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The development of theory for flow and dynamic effects for nematic liquid crystals†

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Today studies of flow phenomena in nematic systems normally employ the so-called Leslie–Ericksen theory. This theory was formulated in its present form in the 1960s, and it represents the culmination of theoretical developments that essentially started at the beginning of this century. This article gives an account of the evolution of the theory.

1. Introduction

Since Reinitzer [1] discovered the liquid crystalline state in 1888 a little more than a century has passed. During these years a vast number of new liquid crystalline phases have been discovered and an intensive study, from the experimental as well as from the theoretical point of view, has been performed on these; today the study of liquid crystals is a well established branch of modern science. When studying the visco-elastic behaviour of nematic liquid crystals, the so-called Leslie–Ericksen theory [2, 3] is the theory that is normally employed. In this article we discuss how different theoretical models for the static and dynamic behaviour of nematics ultimately converged into this theory. We want to gain some insight into the historical development of the theory, and the goal is to shed some light on questions such as ‘why did a certain researcher contribute with a certain theory at a certain time?’. To do so one must compare the level to which the development of theoretical physics had reached at the time when a certain theory was proposed.

The first serious attempt to derive a theoretical model of liquid crystals was presented by Bose [4–6] in a series of articles in 1907–1909. These papers were mainly qualitative and contained discussions employing the recently developed statistical physics by Boltzmann. The core of the theory by Bose is the swarm theory, i.e. the idea that liquid crystals consist of small domains

with diameter in the micron range, within which the director stays constant. The swarm theory dominated the theoretical modelling of liquid crystals for several decades, and in fact one of the key questions discussed during the famous discussions of the Faraday Society in 1933 [7] was the existence of swarms. It is interesting to note what de Gennes writes about swarms in his book [8] ‘...the so-called swarm theory which has plagued the field of nematic liquid crystals for thirty years’.

In 1916 Born presented the first quantitative theory of liquid crystals [9]. He assumed that the origin of liquid crystalline phases was interactions between permanent dipoles attached to the molecules in the liquid, an assumption which we know today is wrong [10]. By symmetry arguments Oseen could show that Born’s model was incorrect and in a series of articles [11, 12] Oseen derived a theoretical description of the liquid crystalline phases that is essentially correct regarding their static behaviour. However, Oseen initially was also one of the advocates of the swarm theory and in his own words [13] the theory by Oseen puts the mathematical–physical lighting on Bose’s hypothesis. In 1933 one of Oseen’s students, Anzelius, published the first attempt [14] at a dynamic theory for the nematic phase. The theory by Anzelius, however, does not describe the dynamical behaviour of nematics correctly, even if it contains many elements which in retrospect can be proven to be sound. Following the development of the modern theory of rational mechanics in the fifties it was possible to reformulate a correct version of Anzelius’s theory, finally establishing the dynamic theory of nematics

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as it is known today, the Leslie–Ericksen theory [2, 3]. This article gives an account of how the development that finally produced this theory took place.

2. Carl Wilhelm Oseen

Carl Wilhelm Oseen [15] was born on 17 April 1879 in Lund, Sweden. He was from a family with a strong scientific tradition. Already in school he read a French textbook on mechanics after which he expressed ‘I learned to know the strange joy of the French mathematical–mechanics literature, i.e. the great clearness’. Oseen started his university studies in Lund during the autumn of 1896, and passed his first exam in mathematics, mechanics, astronomy, theoretical and practical philosophy in December 1897. In April 1900 he obtained his master degree in mathematics and mechanics, the subject of which was the generalization of work done by Sophus Lie. Lie had formulated a problem of contact transformations and solved two special cases of this problem himself. Oseen succeeded in solving the corresponding general problem, a work which became his diploma work: ‘Über die allgemeinste Abbildung der geodätischen Kreise einer Fläche durch Berührungstransformationen’.

The winter 1900–1901 Oseen spent in Göttingen where the leading mathematicians at the time were Felix Klein and David Hilbert. The strongest impressions from

his visit to Göttingen, Oseen later said, were a series of lectures by Hilbert on partial differential equations. In December 1901 Oseen was awarded his doctor’s degree for a dissertation consisting of a group theoretical classification of contact transformations: ‘Über die endlichen, kontinuierlichen, irreduciblen Berührungstransformationsgruppen im Raume’. After the dissertation he worked as a researcher and teacher at the University of Lund until in 1909 he became a professor of mechanics and mathematical physics at the University of Uppsala. It was during this period that he started to gain interest in the subject where he made his most remarkable contributions, hydrodynamics. Already during his first visit to Göttingen, Oseen later told how often he stopped on one of the bridges over the river Leine to study the eddies of the flowing water. Perhaps it is from this time that his devotion to master the mathematics of hydrodynamics emanated.

One example of Oseen’s work on hydrodynamics is the resolution of the so-called d’Alembert’s paradox. During the nineteenth century the Navier–Stokes equations, which are the equations of motion regulating the flow of viscous media, had been established. Due to the non-linear character of these equations they were still, at this time, not very well studied except in some cases where severe approximations had been made. For instance, Stokes, by neglecting the non-linear inertial terms in the equations, had calculated the drag force acting on a sphere with radius a moving with velocity v through a liquid with viscosity μ to be

$$F = 6\pi\mu av. \quad (1)$$

Another outcome of Stokes’s calculation is the result that the velocity field of the liquid around the sphere is symmetric. The consequence of this calculation is that for an ideal liquid, i.e. if $\mu = 0$, the drag force disappears and the velocity field is regular, without a wake. This is obviously incorrect as Oseen had already concluded from the bridges in Göttingen. Contrary to what Stokes had done, when studying this problem, Oseen kept the non-linearities in the calculations. One delicate problem when studying a liquid in the limit of zero viscosity is when to take the limit $\mu \rightarrow 0$. The authors before Oseen had taken this limit before solving the equations, while Oseen did the reverse, performing the limit $\mu \rightarrow 0$ only at the end of the calculations. In this way Oseen succeeded in calculating a better approximation of the velocity field around the sphere, as well as the drag force, and for the first time the solution to the problem indicated a non-symmetric flow field, containing a tail of eddies in the wake of the flow. The drag force in Oseen’s calculation was found to be

$$F = 6\pi\mu av \left(1 + \frac{3\rho a|v|}{8\mu} \right) \quad (2)$$

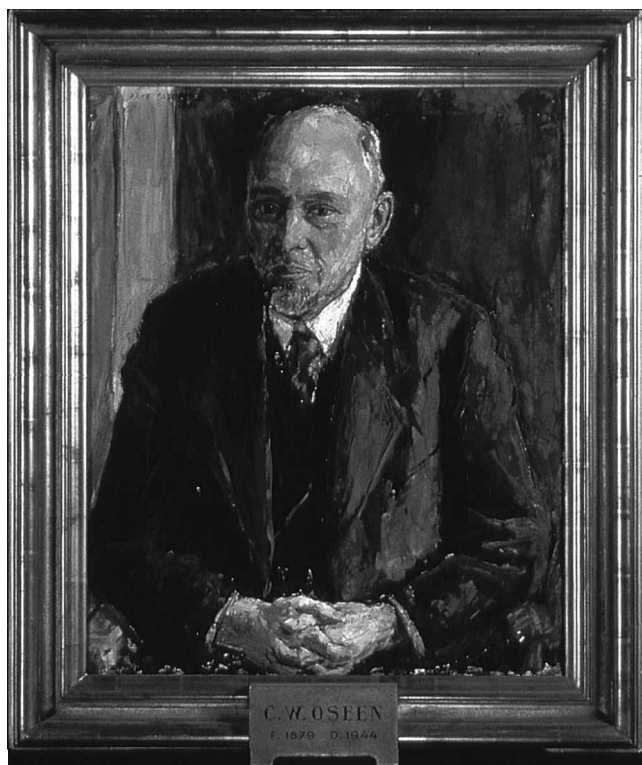


Figure 1. Carl Wilhelm Oseen in 1938. Oil painting by Arne Cassel.

with ρ being the density of the liquid, a result which is non-zero also in the limit $\mu \rightarrow 0$. One notices that Oseen's result is a first correction of Stokes's formula, the correction term of which can be written as $3R/16$, where

$$R = \frac{2\rho a|v|}{\mu} \quad (3)$$

is the Reynolds number. Oseen's work on hydrodynamics, which to a large extent was concentrated on the study of non-linear solutions of Navier–Stokes equations is summarized in his book [16] 'Neuere Methoden und Ergebnisse in der Hydrodynamik', a book which for a long time was one of the standard books in hydrodynamics.

In addition to his work on hydrodynamics, Oseen contributed to many branches of modern physics. In 1915 he published a paper [17] *Über die Wechselwirkung zwischen zwei elektrischen Dipolen und über die Drehung der Polarisationssebene in Kristallen und Flüssigkeiten*. This paper was published at the same time as a similar paper by Born and presented the first satisfactory theory of the rotation of polarized light and optical activity. The paper also contained a highly appreciated treatment of dispersion, similar to the one Ewald later used as a basis for his research within the field of crystallography. Oseen also contributed to the mathematical treatment of relativity and quantum theory. He even tried, although unsuccessfully, to develop a consistent theory of quantum-electrodynamics.

The subject into which Oseen, apart from the hydrodynamics of isotropic media, put most of his effort was the theory of liquid crystals. The only serious theory for liquid crystals which existed around 1920 when Oseen started his investigations was the work by Born [9], who assumed that dipole–dipole interactions between the molecules were responsible for the formation of the liquid crystalline phases. It is easy to understand that Oseen had the right skill and background for undertaking the work to formulate a theory for liquid crystals. He had good knowledge in symmetry analysis and group theory, he was a skilled mathematician who did not hesitate to perform lengthy calculations, he was an expert in hydrodynamics and elasticity, and he had contributed to the field of optics of anisotropic media. During a period of almost twenty years Oseen published more than twenty papers on liquid crystals [18], mostly regarding the optics of twisted nematics as well as the static behaviour of nematics, but he also touched [12, 19] (to a large extent through his student Anzeli) upon dynamic behaviour.

The starting point for Oseen's studies on liquid crystals was the use of symmetry reasoning to prove that Born's model of interaction between the molecules was not

consistent with the symmetry of the system. Instead of Born's dipolar interaction he constructed a model within which the interaction between the molecules is assumed to consist of two parts: attractive forces between the centres of mass between the molecules, and torques which tend to orient the molecules in certain directions with respect to each other. The molecules were assumed to consist of elongated, rigid bodies. One important feature of Oseen's theory is that the potential corresponding to the attractive forces falls faster than $1/r^3$, where r is the distance between the molecules. Applying statistical physics in the sense of Boltzmann, he then succeeded in deriving the first consistent static theory of liquid crystals.

Oseen appears to have been the first to appreciate that one can employ a unit vector field to describe the orientational order in a liquid crystal, thus ignoring variations in the degree of alignment, which presumably are quite small in reasonably well-ordered samples. While this simplified model may have attracted criticism, it does lead to a simpler and therefore more tractable theory. With this assumption he calculated a local stored energy density, which he required to give a minimum energy for the volume of liquid crystal in equilibrium configurations. After some consideration he ultimately adopted for nematics [20] an expression of the form

$$\int_V \rho^2 \{K_{11}(\mathbf{n} \cdot \text{curl } \mathbf{n})^2 + K_{22}(\text{div } \mathbf{n})^2 + K_{33}[(\mathbf{n} \cdot \text{grad})\mathbf{n}]^2\} dV \quad (4a)$$

where ρ denotes density, \mathbf{n} the unit vector field, and V the volume of nematic. However, in an earlier paper [21] he does consider some additional nematic terms, and of particular interest to present day readers are the two

$$\text{div}[(\mathbf{n} \cdot \text{grad})\mathbf{n} - (\text{div } \mathbf{n})\mathbf{n}] \quad \text{and} \quad (\mathbf{n} \cdot \text{grad}) \text{div } \mathbf{n} \quad (4b)$$

which he chooses to discard on the grounds that they lead to surface integrals, and therefore make no contribution to his equilibrium considerations. Thus Oseen did derive all of the terms currently employed in the static energy for nematics, plus some additional terms that we now exclude on grounds of material symmetry, a concept less well established in Oseen's time.

In 1933 Oseen left the professorship at Uppsala to become the first director of the recently founded Nobel Institute of theoretical physics, where he remained for the rest of his life. Although he put much devotion into his research, he had plenty of time left over for his other intellectual interests which included philosophy, history, literature and art. He was not only unusually many-sided, but also very straight-backed. As an example of

this one can mention that although German was the language which besides Swedish he mastered best, due to political developments in Germany around 1932 he stopped writing in German and instead started to publish in French. Oseen died on 7 November 1944.

3. Viscometry in the twenties—Newtonian and non-Newtonian liquids

The classical treatment of fluid mechanics rests upon a hypothesis by Newton, supposing that in a non-uniform flow, fluid layers slide relative to adjacent layers in the direction of the flow and that the relative 'slip' velocity (velocity gradient) between such adjacent layers varies across the flow. From this assumption Newton defined the viscosity μ of a liquid as the ratio between the shear stress τ and the velocity gradient dv/dy

$$\mu = \frac{\tau}{dv/dy}. \quad (5)$$

In this model the viscosity μ of an incompressible liquid is a material constant, depending only on the temperature of the system. A straightforward result of Newton's hypothesis is the derivation of the Navier–Stokes equation, which is the equation of motion governing the flow of an incompressible, Newtonian liquid. This equation can be solved exactly for Poiseuille flow and Couette flow, i.e. the flow through a long tube driven by a pressure gradient and the flow between concentric cylinders driven by the rotation of one of the cylinders with respect to the other. For Poiseuille flow the driving pressure gradient is found to be proportional to the total flux of liquid through the tube, while in the case of Couette flow, a similar proportionality is found between the driving torque applied to the moving cylinder and the corresponding angular velocity. In both cases the proportionality constant is found to be equal to (apart from some geometrical factors depending on the experimental set up) the viscosity of the liquid.

At the beginning of the twentieth century the viscous properties of colloids and more complicated substances such as gelatin, cellulose, lubricants and liquid crystals started to be investigated, and already in 1913 the Faraday Society held a general discussion devoted to such systems [22]. Ostwald reported [23, 24] the viscosity of such substances as depending not only on temperature, but also on a vast number of other variables such as concentration, charge or ionization, thermal and mechanical pretreatment of the sample as well as its age. He also found that the measured viscosity in many cases was irreproducible and apparatus-dependent. Ostwald called these anomalies structural viscosity. Similar behaviour for the viscosity of gelatin, albumen and colloids was reported by Garrett [25] and du Pré Denning [26]. Deviations from Poiseuille's law for the flow of blood

and blood serum was reported by Hess [27], and the complex behaviour of lubricants in contact with metal surfaces was studied in detail by Woog [28] and Trillat [29]. Structural viscosity or, as we say today, non-Newtonian behaviour was also observed early in liquid crystalline systems by, among others, Lehmann [30], Ostwald [31], Svedberg [32] and Vorländer [33]. Ostwald for example had observed deviations from Newtonian behaviour in Couette flow and already in 1900 Lehmann had reported peculiar behaviour in thermally driven flow in a liquid crystal. If a cholesteric liquid crystal, spread out between two glass surfaces, is submitted to a thermal gradient, under some circumstances a violent motion is observed. This motion, as Oseen later explained [12], is not due to the motion of the liquid itself, but due to the uniform rotation of the local molecular axis. In 1925 Kruyt [34] concluded that systems for which the building blocks can be assumed to consist of spherical particles exhibit Newtonian behaviour, while non-Newtonian behaviour is exhibited by systems where the shapes of the building blocks deviate from spheres.

One of the first attempts to derive a theoretical model for the viscous behaviour of nematics was presented by Herzog and Kuda [35, 36]. They divided the total viscosity of polyatomic molecules into two parts, one due to the translational motion and one due to the rotation of the molecules. However, they did not develop their theory much beyond this point, so it is not of much help when studying nematic liquid crystals. Generally, it seems that virtually all experimental observations and rather rare theoretical descriptions of the viscous behaviour of liquid crystals before 1930 neglected to keep track of the behaviour of the director. The first serious attempt to derive a consistent dynamical theory for nematics was presented by Anzelius [14], who generalized the Navier–Stokes equation by incorporating the director, and who realized that one also needs one equation for the conservation of angular momentum in addition to the Navier–Stokes equation. Anzelius was probably also one of the first in realizing the importance of imposing boundary conditions on the director when performing calculations. In the next section Anzelius and his dynamical theory of nematics are presented.

4. Adolf Anzelius

As seen in §2, Oseen devoted much research to the static theory of liquid crystals [18]. He also touched upon the dynamic properties [12, 19], but the first consistent dynamic theory of liquid crystals was put forward by one of Oseen's students, Adolf Anzelius. Anzelius was born on 29 April 1894 in Växjö, Sweden. After studies at the Royal Institute of Technology (School of Mechanical Engineering), he became a civil engineer in 1916. He



Figure 2. Adolf Anzelius at his desk at Chalmers. Anzelius was an eager cigar smoker and according to his students one could decide by the smell whether or not Anzelius was present at the department.

continued studies at the University of Stockholm for another two years and was awarded degrees in astronomy, mathematics and mechanics. During the years 1916–1940 he was a lecturer at the Royal Institute of Technology, teaching topics such as mechanics, water-motions and pumps, heat and powder technology. He also taught mathematics and mechanics part time at the Royal College of Artillery and Engineering Sciences and at the Royal Naval Academy. Anzelius everywhere obtained excellent testimonials for his pedagogical merits.

Although Anzelius during these years had a heavy teaching load, he also had time for research, the main topics of which were within the field of mathematical physics, including the application of potential theory and integral equations to, among other things, boundary value problems for the deformation of elastic bodies, heat conduction and sedimentation problems. In 1931, under the supervision of Oseen, he was awarded the doctor's degree in mechanics at the University of Uppsala for a dissertation [14] with the title *Über die Bewegung der anisotropen Flüssigkeiten*. The thesis, which consists of an eighty-four page booklet, represents the only research which Anzelius published on liquid crystals. Nevertheless, this work was the first serious and consistent attempt to derive a dynamical theory for nematics. As discussed in the last section, at the end of the twenties a vast number of experimental data [22–36] for the dynamic behaviour of anisotropic liquids existed, mainly considering viscometry. Anzelius starts his thesis by presenting a review of these. He then, basing his derivation upon the theory of Oseen, derives a set of dynamical equations, consisting of one equation which is a generalization of the Navier–Stokes equation and one which is an equation of motion for the rotation of the director. By solving these equations he then, for the first time, succeeds in

giving a theoretical explanation of the observed flow behaviour of nematics for some special flow geometries like shear flow, Poiseuille flow and Couette flow. He also presents a short treatment of heat conduction in anisotropic liquids, and in this connection he presents the first theoretical discussion of the Lehmann effect [30]. This attempt by Anzelius to formulate a theory for flow behaviour in liquid crystals was a somewhat ambitious project at that time, given the general level of understanding of continuum models for non-linear flow phenomena in the first half of this century. However, in spite of the difficulties he made a tolerably good attempt, demonstrating that it was possible to model certain aspects in this manner.

Clearly, the Navier–Stokes equations for isotropic liquids had to be extended, the stress now dependent upon the local alignment as well as the rate of strain tensor. Anzelius [14] assumed the existence of a dissipation function, now a function of the rate of strain and the orientation of the anisotropic axis in the liquid crystal, again described by a unit vector field \mathbf{n} . In view of the fact that his theory preceded Tsvetkov's experiment [37] with a rotating magnetic field by a number of years, it was excusable that he did not include the rate of change of the vector field \mathbf{n} in his expression for the dissipation function. In any event, restricting attention to the incompressible case, he found that his dissipation function f must take the form

$$f = A_1 \operatorname{tr} \mathcal{D}^2 + A_2 \mathcal{D}\mathbf{n} \cdot \mathcal{D}\mathbf{n} + A_3 (\mathbf{n} \cdot \mathcal{D}\mathbf{n})^2 \quad (6a)$$

where

$$D_{ij} = \frac{1}{2} \left[\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right]. \quad (6b)$$

Here \mathbf{u} denotes the velocity vector, and A_1 , A_2 and A_3 are constants. The viscous stress tensor or matrix \mathcal{T} then follows from

$$\mathcal{T} = -p\mathcal{I} + \frac{1}{2} \frac{\partial f}{\partial \mathcal{D}} \quad (7)$$

where p is an arbitrary pressure, and so

$$\mathcal{T} = -p\mathcal{I} + A_1 \mathcal{D} + \frac{1}{2} A_2 (\mathcal{D}\mathbf{n} \otimes \mathbf{n} + \mathbf{n} \otimes \mathcal{D}\mathbf{n}) + A_3 \mathbf{n} \cdot \mathcal{D}\mathbf{n} \otimes \mathbf{n} \quad (8)$$

clearly giving a symmetric viscous stress. In the above, $\mathbf{a} \otimes \mathbf{b}$ denotes the 3×3 matrix with (i, j) th element $a_i b_j$.

Given the symmetry of his dynamic stress, Anzelius had to look elsewhere for torques to influence the alignment in flow. Not surprisingly therefore he attempted to generalize Oseen's static theory by adding a viscous

moment, which he chose in the form

$$\begin{aligned} \mathbf{M} = & [(C_1 - C_2) \underline{D}\mathbf{n} + \frac{1}{2}(C_1 + C_2)\mathbf{n} \times \text{curl } \mathbf{u} \\ & + (C_{11} + C_{12})\mathbf{n} \times \underline{D}\mathbf{n} - \frac{1}{2}(C_{11} - C_{12}) \text{curl } \mathbf{u}] \times \mathbf{n} \end{aligned} \quad (9)$$

where C_1 , C_2 , C_{11} and C_{12} are constants. Anzelius does remark that C_{11} and C_{12} must be zero for 'symmetric molecules', and this can be taken as either \mathbf{n} and $-\mathbf{n}$ being equivalent, or the material being nematic. He then adds this to the Euler–Lagrange equation obtained earlier by Oseen. For boundary conditions he adopts the familiar no-slip assumption of hydrodynamics and assumes that the plates dictate a particular orientation of the alignment.

Anzelius was unfortunate in that this formulation preceded the experiments by Miesowicz [38] and Tsvetkov [37], particularly the latter which clearly points to the need to include the time derivative of the alignment vector \mathbf{n} . The inclusion of this quantity would have allowed him the opportunity to make his torque \mathbf{M} properly invariant, but he may not have seen the need for this. Nonetheless, his theory did show that one could analyse flow effects by developing an appropriate generalization of the hydrodynamics for isotropic liquids.

Although Oseen gave Anzelius's theory strong support, he raised some criticism against it [12, 19], the most severe of which was the fact that the work produced by the torque \mathbf{M} was not discussed within the context of conservation of energy. With this as a starting point, Oseen tried to generalize Anzelius's theory, deriving an alternative formulation of the governing equations [19]. However, he never performed any calculations with this alternative theory. Whether this was a consequence of Oseen not believing completely in his version of the dynamic theory, or simply that the resulting equations were too complicated to deal with is beyond the scope of this discussion.

As stated above, Anzelius contributed with only one article to research on liquid crystals. This might partially have been a result of his heavy teaching load, but also probably due to the fact that at the time Anzelius published his theory, research on liquid crystals almost died out for twenty-five years. Anzelius ended his career as a professor in mechanics at Chalmers University of Technology in Göteborg during the years 1945–1960. It is amusing to see what one of the reviewers wrote about Anzelius's dissertation, when Anzelius applied for the professorship at Chalmers: 'The dissertation should have technical importance for lubrication problems for semi-dry fabrication'. After retiring in 1960, Anzelius returned

to Stockholm, where he lived until he died on 3 January 1979.

5. The empty years: 1933–1958

During the first three decades of the twentieth century liquid crystal research flourished. The experimental observations covered a broad range of aspects such as X-ray and electron diffraction studies, chemistry, biology, magnetic and dielectric properties, viscometry, studies of orientational effects of electric and magnetic fields, etc. Gradually, also the theoretical description of the system began to take shape. From Bose's very qualitative discussions [4–6], followed by Born's dipolar model [9], ultimately Oseen presented the first coherent model [11] of liquid crystals, based on a realistic interaction potential between the molecules and treated in the proper way by statistical mechanics. In 1933 the Faraday Society held a general discussion devoted to liquid crystals [7]. Most of the leading scientists within the field of liquid crystals contributed to the 1933 discussions and the proceedings from the meeting provide a nice overview of the status of liquid crystal research at that time. Many of the discussions at the meeting were devoted to the conflict between the swarm theory and distortion theory (continuum model in modern words). The swarm theory, which seemed to have the largest support at this time was first suggested by Bose in a qualitative discussion [4–6] and later put into an exact mathematical theory by Ornstein and Zernike [39] and by Oseen [13]. The core of the swarm theory was the idea that the system consists of a polycrystalline structure of small droplets containing $\sim 100\,000$ molecules, i.e. with a diameter of the order of $0.1\ \mu\text{m}$.

The first contribution at the meeting was the paper by Oseen [12] where he presented a brief review of the extensive work he had performed on the theoretical description of liquid crystals during the preceding fifteen years. Vorländer [40] presented a number of crystallographic results and suggested that liquid crystals could not be treated as a one-, two- or three-dimensional system, but rather as a system of mixed dimensionality. The contribution by Fréedericksz and Zolina [41] was devoted to a long argument in favour of the swarm theory, and also to the experimental observation of the behaviour in a thin slab of a nematic liquid crystal across which an electric field had been applied. Ornstein and Kast [42] presented a contribution entitled *New Arguments for the Swarm Theory of Liquid Crystals*. By a series of arguments they advocate a swarm theory and summarize their results in a number of conclusions of which a few can be mentioned: (1) Liquid crystals are aggregates of sub-microscopic, homogeneous ranges, the so-called swarms, in which the molecules all lie parallel. (2) The structure is to be visualized as of a polycrystalline

nature, the swarms having an individual existence of considerable duration. (3) The swarms lie perfectly irregularly: their axes are distributed at random. (4) Any further arrangements of the swarms into larger homogeneous ranges is caused by external forces.' The contribution by Zocher [43] focused on the controversy between the swarm theory and the distortion theory. In the first part he gives a full mathematical treatment of a Fréedericksz transition in a magnetic field. The governing equation of the system is obtained by balancing the magnetic torque by an intuitive elastic one. However, he does not differentiate between different kinds of elastic deformations, and thus the approach is similar to the one-constant approximation. In the second part experimental data are presented, and from these the elastic constant is determined to be of the order of 10^{-11} N. In the end Zocher decided [44] that swarm theory is inadequate to describe nematic liquid crystals. Föex [45] presented more investigations of the magnetic properties of nematics; among other things, he investigates the orientational effect of a magnetic field as a function of field strength. He also discusses the analogue behaviour of nematics and ferromagnets. Further X-ray studies of liquid crystals were presented by Malkin [46] and Stewart [47], while electron diffraction studies were discussed by Trillat [48]. Dynamical studies, were described by Lawrence [49], who reported the observation of a non-Newtonian velocity profile in Poiseuille flow and by Ostwald [50], who presented a contribution with the title *Anomalous viscosity in mesomorphic melts*. An immature theoretical description of the viscosity of liquid crystals was given by Herzog and Kudar [36]. Finally Bragg, after the meeting, submitted a contribution [51] regarding the mathematical description of focal-conic structures. In the rather lengthy discussion which is reported in the proceedings the arguments for and against swarms is a frequently occurring question.

As seen above, the standard of liquid crystal research in 1933 was excellent. Many distinguished researchers were active within the field, and the general understanding of the system was good. However, it is a mystery that the research on liquid crystals almost died out after the Faraday meeting in 1933. Concerning viscometry, only two publications of interest after the meeting are worth mentioning, both of which were published in 1935 and thus probably on work already performed at the time of the meeting. One of these concerns the famous viscosity measurements by Miesowicz [38] and the other the experiment by Tsvetkov [37], where the rotation of the director under the influence of a rotating magnetic field was studied. The period of relative inactivity lasted until 1958, when another meeting of the Faraday Society [52] devoted part of the discussions to liquid crystals, before the research within this field took off with an

avalanche-like activity. The reason for these twenty-five lost years is hard to understand. It is obvious that the war might provide part of the answer. One can also speculate whether the researchers in 1933 committed the classical mistake of believing that the subject was mature and nothing more was to be added, or if perhaps the swarm theory obstructed the development of the theoretical description to such an extent that researchers abandoned the subject. However, with Frank's celebrated re-formulation [53] of Oseen's elastic theory, liquid crystals re-entered the scientific world and it took less than ten years until the viscoelastic theory as we know it today, the Leslie–Ericksen theory [2, 3], was developed. One of the tools necessary for this development to take place was the modern theory of rational mechanics which had emerged in the early fifties [54–63]. This theory formulates rheological equations of state for non-linear viscoelastic materials and is needed in formulating the proper dynamical equations for anisotropic systems, such as liquid crystals. It is reviewed in the next section.

6. The development of non-linear continuum mechanics

Before the Second World War continuum mechanics consisted of the Navier–Stokes equations for linearly viscous liquids and linear elasticity for solids, and not a great deal more, no great need being then perceived for more general theories. However, the growth of new technology following the Second World War led to an increasing interest in new materials, and the need for theories of solids undergoing finite deformations and of liquids incorporating viscoelastic behaviour began to become apparent. The early developments in this growth of interest in non-linear continuum mechanics essentially began in England, but later much of this activity was to be concentrated in the USA.

Ronald Rivlin was born in London in 1915, and was educated at Cambridge, graduating BA in 1937. Thereafter he joined the Research Laboratories of the General Electric Company prior to war service as a scientific officer at the Ministry of Aircraft Production. After the war he joined the British Rubber Producers Research Association remaining in this employment until 1952, where he was prominent in the development of the theory of non-linear elasticity [54–56], no doubt motivated by the needs of his employers. However, in this period he also wrote an important paper on a theory of non-linear viscous liquids [57]. In 1952 he moved to the USA to the US Naval Research Laboratory in Washington for a year before taking an academic post at Brown University in 1953, where he remained until 1967, then moving to Lehigh in Pennsylvania. Throughout his career in the USA Rivlin has been a leading figure in continuum mechanics, best known for his work on

non-linear theories of solids and viscoelastic materials, and noted for many outstanding contributions.

James Oldroyd was born in Yorkshire in England in 1921. His education at Cambridge was interrupted by war service in the Ministry of Supply, but at the end of hostilities he joined Courtaulds Ltd as a research scientist, although continuing his association with Cambridge, being a Fellow of Trinity College during the period 1947–51 and obtaining his PhD in 1949. His work at Courtaulds led to his interest in the formulation of non-linear theories to model the spinning of artificial fibres. In 1950 he published an important paper in the *Proceedings of the Royal Society* concerning the formulation of relationships between stress and deformation in non-linear continuum theories [58], which had much influence upon thinking in this area. In 1953 Oldroyd moved to a professorship at Swansea and in 1965 he moved to Liverpool; he died at a relatively early age in 1982. His 1950 paper was in many respects the high water mark of his career, and thereafter his research appeared to suffer at the expense of other academic responsibilities.

A leading American in the growth of continuum mechanics was Clifford Truesdell, born in Los Angeles in 1919. He was educated at the California Institute of Technology, graduating in 1941 and proceeding to Princeton to complete his PhD in 1943. After short stays at Princeton, Ann Arbor and MIT, he became head of the Theoretical Mechanics section at the US Naval Research Laboratory in Washington from 1948 to 1951, but moved to Indiana University in 1950, although remaining as a consultant at the Naval Research Laboratory until 1955. In 1961 he moved to John Hopkins University in Baltimore. Truesdell's greatest achievements were possibly his books and critical essays. His critical review *The Mechanical Foundation of Elasticity and Fluid Dynamics* first published in the *Journal of Rational Mechanics and Analysis* in 1952 [55] had considerable impact, and his article with Toupin, *Classical Field Theories*, published in 1960 in the *Handbook of Physics* [59] was a major achievement. A colourful and articulate personality he was to have considerable influence, particularly on mathematicians looking for a more axiomatic approach to mechanics. One such mathematician was Walter Noll who was born in Berlin in 1925 and educated at the University of Paris and the Technical University in Berlin before completing his PhD at Indiana under Truesdell's supervision in 1955 [60]; he became an important figure in rational mechanics, co-authoring with Truesdell their influential account of developments published in the *Handbook of Physics* in 1965 [61].

While Truesdell and Rivlin did not work at the same time at the Naval Research Laboratory, the former was

a consultant to this Laboratory when the latter spent a year working there. Clearly they must have met and interacted, and influenced colleagues around them. One such colleague was Jerald Ericksen who worked at this Laboratory from 1951 to 1957, and who wrote important papers with both (see next section). Rivlin in moving to Brown joined a strong group in mechanics, and Truesdell in moving to Indiana and then John Hopkins joined good departments, and so both were in positions to influence other researchers and students. In their different ways they were at the forefront of a growth of interest in the mechanics of continuous media that saw a period of exciting developments in this general area. Rivlin's formulation of this theory for finite elastic deformations and other subsequent developments demonstrated very clearly that general theories of material behaviour can be formulated in relatively simple closed form through appeal to invariance, material symmetry and mathematical theory of invariants [62, 63]. Moreover, Rivlin in his work on elasticity and non-linear viscous liquids also showed that one could solve problems for such theories in exact and general forms. Thus able people in applied mathematics and related areas were attracted to the subject, and encouraged particularly by Truesdell to look critically at perceived wisdoms and question their merits. Also the completeness of the theory of finite elasticity prompted subsequent researchers to seek more general treatments rather than simply pursue special cases. Clearly questions concerning the formulation of continuum models were prominent, so that there was much discussion and debate of issues such as invariance, material symmetry and thermodynamic restrictions. The former two were rather less contentious, with a consensus more or less reached; thermodynamics, however, especially irreversible aspects, was to remain a topic of some controversy, particularly between researchers from continuum mechanics and other backgrounds. During this period continuum mechanics was progressively applied to a diversity of materials and phenomena, and generally with a relatively high standard of mathematics.

Apart from Noll and Ericksen, of whom more later, it is not easy to select names from the many active in this area, and so we mention only a few for reasons that become apparent. In England following Rivlin's departure, Albert Green at Newcastle, and initially John Adkins and later Anthony Spencer at Nottingham, were prominent for their work in continuum mechanics; at first on aspects of finite elasticity influenced by Rivlin, but generally following developments in North America, or in some instances leading new developments. Clearly others were also involved, but the groups at Newcastle and Nottingham had greater influence, certainly in the UK and possibly also on continental Europe.

7. Jerald Ericksen

Jerald Ericksen was born in Portland, Oregon in 1924, and following war service in the USNR in the Pacific from 1943–1946, he completed his first degree at the University of Washington in 1947, going on to obtain his PhD at Indiana University in 1951. From there he went to Washington, DC, to work in the US Naval Research Laboratory, leaving in 1957 for an academic post at John Hopkins. In 1983 he moved to the University of Minnesota, retiring in 1990 and is now living in Oregon.

Apart from a short mathematical note written as a graduate student, Ericksen's first papers published in 1952 are on the flow of gases [64] and thin liquid jets [65], but are followed one year later by papers on finite elasticity [66] and non-Newtonian fluids [67], two areas of mechanics on which he was to become an authority. In the next five years he was to publish several papers which became very well known, one with Baker [68] and one written by himself [69] on inequalities occurring in elasticity (the 'Baker–Ericksen inequalities'), two [70, 71] on solutions possible in every isotropic, incompressible elastic body (Ericksen's universal solutions), a lengthy paper with Rivlin [72] on stress-deformation relations for viscoelastic materials (which introduced the 'Rivlin–Ericksen tensors'), another [73] on the overdetermination of flow in non-Newtonian liquids, and one with Truesdell [74] on theory for rods and shells employing directors. As a consequence, by the time that he moved to John Hopkins he was already well-known for his contribution to non-linear continuum mechanics.

In all, Ericksen has written in excess of one hundred papers (see [75]) virtually all as a sole author, the notable exceptions in which he is co-author do not reach double figures. The great majority are on aspects

of elasticity discussing topics such as finite elastic deformations, theories of rods, shells and plates, stability, thermoelasticity, and symmetries and phase transitions in crystals, in all some seventy papers. Of the remainder about ten deal with isotropic non-Newtonian liquids, a similar number examine continuum models for anisotropic liquids, and some two dozen are on liquid crystal theory.

In his very first paper on anisotropic liquids [76] published in 1960, Ericksen begins by citing a paper by Noll [60] which points out that most theories of anisotropic liquids do not satisfy necessary invariance conditions, and he presents what he believes to be the simplest properly invariant theory of anisotropic liquids, employing a unit vector field to describe the anisotropic axis. In two other papers [77, 78] published in the same year he discusses more general but still relatively simple models in which the vector may vary in magnitude; his motivation was simply to formulate properly invariant theories and examine their predictions, basically because other general theories of isotropic liquids being proposed at that time could not describe anisotropic liquids. In a further paper on this topic published two years later, Ericksen [79] is clearly motivated by consideration of polymer molecules being uncoiled by flow to produce anisotropy in the liquid. In these papers it is possible to assume a symmetric stress, and the orientation of the anisotropic axis is then determined from an equation giving the material time derivative of the director in terms of flow and the director. However, also in the early 1960s he presented static theory for liquid crystals within a mechanical framework [80], allowing him to propose general balance laws for these materials [2]. Since this essentially laid the foundations for the subsequent development of a dynamic theory for liquid crystals, we now discuss his formulation in detail.

For an infinitesimal perturbation of a volume V bounded by a surface S of a nematic liquid crystal, Ericksen [80] assumes that the change in the elastic stored energy W is equal to the work done on the material, and hence postulates that

$$\delta \int_V W dV = \int_V (\mathbf{F} \delta \mathbf{x} + \mathbf{G} \Delta \mathbf{n}) dV + \int_S (\mathbf{t} \delta \mathbf{x} + \mathbf{s} \Delta \mathbf{n}) dS \quad (10)$$

where $W = W(\mathbf{n}, N)$, $N = [\partial n_i / \partial x_j]$, $\Delta \mathbf{n} = \delta \mathbf{n} + (\delta \mathbf{x} \cdot \text{grad}) \mathbf{n}$, and $\delta \mathbf{x}$ and $\delta \mathbf{n}$ denote the infinitesimal changes in displacement and alignment; \mathbf{F} is body force per unit volume, \mathbf{t} is surface force per unit area, and \mathbf{G} and \mathbf{s} are generalized body and surface forces, respectively. Here \mathbf{n} is a unit vector field, and also the nematic is assumed to be incompressible, so that

$$\mathbf{n} \delta \mathbf{n} = 0, \quad \mathbf{n} \Delta \mathbf{n} = 0, \quad \text{div} \delta \mathbf{x} = 0. \quad (11)$$



Figure 3. Jerald Ericksen (left) and Frank Leslie (right). Photograph taken during a visit by Ericksen to Strathclyde University in the 1970s.

Consideration firstly of an arbitrary, infinitesimal rigid displacement \mathbf{a} in which $\delta\mathbf{x} = \mathbf{a}$, $\Delta\mathbf{n} = 0$, quickly yields

$$\int_V \mathbf{F} dV + \int_S \mathbf{t} dS = 0 \quad (12)$$

and secondly of an arbitrary, infinitesimal rigid rotation ω in which $\delta\mathbf{x} = \omega \times \mathbf{x}$, $\Delta\mathbf{n} = \omega \times \mathbf{n}$, soon leads to

$$\int_V (\mathbf{x} \times \mathbf{F} + \mathbf{n} \times \mathbf{G}) dV + \int_S (\mathbf{x} \times \mathbf{t} + \mathbf{n} \times \mathbf{s}) dS = 0 \quad (13)$$

following re-arrangement of the triple scalar products, and in each case assuming that the stored energy is unchanged. Equation (12) undoubtedly represents balance of forces, while (13) is a balance of moments. Thus one can identify the generalized forces with a body moment \mathbf{K} and surface moment \mathbf{I} through

$$\mathbf{K} = \mathbf{n} \times \mathbf{G}, \quad \mathbf{I} = \mathbf{n} \times \mathbf{s}. \quad (14)$$

Returning to the original hypothesis (10), since V is a material volume, one has

$$\begin{aligned} \delta \int_V W dV &= \int_V \delta W dV + \int_S W \delta \mathbf{x} \cdot \mathbf{v} dS \\ &= \int_V [\delta W + (\delta \mathbf{x} \cdot \text{grad}) W] dV \end{aligned} \quad (15)$$

\mathbf{v} denoting the unit outwards normal, and using incompressibility. A straightforward but tedious calculation then leads to

$$\begin{aligned} \delta \int_V W dV &= \int_S (\delta \mathbf{x} \cdot \mathcal{T} \mathbf{v} + \Delta \mathbf{n} \cdot \mathcal{S} \mathbf{v}) dS \\ &\quad - \int_V \left[\delta \mathbf{x} \cdot \text{div } \mathcal{T} + \Delta \mathbf{n} \cdot \left(\text{div } \mathcal{S} - \frac{\partial W}{\partial \mathbf{n}} \right) \right] dV \end{aligned} \quad (16)$$

where $\mathcal{T} = -N^T \partial W / \partial N$, $\mathcal{S} = \partial W / \partial N$, and the divergence is with respect to the second index. Hence, recalling the constraints (11), a comparison between the equations (10) and (16) provides expressions for the surface and generalized surface forces

$$\mathbf{t} = -p\mathbf{v} + \mathcal{T}\mathbf{v}, \quad \mathbf{s} = \beta\mathbf{n} + \mathcal{S}\mathbf{v} \quad (17)$$

where p is an arbitrary pressure and β an arbitrary scalar. In addition one obtains two equations

$$\mathbf{F} - \text{grad } p + \text{div } \mathcal{T} = 0, \quad \text{div } \mathcal{S} - \frac{\partial W}{\partial \mathbf{n}} + \mathbf{G} + \gamma\mathbf{n} = 0, \quad (18)$$

where γ is an arbitrary scalar. The former is clearly the point form of the balance of forces (12), and the latter

can be shown to be equivalent to the balance of moments (13). This last result requires the identity

$$\begin{aligned} \mathbf{n} \otimes \frac{\partial W}{\partial \mathbf{n}} + N \frac{\partial W^T}{\partial N} + N^T \frac{\partial W}{\partial N} \\ = \frac{\partial W}{\partial \mathbf{n}} \otimes \mathbf{n} + \frac{\partial W}{\partial N} N^T + \frac{\partial W^T}{\partial N} N \end{aligned} \quad (19)$$

which follows from the invariance requirement $W(\mathbf{n}, N) = W(\mathcal{P}\mathbf{n}, \mathcal{P}N\mathcal{P}^T)$, in which \mathcal{P} is any proper orthogonal matrix, by choosing $\mathcal{P} = \mathcal{I} + \varepsilon\mathcal{R}$, $\mathcal{R} + \mathcal{R}^T = 0$, where ε is an infinitesimal constant. In the above, $\mathbf{a} \otimes \mathbf{b}$ denotes the 3×3 matrix with (i, j) th element $a_i b_j$.

Generalizing the above, Ericksen [2] proposes balance laws for dynamic and flow effects in nematic liquid crystals. Straightforwardly, the obvious generalization of equation (12) or the first of equations (18) representing conservation of linear momentum is

$$\frac{d}{dt} \int_V \rho \mathbf{u} dV = \int_V \mathbf{F} dV + \int_S \mathbf{t} dS \quad (20)$$

ρ denoting density, \mathbf{u} the velocity vector, and d/dt the material time derivative. Also the balance of moments (13) suggests that one assumes, for conservation of angular momentum

$$\begin{aligned} \frac{d}{dt} \int_V \left(\rho \mathbf{x} \times \mathbf{u} + \sigma \mathbf{n} \times \frac{d\mathbf{n}}{dt} \right) dV \\ = \int_V (\mathbf{x} \times \mathbf{F} + \mathbf{n} \times \mathbf{G}) dV + \int_S (\mathbf{x} \times \mathbf{t} + \mathbf{n} \times \mathbf{s}) dS. \end{aligned} \quad (21)$$

Ericksen motivated the extra contribution to the nematic's angular moment through a simple model, σ being a small constant. Finally, motivated by Oseen [12] and the second of equations (18) he adds

$$\frac{d}{dt} \int_V \sigma \frac{d\mathbf{n}}{dt} dV = \int_V (\mathbf{G} + \mathbf{g}) dV + \int_S \mathbf{s} dS \quad (22)$$

where in equilibrium essentially one has

$$\mathbf{g} = \gamma\mathbf{n} - \frac{\partial W}{\partial \mathbf{n}}, \quad \mathbf{s} = \mathcal{S}\mathbf{v}. \quad (23)$$

8. Frank Leslie

Frank Leslie was born in 1935 in Dundee, Scotland, where he was educated, studying at Queen's College, then a college of St Andrews University. Graduating in 1957, he moved to Manchester University, where he joined the staff in 1959, obtaining his PhD in 1961. The following academic year he spent at MIT, before returning to England to a university post at Newcastle. In 1966 he returned to the USA to spend a sabbatical

year at John Hopkins, and a year after his return to Newcastle he left for a post at the University of Strathclyde in Glasgow in 1968.

For the first year of his PhD, Leslie worked on a problem concerning the cooling of turbine blades but spent the remainder of his time at Manchester investigating predictions based on Oldroyd's theory of visco-elastic liquids [81], Oldroyd being the external examiner for his PhD. During his year at MIT he saw some of Ericksen's papers on anisotropic liquids in the literature, and his curiosity was sufficiently aroused to write for off-prints. At Newcastle he readily agreed to Green's suggestion that he work on some problems for anisotropic liquids using Ericksen's theory; later, encouraged by Green, he began to consider the formulation of such theories employing an approach proposed by Green and co-workers [82]. The outcome was a paper [83] essentially deriving Ericksen's theory from a rather more general starting point. Soon thereafter he went to Baltimore for a sabbatical at John Hopkins, where Ericksen suggested that he attempt to derive a theory for nematic liquid crystals somewhat similar to that proposed by Anzelius [14], but exploiting recent developments in continuum mechanics.

Accepting Ericksen's conservation laws described in the previous section, Leslie [3] adopted a thermodynamic approach proposed by Coleman and Noll [84] to complete the theory. He therefore required Ericksen's balance law of energy given by

$$\begin{aligned} \frac{d}{dt} \int_V \left(\frac{1}{2} \rho \mathbf{u} \cdot \mathbf{u} + \frac{1}{2} \sigma \mathbf{w} \cdot \mathbf{w} + U \right) dV \\ = \int_V (r + \mathbf{F} \cdot \mathbf{u} + \mathbf{G} \cdot \mathbf{w}) dV + \int_S (\mathbf{t} \cdot \mathbf{u} + \mathbf{s} \cdot \mathbf{w} - h) dS \end{aligned} \quad (24)$$

where $\mathbf{w} = d\mathbf{n}/dt$, \mathbf{n} once more a unit vector; U denotes the internal energy per unit volume, r the heat supply per unit volume per unit time, and h the flux of heat out of the volume per unit area per unit time. He further assumed an entropy production inequality of the form

$$\frac{d}{dt} \int_V E dV - \int_V \frac{r}{T} dV + \int_S k dS \geq 0 \quad (25)$$

where E denotes entropy per unit volume, T is temperature, and k the flux of entropy out of the volume per unit area per unit time. The latter is used to limit the choice of constitutive relations.

By appeal to the familiar tetrahedron argument one can obtain from the balance laws and the above inequality the results

$$\mathbf{t} = \mathcal{T}\mathbf{v}, \quad \mathbf{s} = \mathcal{S}\mathbf{v}, \quad h = \mathbf{q} \cdot \mathbf{v}, \quad k = \mathbf{p} \cdot \mathbf{v} \quad (26)$$

involving \mathcal{T} the stress matrix, \mathcal{S} the generalized stress matrix, and \mathbf{q} and \mathbf{p} the heat and entropy flux vectors, respectively. As a consequence the balance law for linear momentum, and the director balance law can be written as

$$\rho \frac{d\mathbf{u}}{dt} = \mathbf{F} + \text{div } \mathcal{T}, \quad \sigma \frac{d\mathbf{w}}{dt} = \mathbf{G} + \mathbf{g} + \text{div } \mathcal{S} \quad (27)$$

while angular momentum simply reduces to

$$\mathcal{L} = \mathcal{L}^T, \quad \text{where } \mathcal{L} = \mathcal{T} + \mathcal{S}\mathcal{N}^T + \mathbf{n} \otimes \mathbf{g}. \quad (28)$$

Also, following some manipulation employing the above, the energy balance can be expressed in point form as

$$\frac{dU}{dt} = r - \text{div } \mathbf{q} + \text{tr}(\mathcal{T}\mathcal{D}) + \text{tr}(\mathcal{S}\mathcal{W}^T) - \mathbf{g} \cdot \hat{\mathbf{n}} \quad (29)$$

where

$$\hat{\mathbf{n}} = \mathbf{w} - \mathcal{Q}\mathbf{n}, \quad \mathcal{W} = \nabla \mathbf{w} - \mathcal{Q}\mathcal{N}, \quad 2\mathcal{Q} = \nabla \mathbf{u} - (\nabla \mathbf{u})^T. \quad (30)$$

Further, introducing the Helmholtz free energy W and a vector \mathbf{f} by

$$W = U - TE, \quad \mathbf{f} = \mathbf{q} - T\mathbf{p} \quad (31)$$

the entropy inequality (25) with the aid of equation (29) can be written in the form

$$\begin{aligned} \text{tr}(\mathcal{T}\mathcal{D}) + \text{tr}(\mathcal{S}\mathcal{W}^T) - \mathbf{g} \cdot \hat{\mathbf{n}} - \mathbf{p} \cdot \nabla T \\ - \left(\frac{dW}{dt} + E \frac{dT}{dt} \right) - \text{div } \mathbf{f} \geq 0 \end{aligned} \quad (32)$$

which is employed below to limit options.

At any material point of the nematic liquid crystal at time t , Leslie [3] assumes that

$$\mathcal{T}, \mathcal{S}, \mathbf{g}, \mathbf{q}, \mathbf{p}, U \text{ and } E \quad (33)$$

are all functions of

$$\nabla \mathbf{u}, \mathbf{n}, \mathcal{N}, \mathbf{w}, T \text{ and } \nabla T \quad (34)$$

evaluated at that point at that time. However, invariance to superimposed rigid body motions requires that one replaces the latter by

$$\mathcal{D}, \mathbf{n}, \mathcal{N}, \hat{\mathbf{n}}, T \text{ and } \nabla T. \quad (35)$$

Also a rather tedious calculation shows that the inequality (32) limits the free energy, entropy and entropy flux to be of the forms

$$W = W(\mathbf{n}, \mathcal{N}, T), \quad E = - \frac{\partial W}{\partial T}, \quad \mathbf{p} = \frac{\mathbf{q}}{T} \quad (36)$$

with the inequality reducing to

$$\text{tr}[(\mathcal{T} - \mathcal{T}^e)\mathcal{D}] + \text{tr}[(\mathcal{S} - \mathcal{S}^e)\mathcal{W}^T] - \left(\mathbf{g} + \frac{\partial W}{\partial \mathbf{n}}\right) \hat{\mathbf{n}} - \mathbf{q} \frac{\nabla T}{T} \geq 0 \quad (37)$$

where

$$\mathcal{T}^e = -N^T \frac{\partial W}{\partial N}, \quad \mathcal{S}^e = \frac{\partial W}{\partial N}. \quad (38)$$

Hence, recalling the constraints of incompressibility and the director \mathbf{n} having fixed magnitude, Leslie obtained equilibrium contributions of the form

$$\begin{aligned} \mathcal{T} &= -p\mathcal{I} + \mathcal{T}^e, \quad \mathcal{S} = \mathbf{n} \otimes \beta + \mathcal{S}^e, \\ \mathbf{g} &= \gamma \mathbf{n} - (\beta \text{ grad})\mathbf{n} - \frac{\partial W}{\partial \mathbf{n}} \end{aligned} \quad (39)$$

with β an arbitrary vector and γ an arbitrary scalar, essentially in agreement with the results of Ericksen. Also, under isothermal conditions the heat flux must be zero.

Given the results (39), the dissipative contributions to stress \mathcal{T}^d , generalized stress \mathcal{S}^d , the intrinsic generalized body force \mathbf{g}^d , and heat flux \mathbf{q}^d must satisfy

$$\text{tr}(\mathcal{T}^d \mathcal{D}) + \text{tr}(\mathcal{S}^d \mathcal{W}^T) - \mathbf{g}^d \cdot \hat{\mathbf{n}} - \mathbf{q}^d \frac{\nabla T}{T} \geq 0. \quad (40)$$

However, given the constitutive assumptions (33) and (35) it quickly follows from this inequality that

$$\mathcal{S}^d = 0. \quad (41)$$

Thus the generalized surface force is always given by the second of equations (39) and the inequality simplifies further to

$$\text{tr}(\mathcal{T}^d \mathcal{D}) - \mathbf{g}^d \cdot \hat{\mathbf{n}} - \mathbf{q}^d \frac{\nabla T}{T} \geq 0. \quad (42)$$

Also, with the aid of the identity (19) of the previous section, the relationship (28) above now reduces to

$$\mathcal{L}^d = \mathcal{L}^{dT}, \quad \text{where } \mathcal{L}^d = \mathcal{T}^d + \mathbf{n} \otimes \mathbf{g}^d. \quad (43)$$

To obtain expressions for the remaining dynamic or dissipative contributions, Leslie introduced an additional assumption, basically motivated by the theory of Anzelius. Rather than pursue general relationships for these quantities as functions of the variables (35), he replaced (35) by

$$\mathcal{D}, \mathbf{n}, \hat{\mathbf{n}}, T \text{ and } \nabla T \quad (44)$$

thus omitting terms containing director gradients from these contributions. Also, largely motivated by the experimental results of Miesowicz [38] and Tsvetkov [37], he further assumed a linear dependence upon the

velocity and temperature gradients. In this way, since nematic material symmetry requires isotropic relationships independent of the change of sign of the director, using the results of Smith [62] Leslie obtained

$$\begin{aligned} \mathcal{T}^d &= \alpha_1 \mathbf{n} \mathcal{D} \mathbf{n} \mathbf{n} \otimes \mathbf{n} + \alpha_2 \hat{\mathbf{n}} \otimes \mathbf{n} + \alpha_3 \mathbf{n} \otimes \hat{\mathbf{n}} + \alpha_4 \mathcal{D} \\ &\quad + \alpha_5 \mathcal{D} \mathbf{n} \otimes \mathbf{n} + \alpha_6 \mathbf{n} \otimes \mathcal{D} \mathbf{n} \end{aligned} \quad (45)$$

and

$$\mathbf{q}^d = \kappa_1 \nabla T + \kappa_2 \nabla T \cdot \mathbf{n} \mathbf{n} \quad (46)$$

the α s and κ s at most functions of temperature. Also, it follows from the residual form of angular momentum (43) that

$$\mathbf{g}^d = -\gamma_1 \hat{\mathbf{n}} - \gamma_2 \mathcal{D} \mathbf{n}, \quad \gamma_1 = \alpha_3 - \alpha_2, \quad \gamma_2 = \alpha_6 - \alpha_5. \quad (47)$$

Hence, in summary, Leslie [3] obtained the following relationships for stress, generalized stress, and the generalized intrinsic body force

$$\begin{aligned} \mathcal{T} &= -p\mathcal{I} - N^T \frac{\partial W}{\partial N} + \mathcal{T}^d, \quad \mathcal{S} = \mathbf{n} \otimes \beta + \frac{\partial W}{\partial N} \\ \mathbf{g} &= \gamma \mathbf{n} - (\beta \text{ grad})\mathbf{n} - \frac{\partial W}{\partial \mathbf{n}} + \mathbf{g}^d \end{aligned} \quad (48)$$

where the dynamic contributions are as above, and the energy W is that derived by Oseen [21], but more straightforwardly by Frank [53].

9. Discussion

The theory described in the two preceding sections represents a macroscopic approach, based on classical mechanics, but it should be mentioned that a few years after this theory was finally developed, a group at Harvard proposed a microscopic approach, based on a study of correlation functions and invoking irreversible thermodynamics [85]. The resulting theory is restricted to fluctuations around equilibrium, but it has been shown by de Gennes [86] that the two theories are essentially identical within this limitation. However, the former theory is more suitable for performing calculations in specific cases, not being restricted to small perturbations of a uniformly aligned nematic, and thus most existing data have been analysed within this framework. For this reason we do not present the Harvard approach to nematodynamics in this article.

The derivation of the theory presented above coincided with the beginning of a significant upsurge of interest in thermotropic liquid crystals, and as a consequence it was rather quickly found to describe many effects associated with nematic liquid crystals rather well, as can be seen, for example, from the reviews by Jenkins [87], Leslie [88], and Dubois-Violette *et al.* [89]. However, possibly

the most striking vindication of the theory lay in the experimental confirmation (1) by Fisher and Frederickson [90] of the unusual scaling for Poiseuille flow predicted by Atkin [91], and also (2) by Wahl and Fischer [92] of the scaling for simple shear first noted by Ericksen [93], with the result that the theory is now widely accepted.

In time it has proved possible to develop and re-interpret the theory. Fairly soon after its derivation Parodi [94] appealed to thermodynamics to reduce the number of independent viscous coefficients from six to five through

$$\gamma_2 = \alpha_6 - \alpha_5 = \alpha_2 + \alpha_3 \quad (49)$$

as it transpired, not an insignificant reduction in terms of their measurement, because this allows their determination from the four independent viscosities measured in shear flow [38] and the rotational viscosity measured in the Tsvetkov experiment [37]. With this additional assumption Ericksen [95] was able to show that one could derive the dissipative contributions to stress \mathcal{T}^d and the intrinsic torque \mathbf{g}^d from a dissipation function, using

$$\mathcal{T}^d = \frac{1}{2} \frac{\partial \Delta}{\partial \nabla \mathbf{u}}, \quad \mathbf{g}^d = -\frac{1}{2} \frac{\partial \Delta}{\partial \mathbf{w}}, \quad \mathbf{w} = \frac{d\mathbf{n}}{dt} \quad (50)$$

thus allowing him to recast the equations in a neat and efficient manner. Also, in the original derivation, Leslie and Ericksen employ equation (22) similar to that proposed by Oseen [12] for the director, and regard the outcome from conservation of angular momentum (28) as a constraint upon relations for stress and generalized forces. However, it was soon realized that one can simply assume conservation of angular momentum, equation (21), and derive the Oseen equation from it by introducing the generalized intrinsic torque through what was previously the outcome of angular momentum (28), or effectively (43). In this way it is possible to regard the theory as based on the balance laws for linear and angular momentum, and more recently Leslie [96] has shown how the theory can be based solely upon linear and angular momentum without recourse to generalized forces.

Given that Leslie [3] was to some extent motivated by the theory proposed by Anzelius [14], it is of some interest to conclude by comparing the theories. There are essentially two main differences between them. The first is that Anzelius assumed a symmetric viscous stress, and the second is that he did not include terms involving the material time derivative of the director in his equations. As a consequence of the former there is no intrinsic viscous torque in this theory due to the asymmetric stress, and on account of the latter his viscous stress depends solely upon the alignment and

the rate of strain tensor. Realizing that flow has an aligning influence, Anzelius introduces such a viscous torque, but independent of his viscous stress, and not properly invariant on account of its explicit dependence upon vorticity. Also, Oseen [12] remarks that its role with respect to work and energy is not clear. Had Anzelius introduced the time derivative of the director, he then had the means to make his torque properly invariant, but he may not have seen the need to do this. Also, he would then have been able to invoke simple thermodynamic arguments concerning viscous dissipation, but he had little by way of precedents to follow in this respect. Of course, had Anzelius known of the work by Tsvetkov [37], he may well have included the time derivative of the director, but it is difficult to speculate as to whether or not he would then have reconsidered the other aspects mentioned above.

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