remark made after (7.9) here implies that  $\Delta$  and  $\chi - \frac{\pi}{4}$  are odd functions of  $\kappa$ .

## 8. Couette Flow

As we have already mentioned, of the various shearing flows that can be obtained in the laboratory, Couette flow is the one most widely used for studying streaming birefringence. This shearing flow is particularly suited for determination of the material functionals  $\mathcal{Y}_N$  and  $\mathcal{W}_N^{(1)}$  in (7.2).

### a) General Theory

In a Couette flow the velocity field has the contravariant components,

$$\dot{r} = 0, \quad \dot{\theta} = v(r, t), \quad \dot{z} = 0, \quad (8.1)$$

---

\* *Cautionary note:* The function  $\sigma_S^{(1)}$  defined by (7.11) does *not* equal the function  $\sigma_1$  introduced by COLEMAN & NOLL [1959, 2 & 3] and subsequently employed in many papers, e.g. [1961, 1] [1962, 1] [1964, 2 & 5] [1965, 1] [1966, 1]. The present  $\tau_S$ ,  $\sigma_S^{(1)}$ , and  $\sigma_S^{(2)}$  are related as follows to the viscometric functions  $\tau$ ,  $\sigma_1$ , and  $\sigma_2$  discussed in the cited references:

$$\tau_S(\kappa) = \tau(\kappa), \quad \sigma_S^{(1)}(\kappa) = \sigma_2(\kappa) - \sigma_1(\kappa), \quad \sigma_S^{(2)}(\kappa) = \sigma_2(\kappa).$$

Our choice of notation is based on the observation that differences of the type  $I_{11} - I_{22}$  ( $I = N, S$ ) occur more frequently in the present subject than do the differences  $I_{11} - I_{33}$ .

\*\* Cf. COLEMAN & NOLL [1959, 2].

relative to a cylindrical coordinate system  $r, \theta, z$ , and the fluid is contained between two coaxial cylinders, located at  $r=R_1$  and  $r=R_2$ , which rotate about their common axis ( $r=0$ ) with angular velocities  $\Omega_1(t)$  and  $\Omega_2(t)$ .

It is easy to show that any motion obeying (8.1) is a shearing flow with

$$\zeta(t-s) = r \int_t^{t-s} \frac{\partial}{\partial r} v(r, \tau) d\tau = \zeta^t(s), \quad 0 \leq s < \infty. \quad (8.2)$$

Moreover, in such a flow, for each material point  $P$  and time  $t$ ,

$$\mathbf{h}_1 = \mathbf{e}_r, \quad \mathbf{h}_2 = \mathbf{e}_\theta, \quad \mathbf{h}_3 = \mathbf{e}_z, \quad (8.3)$$

where  $\mathbf{e}_r, \mathbf{e}_\theta, \mathbf{e}_z$  is the orthonormal basis of unit vectors pointing along the coordinate directions  $r, \theta, z$ , at the point occupied by  $P$  at time  $t$ . Thus, (7.2) and (7.3) here assert that the physical components of  $N(t)$  and  $S(t)$  in the coordinate system  $r, \theta, z$  (*i.e.* the components of  $N(t)$  and  $S(t)$  with respect to  $\mathbf{e}_r, \mathbf{e}_\theta, \mathbf{e}_z$ ) obey the relations

$$\begin{aligned} N_{r\theta}(t) &= \gamma_N(\zeta^t), \\ N_{rr}(t) - N_{\theta\theta}(t) &= w_N^{(1)}(\zeta^t), \\ N_{\theta\theta}(t) - N_{zz}(t) &= w_N^{(2)}(\zeta^t), \\ N_{rz}(t) &= N_{\theta z}(t) = 0, \end{aligned} \quad (8.4)$$

$$\begin{aligned} S_{r\theta}(t) &= \gamma_S(\zeta^t), \\ S_{rr}(t) - S_{\theta\theta}(t) &= w_S^{(1)}(\zeta^t), \\ S_{\theta\theta}(t) - S_{zz}(t) &= w_S^{(2)}(\zeta^t), \\ S_{rz}(t) &= S_{\theta z}(t) = 0. \end{aligned} \quad (8.5)$$

It follows from (8.5) that, in a Couette flow of an incompressible fluid, the equation of motion (5.1) reduces to the scalar functional-differential equation

$$r^3 \frac{\partial}{\partial t} v(r, t) = \frac{\partial}{\partial r} [r^2 \gamma_S(\zeta^t)] \quad (8.6)$$

with  $\zeta^t$  given by (8.2). One may presume that the equation (8.6), appropriate initial data, and the adherence conditions

$$v(R_1, t) = \Omega_1(t), \quad v(R_2, t) = \Omega_2(t), \quad (8.7)$$

uniquely determine the angular velocity  $v$  as a function of  $r$  and  $t$ , but a general theory to this effect, valid for non-linear functionals  $\gamma_S$  and non-steady motions, is lacking.

If the relative gap  $(R_2 - R_1)/R_1$  is small, and if  $\Omega_1(t), \Omega_2(t), \dot{\Omega}_1(t)$ , and  $\dot{\Omega}_2(t)$  are small for all  $t$ , one expects that  $v$  should be such that the rate of shear,  $r \frac{\partial v}{\partial r}$ , is independent of  $r$  and given to a high degree of accuracy by the first term on the right in the formula

$$r \frac{\partial}{\partial r} v(r, t) = \frac{R_1(\Omega_2(t) - \Omega_1(t))}{R_2 - R_1} + O\left(\frac{R_2 - R_1}{R_1}\right), \quad (8.8)$$

which yields

$$\zeta^t(s) = \frac{R_1}{R_2 - R_1} \int_t^{t-s} \Omega(\tau) d\tau + O\left(\frac{R_2 - R_1}{R_1}\right), \quad (8.9)$$

with

$$\Omega = \Omega_2 - \Omega_1. \quad (8.10)$$

Thus, when the term  $O((R_2 - R_1)/R_1)$  in (8.8) can be neglected, *i.e.* in the “small gap approximation”, the history  $\zeta^t$  is independent of  $r$  at each time  $t$ , and so also are all the terms in (8.4) and (8.5) as well as the principal axes of refraction  $k_i$  and the principal axes of stress  $s_i$ .

Optical measurements on Couette flow are usually performed with a plane polariscope mounted with its axis parallel to the axis of the bounding cylinders. Thus the light travels perpendicular to the planes  $z = \text{const.}$ ; these planes contain the circular streamlines, two of the axes of stress ( $s_1, s_2$ ), and two of the axes of refraction ( $k_1, k_2$ ). The observed isoclinic lines are the locus of points at which  $k_1$  or  $k_2$  is parallel to a polarizing axis of the analyzer. When (8.8) holds,  $k_1$  and  $k_2$  are independent of  $r$ , and as one sees in Figure 8.1, each isocline is a straight radial line. Indeed, in this case, at each instant  $t$  the isoclines form a cross (called the “cross of isocline”) whose two branches obey equations of the form

$$\begin{aligned} \theta = -\chi(t), \quad \theta = \frac{\pi}{2} - \chi(t), \quad \theta = \pi - \chi(t), \\ \theta = \frac{3\pi}{2} - \chi(t), \quad 0 \leq \chi(t) < \frac{\pi}{2}, \end{aligned} \quad (8.11)$$

with  $\theta$  measured counterclockwise from the direction of the polarizing axis of the polarizer.  $\chi$  is called the *extinction angle* and equals the clockwise angle from the axis of the polarizer to that arm of the cross which is nearest to it. In view of Figure 8.1, however,  $\chi$  equals also the counterclockwise angle from  $e_r$  to  $k_1$  and, by (8.3),  $\chi$  here has the same meaning as in the previous section. Thus, the equa-

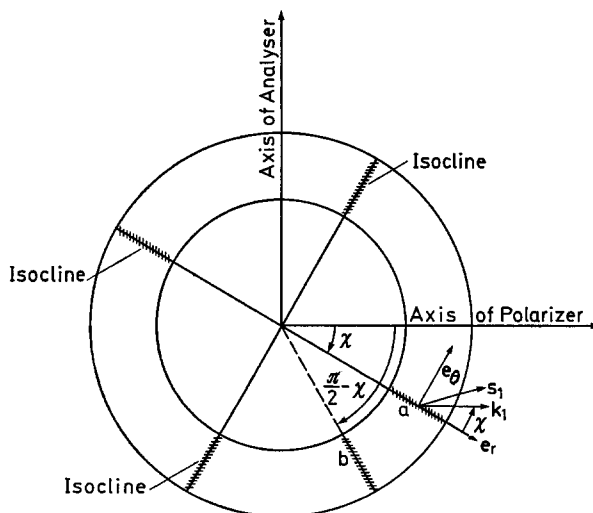


Fig. 8.1. Geometry of the cross of isocline

tions (7.6) and (7.8) here yield

$$\chi(t) = \frac{1}{2} \cot^{-1} \frac{N_{rr}(t) - N_{\theta\theta}(t)}{2N_{r\theta}(t)} = \frac{1}{2} \cot^{-1} \frac{w_N^{(1)}(\zeta^t)}{2y_N(\zeta^t)}. \quad (8.12)$$

If the polariscope is employed to measure the relative retardation of polarized light passing through the fluid in the  $z$ -direction, then, by (7.7), the birefringence obtained obeys the formula

$$\Delta = n_1 - n_2 = (N_{rr} - N_{\theta\theta}) \cos 2\chi + 2N_{r\theta} \sin 2\chi \quad (8.13)$$

and, according to (7.9),

$$\Delta(t) = \pm \sqrt{w_N^{(1)}(\zeta^t)^2 + 4y_N(\zeta^t)^2} = w^{(1)}(\zeta^t) \cos 2\chi + 2y_N(\zeta^t) \sin 2\chi. \quad (8.14)$$

In practice the Couette apparatus must be of finite length, and the flow (8.1) cannot be maintained near the ends. Thus there are difficulties in determining the length  $l$  to be used in (2.4), but as experimenters have found ways of overcoming analogous problems in viscometry,\* these difficulties do not appear insuperable.

Since

$$2N_{r\theta} = \Delta \sin 2\chi, \quad N_{rr} - N_{\theta\theta} = 2N_{r\theta} \cot 2\chi, \quad (8.15)$$

it is evident from (8.12) and (8.14) that when the dimensions of the apparatus and the speeds of rotation of the bounding cylinders are such that the approximation shown in (8.8) is valid, measurement of both the extinction angle  $\chi(t)$  and the birefringence\*\*  $\Delta(t)$  for a history  $\Omega^t$  of  $\Omega$ , *i.e.*

$$\Omega^t(s) = \Omega(t-s), \quad s \in [0, \infty), \quad (8.16)$$

permits calculation of  $y_N(\zeta^t)$  and  $w_N(\zeta^t)$  from the relations

$$\begin{aligned} y_N(\zeta^t) &= \frac{1}{2} \Delta(t) \sin 2\chi(t) \\ w_N^{(1)}(\zeta^t) &= \Delta(t) \cos 2\chi(t); \end{aligned} \quad (8.17)$$

$\zeta^t$  is here given by (8.9). Thus, one can, in principle, determine the functionals  $y_N$  and  $w_N^{(1)}$  experimentally.

### b) Oscillatory Couette Flow

If  $v$  in (8.1) is such that there exists a number  $\tau > 0$  for which

$$v(r, t + \tau) = v(r, t), \quad v\left(r, t + \frac{\tau}{2}\right) = -v(r, t), \quad (8.18)$$

for all  $r$  in  $[R_1, R_2]$  and all  $t > -\infty$ , and if  $\tau$  is the smallest positive number for which this is the case, then we say that the Couette flow *oscillates symmetrically with period*  $\tau$ .

\* See, for example, the survey [1966, 1].

\*\* Of course, intensity measurements and eq. (2.7) yield only  $|\Delta|$ .

It is clear from (8.2) that (8.18) yields

$$\zeta^t = \zeta^{t+\tau}, \quad \zeta^{t+\tau/2} = -\zeta^t, \quad (8.19)$$

and, in view of (8.4), (8.5), and (7.4), we have

$$\begin{aligned} N_{r\theta}(t+\tau) &= N_{r\theta}(t), & N_{r\theta}(t+\tau/2) &= -N_{r\theta}(t), \\ N_{rr}(t+\tau/2) - N_{\theta\theta}(t+\tau/2) &= N_{rr}(t) - N_{\theta\theta}(t), \\ N_{\theta\theta}(t+\tau/2) - N_{zz}(t+\tau/2) &= N_{\theta\theta}(t) - N_{zz}(t), \end{aligned} \quad (8.20)$$

and similar relations for the components of  $S$ .<sup>\*</sup> As a consequence of (8.19) and the remark made after (7.9), we have

$$\begin{aligned} \chi(t+\tau/2) &= \frac{\pi}{2} - \chi(t), & \Delta(t+\tau/2) &= -\Delta(t), \\ \chi(t+\tau) &= \chi(t), & \Delta(t+\tau) &= \Delta(t). \end{aligned} \quad (8.21)$$

Thus, in a Couette flow which oscillates symmetrically with period  $\tau$ , the birefringence  $\Delta$  and the extinction angle  $\chi$  are periodic functions of time with period  $\tau$ ; furthermore, in Figure 8.1, an isocline which lies on the line  $a$  at time  $t$  will be on the line  $b$  at time  $t + \tau/2$ .

### c) Steady Couette Flow

If (8.1) takes the form

$$\dot{r} = 0, \quad \dot{\theta} = v(r), \quad \dot{z} = 0 \quad (8.22)$$

with  $v$  independent of  $t$  for all  $t$ , then the motion is called a *steady Couette flow*. Of course, in such a flow,  $\Omega_1$ ,  $\Omega_2$ , and hence  $\Omega = \Omega_1 - \Omega_2$ , must be held constant for all  $t$ . It follows from (8.22) that (8.2) reduces to

$$\zeta^t(s) = -\kappa s, \quad s \in [0, \infty), \quad (8.23)$$

where

$$\kappa = \kappa(r) = r \frac{dv(r)}{dr}. \quad (8.24)$$

Since equation (8.23) is the same as equation (7.10), each steady Couette flow is a viscometric flow with a rate of shear  $\kappa$  that is a function of  $r$  alone. It follows from (8.8) that

$$\kappa = \frac{R_1}{R_2 - R_1} \Omega + O\left(\frac{R_2 - R_1}{R_1}\right). \quad (8.25)$$

Thus, in the "small gap approximation",  $\kappa$  is independent of  $r$ .

The equations (8.4) and (8.5) here become

$$\begin{aligned} N_{r\theta} &= \tau_N(\kappa), \\ N_{rr} - N_{\theta\theta} &= \sigma_N^{(1)}(\kappa), \\ N_{\theta\theta} - N_{zz} &= \sigma_N^{(2)}(\kappa), \\ N_{rr} &= N_{\theta z} = 0, \end{aligned} \quad (8.26)$$

<sup>\*</sup> Cf. COLEMAN & NOLL [1961, 1] pp. 698, 699.

$$\begin{aligned}
S_{r\theta} &= \tau_S(\kappa), \\
S_{rr} - S_{\theta\theta} &= \sigma_S^{(1)}(\kappa), \\
S_{\theta\theta} - S_{zz} &= \sigma_S^{(2)}(\kappa), \\
S_{r\theta} &= S_{\theta z} = 0,
\end{aligned} \tag{8.27}^*$$

with all terms independent of  $t$ . The equations (8.12) and (8.14) take the forms shown in (7.12) and (7.13), while (8.17) reduces to

$$\begin{aligned}
\tau_N(\kappa) &= \frac{1}{2} \Delta(\kappa) \sin 2\chi(\kappa), \\
\sigma_N^{(1)}(\kappa) &= \Delta(\kappa) \cos 2\chi(\kappa).
\end{aligned} \tag{8.28}$$

Hence, if one measures both  $\Delta$  and  $\chi$  as functions of  $\kappa$ , one can calculate the functions  $\tau_N$  and  $\sigma_N^{(1)}$ . We have no reason to expect  $\sigma_N^{(1)}$  to be proportional to  $\sigma_S^{(1)}$ .\*\*

It follows from the last sentence of Section 7 that

$$\Delta(-\kappa) = -\Delta(\kappa), \quad \chi(-\kappa) = \frac{\pi}{2} - \chi(\kappa). \tag{8.29}$$

Thus, the transformation  $\kappa \rightarrow -\kappa$  changes the sign of the birefringence and results in a change in the position of an isoclinic line from  $a$  to  $b$  in Figure 8.1. Of course, when  $\chi(\kappa) = \pi/4$  (i.e. if  $\sigma_N^{(1)}(\kappa)/\tau_N(\kappa) = 0$ ),\*\*\* the cross of isocline remains invariant under such reversal of the sign of  $\kappa$ .

## 9. Fading Memory

### a) Smoothness of Constitutive Functionals

For a broad class of materials, including those which are sometimes called "viscoelastic", recent deformations have a greater effect on the present values of  $N$  and  $S$  than earlier deformations of comparable magnitude. Such materials are said to have "fading memory", and the concept can be rendered precise in the following way.\*\*\*\*

A continuous, positive, monotone decreasing function  $h$  on  $[0, \infty)$  is here called an *influence function of order*  $r \geq 0$  if  $s^{r+\frac{1}{2}}h(s)$  is monotone decreasing for large  $s$  and

$$\lim_{s \rightarrow \infty} s^{r+\frac{1}{2}} h(s) = 0. \tag{9.1}$$

For examples,  $(s+1)^{-l}$  is an influence function of order  $r$  if  $0 < r + \frac{1}{2} < l$ , and  $e^{-\beta s}$ , with  $\beta > 0$ , is an influence function of all orders  $> 0$ . For a given influence function  $h$ , the set of all measurable symmetric-tensor-valued functions  $J$ , on  $[0, \infty)$ ,

\* The equations (8.27) were derived, at this level of generality, by COLEMAN & NOLL [1959, 2].

\*\*  $\sigma_S^{(1)}$  can be measured directly with apparatus of the type described by PADDEN & DE WITT [1954, 1].

\*\*\*  $\chi(\kappa)$  does equal  $\pi/4$  in the "limit of slow flow" mentioned in Section 3.

\*\*\*\* We here follow the theory of COLEMAN & NOLL [1960, 1] [1961, 2] [1962, 2], parts of which are generalized in the recent studies of COLEMAN & MIZEL [1966, 2] [1968, 1]. The closest presentation to the present is that of COLEMAN & MARKOVITZ [1964, 2]. Of course, in these earlier treatments attention is confined to mechanical, rather than optical effects.

for which the quantity,

$$\|J\|_h \stackrel{\text{def}}{=} \left[ \int_0^\infty \text{tr}(J(s)^2) h(s)^2 ds \right]^{\frac{1}{2}}, \quad (9.2)$$

is finite, form a Banach space  $\mathfrak{H}$  with norm  $\|\cdot\|_h$ . Indeed, with the inner product,

$$\langle J_1, J_2 \rangle \stackrel{\text{def}}{=} \int_0^\infty \text{tr}(J_1(s) J_2(s)) h(s)^2 ds, \quad (9.3)$$

$\mathfrak{H}$  is a Hilbert space. For each  $J$  in  $\mathfrak{H}$ , the number  $\|J\|_h$ , called the  $h$ -norm of  $J$ , is defined in such a way that it places greater emphasis on the values of  $J(s)$  at small  $s$  than on the values at large  $s$ . Two functions  $J_1$  and  $J_2$  in  $\mathfrak{H}$  are considered the same if  $\|J_1 - J_2\|_h = 0$ ; i.e. if  $J_1(s) = J_2(s)$  for almost all  $s$ .

Let  $\mathfrak{A}$  be the set of all functions  $J$  in  $\mathfrak{H}$  such that  $\mathbf{1} + J(s)$  is positive definite for almost all  $s$ . The physically meaningful part of the domain of the functionals  $\mathcal{N}$  and  $\mathcal{S}$  is the set  $\mathfrak{D}$  of functions  $J^t$  in  $\mathfrak{A}$  obeying (6.22). One may, however, extend  $\mathcal{N}$  and  $\mathcal{S}$  from  $\mathfrak{D}$  to  $\mathfrak{A}$  by putting

$$\mathcal{S}(J) = \mathcal{S}(G), \quad \mathcal{S} = \mathcal{N}, \mathcal{S}, \quad (9.4)$$

where  $G$ , defined by

$$G(s) = [\det(J(s) + \mathbf{1})]^{-\frac{1}{2}} [J(s) + \mathbf{1}] - \mathbf{1}, \quad (9.5)$$

is in  $\mathfrak{D}$ , whenever  $J$  is in  $\mathfrak{A}$ . We now assume that there is an influence function of order  $r > 2$ , which renders  $\mathcal{N}$  and  $\mathcal{S}$  not only continuous on  $\mathfrak{D}$  but also smooth in the sense that the extensions of  $\mathcal{N}$  and  $\mathcal{S}$  to  $\mathfrak{A}$  are twice Fréchet-differentiable at the “rest history” (6.27). This assumption may be called the “principle of fading memory”.

*Principle of Fading Memory:* For each fluid there exists an influence function of order greater than 2 such that if  $\mathfrak{H}$ ,  $\mathfrak{A}$ , and  $\mathfrak{D}$  are constructed as shown above, then  $\mathcal{N}$  and  $\mathcal{S}$  are well defined continuous functions mapping  $\mathfrak{D}$  into the set  $\mathcal{C}_D$  of symmetric tensors with zero trace. Furthermore, there exist continuous linear functions  $\delta\mathcal{N}$ ,  $\delta\mathcal{S}$ , mapping  $\mathfrak{A}$  into  $\mathcal{C}_D$ , and continuous, symmetric, bilinear functions  $\delta^2\mathcal{N}$  and  $\delta^2\mathcal{S}$ , mapping  $\mathfrak{A} \times \mathfrak{A}$  into  $\mathcal{C}_D$ , such that the extensions (9.4) of  $\mathcal{S}$  and  $\mathcal{N}$  obey the relation

$$\mathcal{S}(J) = \delta\mathcal{S}(J) + \frac{1}{2} \delta^2\mathcal{S}(J, J) + o(\|J\|_h^2), \quad \mathcal{S} = \mathcal{N}, \mathcal{S}, \quad (9.6)$$

for each  $J$  in  $\mathfrak{A}$ . Here, as usual,  $o(\|J\|_h^2)$  stands for an unspecified function of  $J$  obeying

$$\lim_{\|J\|_h \rightarrow 0} \frac{o(\|J\|_h^2)}{\|J\|_h^2} = 0. \quad (9.7)$$

The continuous linear functional  $\delta\mathcal{S}$  and the continuous symmetric bilinear functionals  $\delta^2\mathcal{S}$  are called the *first and second Fréchet derivatives of  $\mathcal{S}$  at the point  $\mathbf{0}^\dagger$  in  $\mathfrak{A}$* . The equation (9.6) determines  $\delta\mathcal{S}$  and  $\delta^2\mathcal{S}$  for functions  $J$  in  $\mathfrak{A}$ . However,  $\mathfrak{A}$  contains as a subset the cone  $\mathfrak{C}$  of functions  $J$  in  $\mathfrak{H}$  with  $J(s)$  positive-semidefinite for almost all  $s$ , and since  $\mathfrak{C}$  is an additive cone spanning  $\mathfrak{H}$ ,  $\delta\mathcal{S}$  has a unique continuous linear extension to  $\mathfrak{H}$ , and  $\delta^2\mathcal{S}$  has a unique continuous, symmetric, bilinear extension to  $\mathfrak{H} \times \mathfrak{H}$ . As every continuous linear functional on a

Hilbert space may be written as an inner product, the extension of  $\delta\mathcal{J}$  to  $\mathfrak{H}$  has an integral representation of the form

$$\delta\mathcal{J}(\mathbf{J}) = \int_0^\infty \Gamma_I(s) \{\mathbf{J}(s)\} h(s)^2 ds, \quad \mathcal{J} = \mathcal{N}, \mathcal{S}, \quad (9.8)$$

$$I = N \text{ when } \mathcal{J} = \mathcal{N}, \quad I = S \text{ when } \mathcal{J} = \mathcal{S};$$

here  $\Gamma_I(s)\{\cdot\}$ , is, for each  $s$ , a linear transformation of the space  $\mathcal{C}$  of symmetric tensors into the space  $\mathcal{C}_D$ , and

$$\int_0^\infty |\Gamma_I(s)|^2 h(s)^2 ds$$

is finite. Furthermore, it follows from (6.18), (6.26), (9.6), and (9.7) that for each  $\mathbf{J}$  in  $\mathfrak{H}$  and each constant orthogonal tensor  $\mathbf{Q}$ ,

$$\delta\mathcal{J}(\mathbf{Q}\mathbf{J}\mathbf{Q}^{-1}) = \mathbf{Q}\delta\mathcal{J}(\mathbf{J})\mathbf{Q}^{-1},$$

$$\delta^2\mathcal{J}(\mathbf{Q}\mathbf{J}\mathbf{Q}^{-1}, \mathbf{Q}\mathbf{J}\mathbf{Q}^{-1}) = \mathbf{Q}\delta^2\mathcal{J}(\mathbf{J}, \mathbf{J})\mathbf{Q}^{-1}, \quad \mathcal{J} = \mathcal{N}, \mathcal{S}, \quad (9.9)$$

and it is a consequence of the first of these identities that (9.8) reduces to

$$\delta\mathcal{J}(\mathbf{J}) = I' \mathbf{1} + \int_0^\infty m_I(s) \mathbf{J}(s) ds, \quad (9.10)$$

$$I = N \text{ when } \mathcal{J} = \mathcal{N}, \quad I = S \text{ when } \mathcal{J} = \mathcal{S};$$

where  $m_I$  is a material function determined by  $\mathcal{J}$ , and  $I'$ , chosen to make  $\text{tr } \mathcal{J}(\mathbf{J}) = 0$ , has the form\*

$$I' = -\frac{1}{3} \text{tr} \int_0^\infty m_I(s) \mathbf{J}(s) ds, \quad I = N, S. \quad (9.11)$$

Because the term  $\frac{1}{2} \delta^2(\mathbf{J}, \mathbf{J})$  in (9.6) is  $O(\|\mathbf{J}\|_h^2)$ , (6.20), (6.23), and (9.10) yield the following approximation formulae:

$$N_D(t) = N' \mathbf{1} + \int_0^\infty m_N(s) \mathbf{J}^t(s) ds + O(\|\mathbf{J}^t\|_h^2),$$

$$S_D(t) = S' \mathbf{1} + \int_0^\infty m_S(s) \mathbf{J}^t(s) ds + O(\|\mathbf{J}^t\|_h^2). \quad (9.12)$$

The equation (9.12)<sub>2</sub> was derived by COLEMAN & NOLL [1961, 2]; they called the constitutive equation obtained by striking out the term  $O(\|\mathbf{J}^t\|_h^2)$ , the “equation of finite linear viscoelasticity (for an incompressible fluid)”, and they discussed the sense in which such approximation formulae should be independent of the choice of strain measure. In this connection we remark that (9.12) remains valid if  $\mathbf{J}^t(s)$  is replaced throughout by

$$\mathbf{J}^{*t}(s) = (\mathbf{J}^t(s) + \mathbf{1})^{-1} - \mathbf{1} \quad (9.13)$$

or any other measure of strain which is related to  $\mathbf{J}^t(s)$  by a smoothly invertible transformation and has the property that  $\mathbf{J}^{*t}$  reduces to  $\mathbf{0}^t$  for a medium which has never been distorted.

\* The presence of  $I'$  in (9.10) is a result of the normalizations (5.10) and (6.24). For  $\mathbf{J} \in \mathfrak{D}$ ,  $I'$  is  $O(\|\mathbf{J}\|_h^2)$ .



If one is interested in approximations that include all terms  $O(\|\mathbf{J}'\|_h^2)$ , then, of course, in place of (9.12) one should use the following consequences of (9.6), (9.10), (6.20), and (6.23):

$$\begin{aligned} N_D(t) &= N' \mathbf{1} + \int_0^\infty m_N(s) \mathbf{J}'(s) ds + \frac{1}{2} \delta^2 \mathcal{N}(\mathbf{J}', \mathbf{J}') + o(\|\mathbf{J}'\|_h^2), \\ S_D(t) &= S' \mathbf{1} + \int_0^\infty m_S(s) \mathbf{J}'(s) ds + \frac{1}{2} \delta^2 \mathcal{S}(\mathbf{J}', \mathbf{J}') + o(\|\mathbf{J}'\|_h^2). \end{aligned} \quad (9.14)$$

The constitutive equation which results from neglect of the term  $o(\|\mathbf{J}'\|_h^2)$  in (9.14)<sub>2</sub> has been called the “equation of second-order viscoelasticity (for an incompressible fluid)”.<sup>\*</sup> If it be assumed that  $\delta^2 \mathcal{F}(\mathbf{J}, \mathbf{J})$  ( $\mathcal{F} = \mathcal{N}, \mathcal{S}$ ) is a sum of iterated integrals, then it can be shown that (9.9)<sub>2</sub> implies that  $\delta^2 \mathcal{F}$  has the form

$$\begin{aligned} \frac{1}{2} \delta^2 \mathcal{F}(\mathbf{J}, \mathbf{J}) &= I'' \mathbf{1} + \int_0^\infty \int_0^\infty [a_I(s_1, s_2) \mathbf{J}(s_1) \mathbf{J}(s_2) \\ &\quad + b_I(s_1, s_2) \mathbf{J}(s_1) \text{tr} \mathbf{J}(s_2)] ds_1 ds_2, \quad \mathcal{F} = \mathcal{N}, \mathcal{S}, \end{aligned} \quad (9.15)$$

where  $a_I$  and  $b_I$  ( $I = N, S$ ) are material functions determined by  $\mathcal{F}$ ;  $a_I$  is symmetric in the sense that  $a_I(s_1, s_2) = a_I(s_2, s_1)$ ; the number  $I''$  is chosen to make  $\text{tr} \mathcal{F}(\mathbf{J}, \mathbf{J}) = 0$ .

### b) The Relaxation Theorem

In the next two sections we shall discuss the way the relations (9.12) and (9.14) can be used to obtain the “equation of infinitesimal viscoelasticity” and the “equations of materials of differential type” as asymptotic forms of the constitutive equations (6.20) and (6.23). Before turning to the theory of motions of small amplitude and retarded motions, we should, however, like to state a consequence of the principle of fading memory whose demonstration requires not the differentiability of  $\mathcal{N}$  and  $\mathcal{S}$  but only the much weaker assumption that these functionals are continuous at the point  $\mathbf{J} = \mathbf{0}^\dagger$  in  $\mathfrak{D}$ .

Given any history  $\mathbf{J}$  in  $\mathfrak{D}$  and a positive number  $\delta$ , one may define a function  $\mathbf{J}^{[\delta]}$  as follows:

$$\mathbf{J}^{[\delta]}(s) = \begin{cases} \mathbf{0}, & s \in [0, \delta), \\ \mathbf{J}(s - \delta), & s \in [\delta, \infty); \end{cases} \quad (9.16)$$

$\mathbf{J}^{[\delta]}$  is called the *statical extension of  $\mathbf{J}$  by the amount  $\delta$* .<sup>\*\*</sup> If  $\mathbf{J}$  is the relative strain history up to some time, say  $t$ , then  $\mathbf{J}^{[\delta]}$  is the relative strain history up to time  $t + \delta$ , assuming that no further deformation (*i.e.* at most rigid rotations) occurred between  $t$  and  $t + \delta$ . Now, it is not hard to show that if  $h$  in (9.2) is an influence function of order 0 (or greater), then for each  $\mathbf{J}$  in  $\mathfrak{D}$ ,  $\mathbf{J}^{[\delta]}$  is also in  $\mathfrak{D}$  for all  $\delta > 0$ , and, furthermore,<sup>\*\*\*</sup>

$$\lim_{\delta \rightarrow \infty} \|\mathbf{J}^{[\delta]}\|_h = 0. \quad (9.17)$$

<sup>\*</sup> [1961, 2].

<sup>\*\*</sup> COLEMAN & NOLL [1962, 2] p. 539.

<sup>\*\*\*</sup> COLEMAN & NOLL [1962, 2], WANG [1964, 6], COLEMAN [1964, 1]. Weaker conditions on  $h$  sufficing for (9.17) are given by COLEMAN & MIZEL [1966, 2].

The equation (9.17) is sometimes called the “relaxation theorem”. As  $\mathcal{N}$  and  $\mathcal{S}$  are assumed continuous at the zero function  $\mathbf{0}^\dagger$ , (9.17) implies that

$$\lim_{\delta \rightarrow \infty} \mathcal{J}(\mathbf{J}^{[\delta]}) = \mathcal{J}(\mathbf{0}^\dagger), \quad \mathcal{J} = \mathcal{N}, \mathcal{S}, \quad (9.18)$$

or, by (6.28),

$$\lim_{\delta \rightarrow \infty} \mathcal{N}(\mathbf{J}^{[\delta]}) = \lim_{\delta \rightarrow \infty} \mathcal{S}(\mathbf{J}^{[\delta]}) = \mathbf{0}. \quad (9.19)$$

Thus, for a fluid with fading memory,  $N_D$  and  $S_D$  at a point which has been left undistorted for a long time are approximately  $\mathbf{0}$ , *i.e.* are approximately the same as  $N_D$  and  $S_D$  at a point which has never been distorted.

## 10. On Infinitesimal Deformations

In this and subsequent sections we shall examine certain situations in which the  $h$ -norm,  $\|\mathbf{J}^t\|_h$ , is small, so that the error terms  $o(\|\mathbf{J}^t\|_h^2)$  in (9.14), or the terms  $O(\|\mathbf{J}^t\|_h^2)$  in (9.12), can be neglected.

### *a) Relaxation Functions*

Intuition suggests that  $\|\mathbf{J}^t\|_h$  should be small if all the “strains” experienced by the material have been “infinitesimal”. To make precise the meaning of “infinitesimal strains”, let us pick a fixed reference configuration  $\mathcal{R}$  and put

$$\mathbf{H}(\tau) \stackrel{\text{def}}{=} \mathbf{F}(\tau) - \mathbf{1}, \quad (10.1)$$

$$\varepsilon \stackrel{\text{def}}{=} \sup_{-\infty < \tau < t} [\text{tr}(\mathbf{H}(\tau)\mathbf{H}(\tau)^T)]^{\frac{1}{2}},$$

where  $\mathbf{F}(\tau)$  is the deformation gradient at time  $\tau$  computed relative to  $\mathcal{R}$ . We say that *the strain relative to  $\mathcal{R}$  has been infinitesimal up to time  $t$  if*

$$\varepsilon \ll 1. \quad (10.2)$$

The displacement gradient tensor  $\mathbf{H}$  introduced in (10.1)<sub>1</sub> has the property that its symmetric part  $\mathbf{E}$  is the familiar *infinitesimal strain tensor* (relative to  $\mathcal{R}$ ):

$$\mathbf{E} = \frac{\mathbf{H} + \mathbf{H}^T}{2}. \quad (10.3)$$

It is not difficult to show that, for each  $s$ ,

$$\mathbf{J}^t(s) = 2\mathbf{E}(t-s) - 2\mathbf{E}(t) + O(\varepsilon^2). \quad (10.4)$$

Furthermore, by (9.2) and (10.1),

$$\|\mathbf{J}^t\|_h = O(\varepsilon). \quad (10.5)$$

Thus, it is indeed true that if  $\varepsilon$  is small, *i.e.* if the strains have been infinitesimal relative to some reference configuration  $\mathcal{R}$ , then  $\|\mathbf{J}^t\|_h$  is small. Of course  $\varepsilon$  is not small in viscometric flows. (In fact, for such motions, by (7.10),  $\varepsilon = \infty$ , for each  $\mathcal{R}$ .) But, in oscillatory motions of small amplitude there do exist references  $\mathcal{R}$  which

render  $\varepsilon$  small;  $\varepsilon$  is small also in relaxation tests for which the initial deformation,  $E^\circ$ , is small. [See (10.8) below.]

Now, it follows from (10.4) and (10.5) that, for small  $\varepsilon$ , the equations (9.12) reduce to\*

$$\begin{aligned} N_D(t) &= 2G_N(0)E(t) + 2\int_0^\infty \frac{dG_N(s)}{ds} E(t-s) ds + O(\varepsilon^2), \\ S_D(t) &= 2G_S(0)E(t) + 2\int_0^\infty \frac{dG_S(s)}{ds} E(t-s) ds + O(\varepsilon^2), \end{aligned} \quad (10.6)$$

where  $G_N$  and  $G_S$  are scalar valued functions related as follows to the functions  $m_N$  and  $m_S$  in (9.10)–(9.12):

$$G_I(s) \stackrel{\text{def}}{=} -\int_s^\infty m_I(\sigma) d\sigma, \quad \frac{dG_I(s)}{ds} = m_I(s), \quad I = N, S. \quad (10.7)$$

The absence from (10.6) of the terms  $N' \mathbf{1}$  and  $S' \mathbf{1}$  is due to the fact that in an isochoric motion, to within an error  $O(\varepsilon^2)$ ,  $\text{tr } E = 0$ , and hence the linear terms shown in (10.7) already give  $\text{tr } S_D = \text{tr } N_D = 0$  to within an error  $O(\varepsilon^2)$ .

The terms exhibited explicitly in (10.6)<sub>2</sub> are  $O(\varepsilon)$  and have precisely the forms found in Boltzmann's classical theory of linear viscoelasticity, specialized, of course, to incompressible fluids. The corresponding terms in (10.6)<sub>1</sub> give an analogous linear theory of induced birefringence. We call the theory which results from ignoring the terms  $O(\varepsilon^2)$  in (10.6)<sub>1&2</sub> the *linear theory of infinitesimal viscoelasticity for incompressible fluids*. Rheologists call the material function  $G_S$  the *shear relaxation modulus* or the *stress relaxation function for shear*; we may call  $G_N$  the *optical relaxation function for shear*.

For a "relaxation test", i.e. an experiment in which  $E$  has the special time-dependence

$$E(t) = \begin{cases} \mathbf{0}, & \text{for } t < 0, \\ E^\circ, & \text{for } t \geq 0, \end{cases} \quad (10.8)$$

the equations (10.6) yield, for  $t > 0$ ,

$$\begin{aligned} N_D(t) &= 2G_N(t)E^\circ + O(\varepsilon^2), \\ S_D(t) &= 2G_S(t)E^\circ + O(\varepsilon^2). \end{aligned} \quad (10.9)$$

Thus we see why  $G_N$  and  $G_S$  are called "relaxation functions": in the linear theory of infinitesimal viscoelasticity, if one knows  $G_N$  (or  $G_S$ ) one knows, without much calculation, how  $N_D$  (or  $S_D$ ), gradually "relaxes to zero" after a sudden increment in strain.

### b) Creep Functions

If one assumes that, under neglect of the terms  $O(\varepsilon^2)$ , (10.6)<sub>2</sub> can be inverted, then the inverse has the following form, well known in Boltzmann's theory of

\* Cf. [1961, 2], [1962, 2], [1964, 2], [1964, 4].

viscoelasticity and sometimes called “Volterra’s relation”:

$$2E(t) = J_E(0)S_D(t) + \int_0^{\infty} \frac{dJ_E(s)}{ds} S_D(t-s) ds. \quad (10.10)$$

The material function  $J_E$ , called the *creep compliance function for shear*, obeys the equation,

$$\int_0^t G_S(s) J_E(t-s) ds = t, \quad t \in [0, \infty), \quad (10.11)$$

which, among other things, implies

$$J_E(s) G_S(s) \leq 1, \quad s \in [0, \infty). \quad (10.12)$$

If either  $G_S$  or  $J_E$  is known, the other can be calculated from (10.11).

Employing (10.10) to eliminate  $E(\cdot)$  from (10.6)<sub>1</sub>, we find that, when terms  $O(\varepsilon^2)$  can be neglected,

$$\begin{aligned} N_D(t) = & G_N(0) J_E(0) S_D(t) + \int_0^{\infty} \left[ G_N(0) \frac{dJ_E(s)}{ds} + \frac{dG_N(s)}{ds} J_E(0) \right. \\ & \left. + \int_0^s \frac{dG_N(\tau)}{d\tau} \frac{dJ_E(s-\tau)}{ds} d\tau \right] S_D(t-s) ds. \end{aligned} \quad (10.13)$$

Hence, if we put

$$J_N(s) \stackrel{\text{def}}{=} G_N(0) J_E(s) + \int_0^s \frac{dG_N(\tau)}{d\tau} J_E(s-\tau) d\tau, \quad (10.14)$$

then, in the linear theory of infinitesimal viscoelasticity, we have

$$N_D(t) = J_N(0) S_D(t) + \int_0^{\infty} \frac{dJ_N(s)}{ds} S_D(t-s) ds. \quad (10.15)$$

We call  $J_N$  the *optical creep function for shear*. Through integration by parts, one may show that equivalent to (10.14) is the formula

$$J_N(s) = G_N(s) J_E(0) + \int_0^s G_N(s-\tau) \frac{dJ_E(\tau)}{d\tau} d\tau. \quad (10.16)$$

For a creep test, *i.e.* an experiment in which  $S_D(t)$  is controlled in such a way that

$$S_D(t) = \begin{cases} \mathbf{0}, & \text{for } t < 0, \\ S_D^{\circ}, & \text{for } t \geq 0, \end{cases} \quad (10.17)$$

the equations (10.10) and (10.15) yield, for  $t > 0$ ,

$$2E(t) = J_E(t) S_D^{\circ}, \quad N_D(t) = J_N(t) S_D^{\circ}. \quad (10.18)$$

Thus, when the approximations made in the linear theory of viscoelasticity are valid, in a creep test on a fluid the deviators of the stress and refraction tensors are proportional, but the proportionality depends on time; *i.e. even under neglect of all terms  $O(\varepsilon^2)$ , our theory shows optical creep.*

c) *Harmonic Strains and Loads*

In the linear theory of infinitesimal viscoelasticity, the mechanical and optical response of an incompressible fluid is completely determined by the functions  $G_S$  and  $G_N$ , which may be measured in a relaxation test, or by the functions  $J_E$  and  $J_N$ , which have simple interpretations in creep tests. More common tests for fluids are, however, those involving harmonic strains, and therefore we consider now motions for which the infinitesimal strain tensor  $E$  is a sinusoidal function of time with circular frequency  $\omega$  and phase  $\delta_E$ ,

$$E(t) = E^\circ \sin(\omega t + \delta_E), \quad t \in (-\infty, \infty); \quad (10.19)$$

here  $E^\circ$  is a constant symmetric tensor with  $\text{tr } E^\circ = 0$ . For the special function (10.19), the equations (10.6) yield \*

$$\begin{aligned} N_D(t) &= 2[G'_N(\omega) \sin(\omega t + \delta_E) + G''_N(\omega) \cos(\omega t + \delta_E)] E^\circ + O(\varepsilon^2), \\ S_D(t) &= 2[G'_S(\omega) \sin(\omega t + \delta_E) + G''_S(\omega) \cos(\omega t + \delta_E)] E^\circ + O(\varepsilon^2). \end{aligned} \quad (10.20)$$

For each  $\omega \geq 0$ , the real numbers  $G'_S(\omega)$  and  $G''_S(\omega)$ , called, respectively, the *shear storage modulus* and the *shear loss modulus*, are defined by

$$\begin{aligned} G'_S(\omega) &\stackrel{\text{def}}{=} \omega \int_0^\infty G_S(s) \sin \omega s \, ds, \\ G''_S(\omega) &\stackrel{\text{def}}{=} \omega \int_0^\infty G_S(s) \cos \omega s \, ds. \end{aligned} \quad (10.21)$$

We call  $G'_N(\omega)$  and  $G''_N(\omega)$ , respectively, the *in-phase* and *out-of-phase strain-optical coefficients*; these numbers are given by

$$\begin{aligned} G'_N(\omega) &\stackrel{\text{def}}{=} \omega \int_0^\infty G_N(s) \sin \omega s \, ds, \\ G''_N(\omega) &\stackrel{\text{def}}{=} \omega \int_0^\infty G_N(s) \cos \omega s \, ds. \end{aligned} \quad (10.22)$$

It is clear that, under appropriate assumptions of regularity, the Fourier transforms shown in (10.21) and (10.22) can be inverted; hence  $G_I$  ( $I = N, S$ ) can be determined if either  $G'_I$  or  $G''_I$  is known. Thus, measurement of the response to a sinusoidal input (10.19) supplies a convenient way of obtaining the relaxation functions  $G_N$  and  $G_S$ . Equations equivalent to (10.20) are

$$\begin{aligned} N_D(t) &= N_D^* \sin(\omega t + \delta_N) + O(\varepsilon^2), \\ S_D(t) &= S_D^* \sin(\omega t + \delta_S) + O(\varepsilon^2), \end{aligned} \quad (10.23)$$

where

$$\begin{aligned} N_D^* &= 2E^\circ \sqrt{G'_N(\omega)^2 + G''_N(\omega)^2}, \\ \tan(\delta_N - \delta_E) &= \frac{G''_N(\omega)}{G'_N(\omega)}, \end{aligned} \quad (10.24)$$

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\* The primes in eqs. (10.20)–(10.22) do *not* signify derivatives; unfortunately, the notation,  $G$ ,  $G'$ , and  $G''$ , for relaxation moduli, storage moduli, and loss moduli, has become standard; cf. LEADERMAN [1957, 1].

and

$$\begin{aligned} \mathbf{S}_D^* &= 2E^o \sqrt{\mathbf{G}'_S(\omega)^2 + \mathbf{G}''_S(\omega)^2}, \\ \tan(\delta_S - \delta_E) &= \frac{\mathbf{G}''_S(\omega)}{\mathbf{G}'_S(\omega)}. \end{aligned} \quad (10.25)$$

A direct relation between  $\mathbf{S}_D(t)$  and  $N_D(t)$  can be obtained by substituting (10.23) into (10.15); we find that if one defines an *in-phase stress-optical coefficient*  $J'_N$  by

$$J'_N(\omega) \stackrel{\text{def}}{=} \omega \int_0^\infty J_N(s) \sin \omega s \, ds, \quad (10.26)$$

and an *out-of-phase stress-optical coefficient*  $J''_N$  by

$$J''_N(\omega) \stackrel{\text{def}}{=} \omega \int_0^\infty J_N(s) \cos \omega s \, ds, \quad (10.27)$$

then, to within an error  $O(\varepsilon^2)$ ,

$$N_D(t) = [J'_N(\omega) \sin(\omega t + \delta_S) + J''_N(\omega) \cos(\omega t + \delta_S)] \mathbf{S}_D^*, \quad (10.28)$$

or, equivalently,

$$\begin{aligned} N_D^* &= \mathbf{S}_D^* \sqrt{J'_N(\omega)^2 + J''_N(\omega)^2}, \\ \tan(\delta_N - \delta_S) &= \frac{J''_N(\omega)}{J'_N(\omega)}. \end{aligned} \quad (10.29)$$

If we let  $\mathcal{G}_N(\omega)$ ,  $\mathcal{G}_S(\omega)$ , and  $\mathcal{J}_N(\omega)$  be the complex numbers defined by

$$\begin{aligned} \mathcal{G}_I(\omega) &= \mathbf{G}'_I(\omega) + i\mathbf{G}''_I(\omega), \quad I = N, S, \quad i^2 = -1, \\ \mathcal{J}_N(\omega) &= J'_N(\omega) + iJ''_N(\omega), \end{aligned} \quad (10.30)$$

then

$$\mathcal{J}_N(\omega) = \frac{\mathcal{G}_N(\omega)}{\mathcal{G}_S(\omega)}. \quad (10.31)$$

In (10.23) and (10.29) we see that, when terms  $O(\varepsilon^2)$  are neglected, the principal axes of stress and refraction coincide at each time in harmonic straining of the type (10.19); however, the principal stresses and principal strains have generally different phase angles.

We do not wish to give here an extensive résumé of linear hereditary theories. The student of such theories will recognize that we have introduced only a few ( $\mathbf{G}_S$ ,  $\mathbf{J}_E$ ,  $\mathbf{G}'_S$ ,  $\mathbf{G}''_S$ ) of a long list of the mechanical-response functions to be found in the standard treatises on linear viscoelasticity. The important point is that the mechanical properties of an incompressible fluid are completely characterized in the linear theory of infinitesimal deformations if any one of the functions  $\mathbf{G}_S$ ,  $\mathbf{J}_E$ ,  $\mathbf{G}'_S$ ,  $\mathbf{G}''_S$  is known, and, in our present non-linear theory, each of these functions suffices to determine the Fréchet derivative  $\delta\mathcal{S}$  in (9.6). [See the relations (9.10) and (10.7).] Similarly, the optical properties corresponding to a history with small  $\varepsilon$  are known whenever any one of the functions  $\mathbf{G}_N$ ,  $\mathbf{G}'_N$ ,  $\mathbf{G}''_N$  is known, and each of these functions suffices to determine  $\delta\mathcal{N}$  in (9.6). Furthermore,  $\mathbf{G}_N$ ,  $\mathbf{G}'_N$ , and  $\mathbf{G}''_N$  can be calculated if one knows one function from each of the sets  $\{J_N, J'_N, J''_N\}$ ,  $\{\mathbf{G}_S, \mathbf{J}_E, \mathbf{G}'_S, \mathbf{G}''_S\}$ .

d) *Harmonic Couette Flow*

As an example of a motion in the family (10.19) of harmonic motions, we consider a circumferential flow between rotating coaxial cylinders and employ a cylindrical coordinate system set up so that the line  $r=0$  gives the common axis of the bounding cylinders. Letting  $r, \theta, z$  be the coordinates at time  $t$  of the material point which is located at  $R, \Theta, Z$  in a fixed reference configuration  $\mathcal{R}$ , we suppose that the motion has the form

$$r=R, \quad \theta=\Theta+\beta(r, t)=\Theta+\alpha(r)\sin(\omega t+\delta_E), \quad z=Z. \quad (10.32)$$

This is clearly a Couette flow for which  $v$  in (8.1) is the function

$$v(r, t)=\omega\alpha(r)\cos(\omega t+\delta_E). \quad (10.33)$$

At each instant  $t$  the deformation gradient  $F$  relative to the fixed configuration  $\mathcal{R}$  has the form

$$F(t)=P(r, t)[1+M(r, t)], \quad (10.34)$$

where  $M^2=0$ , and  $P$  is the orthogonal tensor mapping the orthonormal basis  $e_R, e_\Theta, e_Z$  along the coordinate directions in the reference configuration into the orthonormal basis  $e_r, e_\theta, e_z$  along the coordinate directions at the place occupied at time  $t$ ; the components of  $P$  and  $M$  relative to the basis  $e_R, e_\Theta, e_Z$ , have the matrices

$$[P]=\begin{bmatrix} \cos\beta & -\sin\beta & 0 \\ \sin\beta & \cos\beta & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad (10.35)$$

$$[M]=\begin{bmatrix} 0 & 0 & 0 \\ r\frac{\partial\beta}{\partial r} & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}.$$

It follows that  $\varepsilon$ , defined in (10.1), is here given by

$$\varepsilon^2 = \sup_{-\infty < \tau < t} \left[ 4[1 - \cos\beta(r, \tau)] + 2r \frac{\partial\beta(r, \tau)}{\partial r} \sin\beta(r, \tau) + r \left[ \frac{\partial\beta(r, \tau)}{\partial r} \right]^2 \right]. \quad (10.36)$$

Hence,  $\varepsilon$  is small if both  $\alpha$  and  $d\alpha/dr$  are small at all times. Assuming such is the case, we calculate the infinitesimal strain tensor  $E$  of (10.3), and we find that

$$E(t)=E^\circ \sin(\omega t+\delta_E)+O(\varepsilon^2) \quad (10.37)$$

where  $E^\circ$  has components

$$2E_{r\theta}^\circ = r \frac{d\alpha}{dr}, \quad (10.38)$$

$$E_{rr}^\circ = E_{\theta\theta}^\circ = E_{zz}^\circ = E_{rz}^\circ = E_{z\theta}^\circ = 0,$$

with respect to the basis  $e_r, e_\theta, e_z$ . Thus, when terms  $O(\varepsilon^2)$  can be neglected, the motion (10.32) is indeed of the type (10.19). It follows from this, (10.23), and (10.24)

that the components of  $N$  obey the equations

$$\begin{aligned} N_{r\theta}(t) &= N_{r\theta}^{\circ} \sin(\omega t + \delta_N) + O(\varepsilon^2), \\ N_{r,r}(t) - N_{\theta\theta}(t) &= O(\varepsilon^2), \\ N_{\theta\theta}(t) - N_{z,z}(t) &= O(\varepsilon^2), \end{aligned} \quad (10.39)$$

where

$$N_{r\theta}^{\circ} = r \frac{d\alpha}{dr} \sqrt{G'_N(\omega)^2 + G''_N(\omega)^2}, \quad (10.40)$$

$$\delta_N = \delta_E + \tan^{-1}(G''_N(\omega)/G'_N(\omega)). \quad (10.41)$$

Under the "small gap approximation" [see (8.8)–(8.10)], the number

$$E_{r\theta}^{\circ} = \frac{1}{2} r \frac{d\alpha}{dr}$$

is a constant independent of  $r$ ; furthermore, if the inner bounding cylinder ( $r = R_1$ ) is kept at rest while the outer cylinder ( $r = R_2$ ) is constrained to undergo, for all  $t$ , a sinusoidal oscillation of the form

$$\theta(R_2, \Theta, t) = \Theta + A \sin(\omega t + \delta_E), \quad (10.42)$$

then (10.32) holds with

$$E_{r\theta}^{\circ} = \frac{R_1 A}{R_2 - R_1} + O\left(\frac{R_2 - R_1}{R_1}\right). \quad (10.43)$$

It follows from (10.39) and equation (8.12)<sub>1</sub> that, in the linear theory of viscoelasticity (i.e. when the terms  $O(\varepsilon^2)$  in (10.39) are ignored), the extinction angle  $\chi$  is  $\pi/4$ . In addition, (8.13), (10.39), and (10.40) yield the following expression for the birefringence for light traveling in the  $z$ -direction:

$$\Delta = \Delta^{\circ} \sin(\omega t + \delta_N) + O(\varepsilon^2); \quad (10.44)$$

$\delta_N$  is given by (10.41), and

$$\Delta^{\circ} = 4E_{r\theta}^{\circ} \sqrt{G'_N(\omega)^2 + G''_N(\omega)^2}. \quad (10.45)$$

Thus, as  $E_{r\theta}^{\circ}$  and  $\delta_E$  are determined, in principle, by the motion of the bounding cylinders, measurement (for small  $E_{r\theta}^{\circ}$ ) of the amplitude  $\Delta^{\circ}$  and the phase angle  $\delta_N$  of the birefringence  $\Delta$  yields the sum  $G'_N(\omega)^2 + G''_N(\omega)^2$  and the ratio  $G''_N(\omega)/G'_N(\omega)$  and hence permits a calculation of in-phase and out-of-phase strain-optical coefficients  $G'_N(\omega)$  and  $G''_N(\omega)$ . If such measurements are made over a wide range of values of  $\omega$ , it should be possible to calculate, by Fourier-transform methods,  $G_N(s)$  over a comparable range of values of  $s^{-1}$ .

Under the approximations of "small gap" and small  $\varepsilon$ , the torque  $M$  per unit height which must be applied to bounding cylinders to maintain their relative motion (10.39) is given by \*

$$M(t) = M^{\circ} \sin(\omega t + \delta_S), \quad (10.46)$$

where

$$M^{\circ} = 4\pi R_1 R_2 E_{r\theta}^{\circ} \sqrt{G'_S(\omega)^2 + G''_S(\omega)^2}, \quad (10.47)$$

\* For a thorough analysis, within the framework of the linear viscoelasticity, see MARKOVITZ [1952, 1].



and  $\delta_S$  is as in (10.25)<sub>2</sub>. Hence, as every rheologist knows, measurement of amplitude  $M^\circ$  and phase  $\delta_S$  of the torque  $M$  permits a calculation of  $G_S'(\omega)$  and  $G_S''(\omega)$ . Once these storage and loss moduli are known, one can calculate the in-phase and out-of-phase stress-optical coefficients  $J_N'(\omega)$  and  $J_N''(\omega)$  from the strain-optical coefficients  $G_N'(\omega)$  and  $G_N''(\omega)$  as shown in (10.30) and (10.31). Knowledge of  $G_S'(\omega)$ ,  $G_S''(\omega)$ ,  $J_N'(\omega)$ , and  $J_N''(\omega)$  over a wide range of values of  $\omega$  should permit calculation of the creep functions  $J_E$  and  $J_N$  by Fourier methods.

## 11. On Slow Flows

### a) Asymptotic Forms of the Constitutive Equations

For  $\|J^t\|_h$  to be small, *i.e.* for the relations (9.12) and (9.14) to be useful, it is not necessary that there exist an  $\mathcal{R}$  which renders  $\varepsilon$  in (10.1) small. In fact, it follows from (9.2) and our assumptions about  $h$  that for  $\|J^t\|_h$  to be small it is sufficient that the deformation relative to the present configuration be small in the recent past. Such is the case in motions which are "slow".

If  $J$  is a function on  $[0, \infty)$  and  $\alpha$  a number in  $(0, 1)$ , then the function  $J^{(\alpha)}$ , defined by

$$J^{(\alpha)}(s) \stackrel{\text{def}}{=} J(\alpha s), \quad s \in [0, \infty), \quad (11.1)$$

is called the *retardation of J by the factor  $\alpha$* .<sup>\*</sup> If, after a suitable alteration of  $J$  on a set of measure zero, the limits

$$\stackrel{(0)}{J} = \lim_{s \rightarrow 0} J(s), \quad \stackrel{(l)}{J} = \lim_{s \rightarrow 0} \frac{l!}{s^l} \left( J(s) - \sum_{k=0}^{l-1} \frac{s^k}{k!} \stackrel{(k)}{J} \right), \quad (11.2)$$

exist for  $l=0, 1, \dots, n$ , then one says that  $J$  has  $n$  *generalized derivatives at  $s=0$* . The existence of ordinary derivatives implies the existence of the corresponding generalized derivatives, and

$$\stackrel{(0)}{J} = J(0), \quad \stackrel{(l)}{J} = \left. \frac{d^l J(s)}{ds^l} \right|_{s=0}. \quad (11.3)$$

Let an influence function  $h$  be given, let the Hilbert space  $H$  and its subsets  $\mathfrak{A}$  and  $\mathfrak{D}$  be constructed as in Section 9, and for each integer  $n$  let  $\mathfrak{D}_n$  be the set of functions  $J$  in  $\mathfrak{D}$  which have  $n$  generalized derivatives at  $s=0$ . If the influence function  $h$  is of order greater than  $n$ , then the  $n$ 'th-order *Taylor transformation*  $\Pi_n$ , defined by

$$\Pi_n J(s) = \sum_{l=0}^n \frac{s^l}{l!} \stackrel{(l)}{J}, \quad (11.4)$$

maps  $\mathfrak{D}_n$  into  $\mathfrak{A}$ , and, by a theorem of COLEMAN & NOLL,<sup>\*\*</sup> for each  $J$  in  $\mathfrak{D}_n$ .

$$\lim_{\alpha \rightarrow 0} \frac{1}{\alpha^n} \|J^{(\alpha)} - \Pi_n J^{(\alpha)}\|_h = 0. \quad (11.5)$$

If  $J$  is a relative strain history, as in (6.16), then  $J(0)=0$ , and the tensor

$$A_{(n)} \stackrel{\text{def}}{=} (-1)^n \stackrel{(n)}{J}, \quad n = 1, 2, \dots \quad (11.6)$$

<sup>\*</sup> See COLEMAN & NOLL [1960, 1].

<sup>\*\*</sup> [1960, 1], Thm. 1, pp. 359–361.

is called the  $n^{\text{th}}$  Rivlin-Ericksen tensor. By (11.1), the Rivlin-Ericksen tensors  $A_{(n)}^{(\alpha)}$  corresponding to the retarded history  $J^{(\alpha)}$  are related to those corresponding to  $J$  through the formula

$$A_{(n)}^{(\alpha)} = \alpha^n A_{(n)}. \quad (11.7)$$

We here assume that  $A_{(n)}$  exists, for  $n=0, 1, 2$ ; that is, we consider only histories  $J$  for which there are symmetric tensors  $A_{(1)}$  and  $A_{(2)}$  such that (possibly after alteration of  $J$  on a set of measure zero)

$$\begin{aligned} \lim_{s \rightarrow 0} J(s) = \mathbf{0} = J, \quad \lim_{s \rightarrow 0} \frac{J(s)}{s} = -A_{(1)} = J^{(1)}, \\ \lim_{s \rightarrow 0} \frac{2}{s^2} [J(s) + sA_{(1)}] = A_{(2)} = J^{(2)}. \end{aligned} \quad (11.8)$$

It is known that  $A_{(1)} = 2D$ , where  $D$ , the symmetric part of the velocity gradient tensor, is called the *rate of deformation tensor* and is familiar in the theory of Newtonian fluids.  $A_{(2)}$  supplies a "frame-indifferent" measure of the acceleration gradient. As in our original statement of the principle of fading memory in Section 9, we assume here that the influence function is of order greater than 2. It then follows from (11.5) and (11.8)<sub>1</sub> that

$$\|J^{(\alpha)}\|_h = O(\alpha). \quad (11.9)$$

In other words, not only does a slow flow give rise to a small  $h$ -norm, but, if one retards any history that is continuous at  $s=0$ , one will find that the  $h$ -norm goes to zero at least as fast as the retardation factor  $\alpha$ .

From (9.12), (11.8), and (11.5) with  $n=1$ , one may easily derive the following equations which give, to within an error  $o(\alpha)$ , explicit expressions for the deviators of the refraction tensor and the stress tensor corresponding to the retardation  $J^{(\alpha)}$  of a relative strain history  $J$ :

$$\begin{aligned} N_D^{(\alpha)} &= \eta_N A_{(1)}^{(\alpha)} + o(\alpha), \\ S_D^{(\alpha)} &= \eta_S A_{(1)}^{(\alpha)} + o(\alpha); \end{aligned} \quad (11.10)$$

$\eta_N$  and  $\eta_S$  are material constants given by

$$\eta_I = - \int_0^\infty s m_I(s) ds, \quad I = N, S. \quad (11.11)$$

Furthermore, by putting  $n=2$  in (11.5) and employing (9.14), one can calculate  $N_D^{(\alpha)}$  and  $S_D^{(\alpha)}$  to within an error of only  $o(\alpha^2)$ ; we find

$$\begin{aligned} N_D^{(\alpha)} &= \zeta_N^{(\alpha)} \mathbf{1} + \eta_N A_{(1)}^{(\alpha)} + \beta_N A_{(1)}^{(\alpha)^2} + \gamma_N A_{(2)}^{(\alpha)} + o(\alpha^2), \\ S_D^{(\alpha)} &= \zeta_S^{(\alpha)} \mathbf{1} + \eta_S A_{(1)}^{(\alpha)} + \beta_S A_{(1)}^{(\alpha)^2} + \gamma_N A_{(2)}^{(\alpha)} + o(\alpha^2). \end{aligned} \quad (11.12)$$

\* [1955, 1], [1958, 1].

\*\* See Section 3.

\*\*\* Under the change of frame characterized by  $\mathcal{Q}$ ,  $A_{(n)} \rightarrow \mathcal{Q} A_{(n)} \mathcal{Q}^{-1}$ .

\*\*\*\* The results (11.10)<sub>2</sub> and (11.12)<sub>2</sub> were obtained by COLEMAN & NOLL [1960, 1], §7. They pointed out that (11.10)<sub>2</sub> justifies the intuitive notion that in slow flows the behavior of a general, non-linear, viscoelastic fluid should be locally approximated by that of a Newtonian fluid. COLEMAN & MARKOVITZ [1964, 2] made the observation that  $\gamma_S$  and  $\beta_S$ , in (11.12)<sub>2</sub>, are related to  $m_S$  and  $a_S$  as shown in (11.13) and (11.14).

The material constants  $\gamma_N$  and  $\gamma_S$  are given by

$$\gamma_I = \frac{1}{2} \int_0^{\infty} s^2 m_I(s) ds, \quad I = N, S. \quad (11.13)$$

The constants  $\beta_N$  and  $\beta_S$  are determined by the quadratic functionals  $\delta_2 \mathcal{N}$  and  $\delta^2 \mathcal{S}$ , and, when these functionals have an integral form, it follows from (9.15) that

$$\beta_I = \int_0^{\infty} \int_0^{\infty} s_1 s_2 a_I(s_1, s_2) ds_1 ds_2, \quad I = N, S. \quad (11.14)$$

The numbers  $\zeta_N^{(\alpha)}$  and  $\zeta_S^{(\alpha)}$  are normalizers chosen so that  $\text{tr } N_D^{(\alpha)} = \text{tr } S_D^{(\alpha)} = 0$ . (No such normalizers are needed in (11.10), because the condition that the flow be isochoric is equivalent to the condition that  $\text{tr } A_{(1)} = 0$ .) Employing the fact that in an isochoric motion  $\text{tr } A_{(2)} = \text{tr } A_{(1)}^2$ , one may easily show that

$$3\zeta_I^{(\alpha)} = -(\beta_I + \gamma_I) \text{tr } A_{(2)}^{(\alpha)} = -(\beta_I + \gamma_I) \text{tr } A_{(1)}^{(\alpha)2}, \quad I = N, S. \quad (11.15)$$

Motivated by the results given in equations (11.10) and (11.12), we employ the following terminology. An incompressible fluid obeying the constitutive equations

$$N_D = 2\eta_N D, \quad S_D = 2\eta_S D \quad (11.16)$$

is called a *first-order fluid*. An incompressible fluid for which

$$\begin{aligned} N_D &= \zeta_N \mathbf{1} + \eta_N A_{(1)} + \beta_N A_{(1)}^2 + \gamma_N A_{(2)}, \\ S_D &= \zeta_S \mathbf{1} + \eta_S A_{(1)} + \beta_S A_{(1)}^2 + \gamma_S A_{(2)}, \\ \zeta_I &= -\frac{1}{3}(\beta_I + \gamma_I) \text{tr } A_{(2)}, \quad I = N, S \end{aligned} \quad (11.17)$$

is called a *second-order fluid*.

It is clear that the classical theory of streaming birefringence, which we outlined in Section 3, is just the theory of first-order fluids. The equations (11.10) tell us that, within the present general framework, the classical theory is indeed valid “in the limit of slow flow” to within an error  $o(\alpha)$ . The sentence containing the relation (3.4) may here be rendered precise as follows:

$$N_D^{(\alpha)} = \frac{\eta_N}{\eta_S} S_D^{(\alpha)} + O(\alpha^2). \quad (11.18)$$

Thus, *the deviators of the stress and refraction tensors, being both proportional to the rate of deformation tensor, are proportional to each other in very slow motions*. Unfortunately, this proportionality, although it is often employed to calculate stresses from measurement of birefringence in viscoelastic flows, is valid only in the “Newtonian limit” shown in (11.16) and does not hold with precision even for second-order fluids. On looking at (11.12) we see that  $S_D^{(\alpha)}$  and  $N_D^{(\alpha)}$  are not proportional when terms  $O(\alpha^2)$  are taken into account.

The relation of the results of this section to those of the previous two sections may be explained as follows. For a general incompressible simple fluid with fading memory, the terms exhibited in equations (9.12) and (9.14) give good

approximations for  $N_D$  and  $S_D$  whenever the norm  $\|J^t\|_h$  is small. If the norm is small because the motion has small amplitude (regardless of its speed), *i.e.* because there exists a fixed reference configuration  $\mathcal{R}$  which renders small  $\varepsilon$  in (10.1), then (9.12) reduces to (10.6), which shows that a theory analogous to Boltzmann's theory of linear viscoelasticity holds here to within an error  $O(\varepsilon^2)$ . If, on the other hand, the norm  $\|J^t\|_h$  is small because the motion is slow, *i.e.* because the history can be regarded as retardation with small  $\alpha$ , then (9.12) reduces to (11.10), and (9.14) reduces to (11.12), which shows that while the classical theory (11.16) gives  $N_D$  and  $S_D$  to within an error  $o(\alpha)$ , the theory of second-order fluids gives  $N_D$  and  $S_D$  to within an error  $o(\alpha^2)$ . Of particular importance are the equations (11.11) and (11.13) which can be used to relate the material constants  $\eta_I$  and  $\gamma_I$  of the slow-flow approximations to the material functions  $G_I$  of the Boltzmann-type approximations. We shall discuss the equations (11.11) and (11.13) in Part c of this section, but first we should like to suggest a direct experimental method of determining  $\eta_N$  and  $\gamma_N$ .

*b) Measurement of  $\eta_N$  and  $\gamma_N$  in Steady Couette Flow*

In a viscometric flow, the components of tensors  $A_{(1)}$  and  $A_{(2)}$  relative to a canonical basis  $h_i$  are

$$[h_i \cdot A_{(1)} h_j] = \kappa \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad [h_i \cdot A_{(2)} h_j] = 2\kappa^2 \begin{bmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad (11.19)$$

with  $\kappa$  the rate of shear defined in (7.10). On putting (11.19) into (11.17), we find that, for a second-order fluid in a viscometric flow, the components of  $N$  relative to  $h_i$  obey the relations

$$\begin{aligned} N_{12} &= \eta_N \kappa, \\ N_{11} - N_{22} &= 2\gamma_N \kappa^2, \\ N_{22} - N_{33} &= \beta_N \kappa^2, \end{aligned} \quad (11.20)$$

and, of course, similar relations\* hold for the components of  $S$ . Clearly, each retardation of a viscometric flow is, in turn, a viscometric flow with a rate of shear that is proportional to the retardation factor  $\alpha$ ; *i.e.* small  $\kappa$  corresponds to small  $\alpha$ . It therefore follows from (11.12) and (11.20) that, for a general simple fluid with fading memory,

$$\begin{aligned} \eta_N &= \lim_{\kappa \rightarrow 0} \frac{\tau_N(\kappa)}{\kappa}, \\ \gamma_N &= \frac{1}{2} \lim_{\kappa \rightarrow 0} \frac{\sigma_N^{(1)}(\kappa)}{\kappa^2}, \\ \beta_N &= \lim_{\kappa \rightarrow 0} \frac{\sigma_N^{(2)}(\kappa)}{\kappa^2}, \end{aligned} \quad (11.21)$$

where  $\tau_N^{(1)}$ ,  $\sigma_N^{(1)}$ , and  $\sigma_N^{(2)}$  are the material functions defined through the equations (7.2) and (7.11). In Section 8 we discussed steady Couette flow and related

\* Cf. COLEMAN & NOLL [1961, 1], p. 713; MARKOVITZ & COLEMAN [1964, 3], p. 78.

the measurable birefringence  $\Delta$  and extinction angle  $\chi$  to the functions  $\tau_N$  and  $\sigma_N$ . In view of the equations (8.28) [see also (7.12) and (7.13)], the relations (11.21) imply that in the limit of low rates of shearing

$$\Delta = 2\eta_N \kappa + o(\kappa), \quad (11.22)$$

$$\cot 2\chi = \frac{\gamma_N}{\eta_N} \kappa + o(\kappa). \quad (11.23)$$

Hence, if one measures  $\Delta$  and  $\chi$  in steady Couette flow of a simple fluid with fading memory, one should find that plots of  $\Delta$  versus  $\kappa$  and  $\cot 2\chi$  versus  $\kappa$  should, in the limit as  $\kappa$  goes to zero, yield straight lines with slopes  $2\eta_N$  and  $\gamma_N/\eta_N$ , respectively. Employing the Maclaurin expansion of the function  $\cot^{-1}$ , one may derive from (11.23) the simpler relation

$$\lim_{\kappa \rightarrow 0} \frac{\frac{\pi}{2} - 2\chi}{\kappa} = \frac{\gamma_N}{\eta_N}. \quad (11.24)$$

We shall not review here the various methods which have been proposed for determining  $\eta_S$ ,  $\beta_S$ , and  $\gamma_S$ .\*

c) *The Relation of  $G_N$  to  $\gamma_N$  and  $\eta_N$*

It follows from the definition of  $m_I$ , that the function with values  $\mathbf{1}m_I(s)h(s)^{-2}$  must belong to  $\mathfrak{S}$ , i.e. that

$$A_I \stackrel{\text{def}}{=} \frac{1}{2} \|\mathbf{1}m_I h^{-2}\|_h^2 = \int_0^\infty m_I(s)^2 h(s)^{-2} ds$$

must be finite. Since  $h$  is, by assumption, an influence function of order greater than 2, there exist numbers  $\varepsilon > 0$ ,  $B > 0$ , and  $M \geq 0$  such that

$$h(s) < B s^{-(2+\frac{1}{2}+\varepsilon)}$$

for all  $s > M$ , and thus

$$0 < \int_\zeta^\infty m_I(s)^2 s^{5+2\varepsilon} ds < A_I B^2 < \infty,$$

for all  $\zeta > M$ . But, by (10.7) and the inequality of Schwarz, we have

$$\begin{aligned} G_I(\zeta)^2 &= \left| \int_\zeta^\infty m_I(s) ds \right|^2 \leq \int_\zeta^\infty (s^{-(5+2\varepsilon)/2})^2 ds \int_\zeta^\infty (s^{(5+2\varepsilon)/2})^2 m_I(s)^2 ds \\ &= \frac{1}{(4+2\varepsilon)\zeta^{4+2\varepsilon}} \int_\zeta^\infty m_I(s)^2 s^{5+2\varepsilon} ds, \end{aligned}$$

and hence

$$G_I(\zeta)^2 < \frac{A_I B^2}{(4+2\varepsilon)\zeta^{4+2\varepsilon}},$$

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\* Methods for  $\beta_S$  and  $\gamma_S$  are suggested in [1961, 1], [1964, 2], and [1964, 3]; for a survey see [1964, 5].

for all  $\zeta > M$ . This shows that, in the limit of large  $s$ ,

$$\mathbf{G}_I(s) = O\left(\frac{1}{s^{2+\varepsilon}}\right), \quad I = N, S. \quad (11.25)$$

In particular,

$$\lim_{s \rightarrow \infty} s^2 \mathbf{G}_I(s) = 0, \quad (11.26)$$

and if, in (11.11) and (11.13), we replace  $m_I(s)$  by  $d\mathbf{G}_I(s)/ds$ , we obtain, after integration by parts, \*

$$\eta_N = \int_0^{\infty} \mathbf{G}_N(s) ds, \quad (11.27)$$

$$\gamma_N = - \int_0^{\infty} s \mathbf{G}_N(s) ds, \quad (11.28)$$

$$\eta_S = \int_0^{\infty} \mathbf{G}_S(s) ds, \quad (11.29)$$

$$\gamma_S = - \int_0^{\infty} s \mathbf{G}_S(s) ds. \quad (11.30)$$

Employing the functions  $\mathbf{G}'_I$  and  $\mathbf{G}''_I$ , defined in (10.21) and (10.22), one can write (11.27)–(11.30) in the forms:

$$\eta_N = \lim_{\omega \rightarrow 0} \frac{\mathbf{G}''_N(\omega)}{\omega}, \quad (11.31)$$

$$\gamma_N = - \lim_{\omega \rightarrow 0} \frac{\mathbf{G}'_N(\omega)}{\omega^2}, \quad (11.32)$$

$$\eta_S = \lim_{\omega \rightarrow 0} \frac{\mathbf{G}''_S(\omega)}{\omega}, \quad (11.33)$$

$$\gamma_S = - \lim_{\omega \rightarrow 0} \frac{\mathbf{G}'_S(\omega)}{\omega^2}. \quad (11.34)$$

We call attention to the known equation (11.34), \*\* which relates  $\gamma_S$  to the shear storage modulus, and to the new equations (11.31) and (11.32), which relate  $\eta_N$  and  $\gamma_N$  to, respectively, the out-of-phase and the in-phase strain-optical coefficients. The equation (11.31), like equation (11.33), is not at all surprising. But the equation (11.32), like its mechanical analogue (11.34), appears to be a special consequence of the notion of “fluidity” employed in the theory of simple fluids, and an experimental verification of it would be desirable; this is particularly so when one considers that, albeit experimenters have found it difficult to test (11.34), \*\*\*

\* The equations (11.27) and (11.28) are new. Although relations equivalent to (11.29) occur frequently in rheology, the equation (11.30) is not very old, having been derived first by COLEMAN & MARKOVITZ [1964, 2], eq. (2.23b); [1964, 5], eq. (6.11b).

\*\* COLEMAN & MARKOVITZ [1964, 2], eq. (2.34); [1964, 5], eq. (6.22).

\*\*\* In conversations with Messrs. L. J. ZAPAS and H. MARKOVITZ we have been advised that the difficulty lies in the uncertainties involved in the usual methods of measuring differences in normal stresses.

seemingly feasible tests of (11.32) are readily designed; one such test is outlined below.

To test (11.32), one would construct apparatus capable of imposing on the fluid harmonic Couette flows of the type discussed in Part d of Section 10. One would operate the apparatus in such a way that the flow (10.32) is achieved with  $\varepsilon$  [see (10.36)] small enough that the terms  $O(\varepsilon^2)$  can be neglected in (10.37) and (10.39). That is, one would see to it that, for each value of  $\omega$  employed, the number  $A$  in (10.42) is sufficiently small that measurement of the birefringence  $\Delta$  yields a sinusoidal result of the type (10.44) with the amplitude  $\Delta^\circ$  proportional to  $A$  and the phase difference  $\delta_N - \delta_E$  independent of  $A$ . From the observed dependence of  $\delta_N - \delta_E$  and  $\Delta^\circ$  on  $\omega$ , for small  $\omega$ , one would calculate the functions  $G'_N$  and  $G''_N$ , via the equations (10.41) and (10.45), and extrapolate the quantities  $G'_N(\omega)/\omega^2$ ,  $G''_N(\omega)/\omega$  to  $\omega=0$  to obtain \*

$$\eta_N^T \stackrel{\text{def}}{=} \lim_{\omega \rightarrow 0} G''_N(\omega)/\omega, \quad \gamma_N^T \stackrel{\text{def}}{=} -\lim_{\omega \rightarrow 0} G'_N(\omega)/\omega^2. \quad (11.35)$$

After having obtained  $\eta_N^T$  and  $\gamma_N^T$ , one would place the fluid in a conventional streaming birefringence apparatus which maintains a steady Couette flow and permits measurements of  $\chi$  as a function of  $\kappa$ . By extrapolating the observed values of

$$\frac{\frac{\pi}{2} - 2\chi(\kappa)}{\kappa}$$

to zero, one would obtain the ratio  $\gamma_N/\eta_N$ , as shown in (11.24); if (11.31) and (11.32) hold, this value of  $\gamma_N/\eta_N$ , obtained from measurements of  $\chi(\kappa)$  in steady Couette flow, should equal the value of  $\gamma_N^T/\eta_N^T$ , obtained, via (11.35), from measurements of  $\Delta$  in harmonic Couette flow.

Of course, if one measures also the birefringence  $\Delta$  as a function of  $\kappa$  in steady Couette flow, then one can obtain both  $\eta_N$  and  $\gamma_N$ , via (11.22) and (11.24), and then check to see if both  $\gamma_N^T = \gamma_N$  and  $\eta_N^T = \eta_N$ .

## 12. On Non-Steady Shearing Flows in the Second-Order Theory of Viscoelasticity

The theory which results from neglect of the terms  $o(\|\mathbf{J}^t\|_h^2)$  in (9.14) may be called the *theory of second-order viscoelasticity*. If one inserts (7.1) into (9.14), one finds that, for a simple fluid with fading memory in a general shearing flow with canonical basis  $\mathbf{h}_i$ , the non-zero components of  $\mathbf{N}$  and  $\mathbf{S}$  relative to  $\mathbf{h}_i$  must obey the relations

$$\begin{aligned} N_{12}(t) &= \int_0^\infty m_N(s) \zeta^t(s) ds + o(\|\zeta^t\|_h^2), \\ N_{11}(t) - N_{22}(t) &= \int_0^\infty m_N(s) \zeta^t(s)^2 ds + o(\|\zeta^t\|_h^2), \\ N_{22}(t) - N_{33}(t) &= \varphi_N(\zeta^t, \zeta^t) + o(\|\zeta^t\|_h^2), \end{aligned} \quad (12.1)$$

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\* The superscript T means that the numbers  $\eta_N^T$  and  $\gamma_N^T$  defined in (11.35) are "tentative values" of  $\eta_N$  and  $\gamma_N$  to be checked against independent measurements of the ratio  $\gamma_N/\eta_N$ .

$$S_{12}(t) = \int_0^{\infty} m_S(s) \zeta^t(s) ds + o(\|\zeta^t\|_h^2),$$

$$S_{11}(t) - S_{22}(t) = \int_0^{\infty} m_S(s) \zeta^t(s)^2 ds + o(\|\zeta^t\|_h^2), \quad (12.2)$$

$$S_{22}(t) - S_{33}(t) = \varphi_S(\zeta^t, \zeta^t) + o(\|\zeta^t\|_h^2).$$

Here

$$\|\zeta^t\|_h^2 \stackrel{\text{def}}{=} \int_0^{\infty} \zeta^t(s)^2 h(s)^2 ds, \quad (12.3)$$

and  $\varphi_I$  is, for  $I=N, S$ , a real-valued bilinear functional which, when (9.15) holds, has the form

$$\varphi_I(\zeta^t, \zeta^t) = \int_0^{\infty} \int_0^{\infty} a_I(s_1, s_2) \zeta^t(s_1) \zeta^t(s_2) ds_1 ds_2. \quad (12.4)$$

In view of (10.7), it follows from (12.1)<sub>1&2</sub> and (12.2)<sub>1&2</sub> that the functionals  $\varphi_I$  and  $w_I^{(1)}$  in (7.2) and (7.3) obey the relations,

$$\varphi_I(\zeta^t) = \int_0^{\infty} \frac{dG_I(s)}{ds} \zeta^t(s) ds + o(\|\zeta^t\|_h^2),$$

$$w_I^{(1)}(\zeta^t) = \int_0^{\infty} \frac{dG_I(s)}{ds} \zeta^t(s)^2 ds + o(\|\zeta^t\|_h^2), \quad I=N, S. \quad (12.5)$$

We find it remarkable that (for  $I=N, S$ ) the scalar function  $G_I$ , which occurs already in the linear theory of viscoelasticity, determines not only  $\varphi_I(\zeta^t)$ , but also  $w_I^{(1)}(\zeta^t)$ , to within an error  $o(\|\zeta^t\|_h^2)$ .

Relations equivalent to (12.2) were first derived by COLEMAN & NOLL [1961, 3] under assumptions about the motion only slightly less general than the present. They observed, as we have, that to within the approximations behind the theory of second-order viscoelasticity, knowledge of *one* scalar material function,  $m_S = dG_S/ds$ , suffices for the calculation of *both* the shear stress  $S_{12}(t)$  and the normal stress difference  $S_{11}(t) - S_{22}(t)$ . They suggested that an experimenter could test this consequence of (12.2)<sub>1&2</sub> by studying the harmonic Couette flow (10.32). To do so one would first measure the torque  $M$  required to maintain the flow, working, of course, in a range in which  $M$  obeys (10.46) and (10.47); thus one would obtain  $G'_S(\omega)$  and  $G''_S(\omega)$ . It follows from (12.2) that, to within an error  $o(\|\zeta^t\|_h^2)$ , the difference in the normal thrusts on the inner and outer bounding cylinders of a Couette apparatus obeys the formula \*

$$S_{r,r}(R_2) - S_{r,r}(R_1) = \frac{R_2 - R_1}{R_1} \int_0^{\infty} \frac{dG_S(s)}{ds} \zeta^t(s)^2 ds + O\left(\left[\frac{R_2 - R_1}{R_1}\right]^2\right), \quad (12.6)$$

\* [1961, 3] eq. (25). It is assumed here that one bounding cylinder rotates and the other is at rest; otherwise the contribution,  $-\int_{R_1}^{R_2} \rho r v(r)^2 dr$ , of the centrifugal force to  $S_{r,r}(R_2) - S_{r,r}(R_1)$  is not  $O\left(\left[\frac{R_1 - R_2}{R_1}\right]^2\right)$ .



with  $\zeta^t$  as in (8.9). In a Couette flow of the type (10.32), the function  $\zeta^t$  is given (exactly) by

$$\zeta^t(s) = 2E_{r\theta}^0 [\sin(\omega t - \omega s + \delta_E) - \sin(\omega t + \delta_E)], \quad s \in [0, \infty), \quad (12.7)$$

with  $E_{r\theta}^0 = \frac{1}{2} \text{rd } \alpha(r)/dr$ , as in (10.38). Hence

$$\begin{aligned} \zeta^t(s)^2 = 4E_{r\theta}^{0^2} & \left[ \frac{1}{2} [1 - \cos(2\psi) \cos(2\omega s) - \sin(2\psi) \sin(2\omega s)] \right. \\ & \left. - 2 \sin^2 \psi \cos(\omega s) + 2 \sin \psi \cos \psi \sin(\omega s) + \sin^2 \psi \right], \quad \psi \stackrel{\text{def}}{=} \omega t + \delta_E. \end{aligned} \quad (12.8)$$

Placing (12.8) in (12.6), we find, after several integrations and rearrangements of terms, that, to within an error  $o(\|\zeta^t\|_h^2)$ ,

$$\begin{aligned} S_{r,r}(R_2) - S_{r,r}(R_1) \\ = 2E_{r\theta}^{0^2} \frac{R_2 - R_1}{R_1} & \left[ [2G'_s(\omega) - G'_s(2\omega)] \cos(2\omega t + 2\delta_E) \right. \\ & \left. + [G''_s(2\omega) - 2G''_s(\omega)] \sin(2\omega t + 2\delta_E) - 2G_s(\omega) \right] + O \left( \left[ \frac{R_1 - R_2}{R_1} \right]^2 \right), \end{aligned} \quad (12.9)$$

a relation which should be experimentally verifiable. Since (12.7) and (12.3) imply that  $\|\zeta^t\|_h$  is  $O(|E_{r\theta}^0|)$ , the relation (12.9) holds to within an error  $o(|E_{r\theta}^0|^2)$ . COLEMAN & DILL will discuss relations of the type (12.6) and (12.9) at greater length in a future article. Our interest here is in the possibility of verifying (12.5) for the new case  $I=N$ .

By (7.8) and (12.5) with  $I=N$ , in an arbitrary shearing flow of a simple fluid with fading memory

$$\cot 2\chi = \frac{\int_0^\infty \frac{dG_N(s)}{ds} \zeta^t(s)^2 ds}{2 \int_0^\infty \frac{dG_N(s)}{ds} \zeta^t(s) ds} + o(\|\zeta^t\|_h), \quad (12.10)$$

with  $\chi \in [0, \pi/2)$  the counterclockwise angle from  $\mathbf{h}_1$  to  $\mathbf{k}_1$ . In view of (12.10), (12.5), and (7.9), we have, for the birefringence  $\Delta = n_1 - n_2$ , the relation

$$\Delta = 2 \int_0^\infty \frac{dG_N(s)}{ds} \zeta^t(s) ds + o(\|\zeta^t\|_h^2). \quad (12.11)$$

It is interesting that the error term here is  $o(\|\zeta^t\|_h^2)$ , rather than just  $o(\|\zeta^t\|_h)$ . Using (12.11), one may conclude, for example, that the expressions derived in Part d of Section 10 for the birefringence in harmonic Couette flow remain valid when terms  $O(|E_{r\theta}^0|^2)$  are taken into account; *i.e.*

$$\Delta = \Delta^\circ \sin(\omega t + \delta_N) + o(|E_{r\theta}^0|^2) \quad (12.12)$$

with  $\delta_N$  and  $\Delta^\circ$  given again by (10.41) and (10.45). It is clear from (12.10), however, that the conclusion that  $\chi$  equals  $\pi/4$  is valid only to within an error  $o(1)$ ; the first departure of  $\chi$  from  $\pi/4$  is  $O(\|\zeta^t\|_h)$  or, in harmonic Couette flow,  $O(|E_{r\theta}^0|)$ .\*

\* A result of this type was found already in equation (11.23) which tells us that in slow viscometric flows the departure of  $\chi$  from  $\pi/4$  is linear in  $\kappa$ .

Substitution of (12.7) and (12.8) into (12.10) yields the following expression for the extinction angle  $\chi$  in a harmonic Couette flow (10.32), under neglect of terms  $o(E_r^2)$ :

$$\cot 2\chi = \frac{1}{2} E_r^2 \frac{[2G'_N(\omega) - G''_N(2\omega)] \cos(2\omega t + 2\delta_E) + [G''_N(2\omega) - 2G'_N(\omega)] \sin(\omega t + \delta_E) - 2G'_N(\omega)}{G'_N(\omega) \sin(\omega t + \delta_E) + G''_N(\omega) \cos(\omega t + \delta_E)} \quad (12.13)$$

If  $G'_N$  and  $G''_N$  are obtained first from measurements of  $\Delta$ , as discussed after equation (10.45), then the equation (12.13) should be experimentally verifiable.

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