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Nonlinear dissipative effects in the hydrodynamics of liquid crystals

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The influence of dissipative effects on the nonlinear hydrodynamic equations of various types of liquid crystals like nematics, cholesterics, smectics *A*, *C*, is investigated. An experiment for uniaxial nematics is proposed which allows the detection of one group of the novel terms presented. It is shown that the viscous nonlinearities in the stress tensor of uniaxial nematics discussed by Moritz and Franklin do not exist.

I. INTRODUCTION AND GENERAL REMARKS

The derivation of linear hydrodynamic equations by using symmetry arguments and thermodynamics is a well-established procedure which has been pursued for the various types of liquid crystals by many authors, cf., e.g., for uniaxial nematics,[1–3] for cholesterics,[2–8] and for smectics *A* and smectics *C*.[2,3,5,7] However, as it turns out, nonlinear effects are nearly always essential for the description of the experiments. This can be seen easily, e.g., from the fact that the threshold for the onset of shear flow instabilities and thermoconvective instabilities is much lower than in isotropic systems (for the calculation of the threshold values for various types of instabilities in liquid crystals we refer, e.g., to Refs. 9–15 and for the experimental detection to Refs. 16–20). Thus even at small shear rates or temperature gradients the hydrodynamic behavior is governed by nonlinear effects.

Due to the complicated structure of the order parameter in the various types of liquid crystals, the derivation of nonlinear hydrodynamic equations has attracted considerable attention only recently[3,7,8] and this work has concentrated on the examination of reversible effects. Very recently, Volovik and Kats[21] considered reversible macroscopic equations for various types of liquid crystals including hydrodynamic variables and those microscopic variables which are thought to be slow. The above-mentioned points become even more obvious while deriving nonlinear hydrodynamic equations[22,23] for the recently discovered biaxial nematics.[24]

Concerning the nonlinear dissipative hydrodynamic behavior there seems to exist only the work of Moritz and Franklin[25] on quadratic nonlinearities of the stress tensor in uniaxial nematics and related work by the same authors on nonlinear electrohydrodynamic effects in uniaxial nematics.[26] It will be the purpose of the present note to consider nonlinear effects in the dissipative behavior of the various types of liquid crystals and to propose experiments which will yield information about at least some of the novel parameters involved. In addition, it will be pointed out that the viscous effects in the stress tensor considered by Moritz and Franklin do not exist.

As is already well known from the study of simple fluids, there exists a class of dissipative nonlinearities which will be called the "trivial" ones in the following: all dissipative coefficients entering the nonlinear hydrodynamic equations may depend on the scalar quantities which exist in the hydrodynamic system under investigation. For simple fluids, e.g., the thermal conductivity, the two viscosities can depend on temperature and pressure leading to the experimentally and theoretically well-established non-Boussinesq effects.[27,28] For a binary mixture of fluids there appears an additional scalar quantity on which the transport parameters may depend; the concentration *c*. For the various types of liquid crystals to be discussed in the following (uniaxial nematics, cholesterics, smectics *A* and smectics *C*, biaxial nematics), the same as for simple and binary fluids holds true, of course. In addition, all transport parameters of cholesterics, smectics *A*, and smectics *C* can depend on $p_i p_j \nabla_i R_j$, where p_i is the direction parallel to the axis of the helices and R_i is the layer displacement[2,7]; i.e., all transport parameters assume the form

$$\alpha=\alpha_0+\alpha_1 p_i p_j \nabla_i R_j+\alpha_2(p_i p_j \nabla_i R_j)^2+\cdots, \tag{1.1}$$

where the α_i do not depend on the additional scalar quantity. For smectics A, p_i must be replaced by n_i, the normal to the smectic layers.) For smectics C, the transport parameters can depend, in addition, on the quantity $\vec{n}\cdot\vec{p}$. Thus it becomes possible to give an expression for all dissipative parameters which resembles, in structure, Eq. (1.1).

Since there exist preferred directions in liquid crystals, there is a further straightforward, well-known way to obtain nonlinear viscous effects: one has, e.g., only to replace the equilibrium value of the director in uniaxial nematics n_i^0 by its nonequilibrium value $n_i = n_i^0 + \delta n_i + O(\delta n^2)$, a feature which becomes crucial for the thermoconvective instability in nematics when heating is done from above.[9,16]

In the following we will not deal with the straightforward generalizations of the linear viscous effects on the hydrodynamic equations sketched above, but we will concentrate on the study of nonlinear viscosities which arise from new dynamic couplings. As it turns out, it is most convenient to start with the dissipation function R (the source term of the dynamic equation for the entropy density, cf., e.g., Ref. 5) and to expand this function into powers of the thermodynamic conjugate quantities. The contributions to the dissipative currents can then be easily obtained by the derivation of the dissipation function to the thermodynamic forces. In Sec. II we present the various new terms for the different types of liquid crystals and in Sec. III we sketch two experiments to detect one of the viscous nonlinearities in uniaxial nematics.

II. NONLINEAR VISCOSITIES

A. Uniaxial nematics

As has been indicated in Sec. I, we expand the dissipation function R in powers of the thermodynamic conjugates, and we restrict ourselves throughout the present paper to terms which are quadratic or cubic in the thermodynamic forces. In addition, we allow for the occurence of the hydrodynamic variables in the dissipation function and thus in the corresponding currents. To ensure positivity of the entropy production it is necessary, of course, to add appropriate terms, which are of fourth order in the thermodynamic forces, to the dissipation function. Since quadratic supplement is a straightforward procedure we refrain from writing down these terms. For the static behavior, the corresponding procedure has been discussed explicitly by the authors.

For uniaxial nematics we have, as hydrodynamic variables contributing to the dissipative behavior (as always, the density does not contribute to dissipation since its current is a conserved quantity), the density of linear momentum $\vec{g}$, the energy density ϵ (or entropy density σ), and the transverse components of the director field $\vec{n}, \delta\vec{n}$. The corresponding conjugate thermodynamic forces are the velocity $\vec{v}$, the temperature T, and $\vec{h}$. The quadratic terms in the dissipation function are given by[1,2,5]:

$$R^{(2)} = \kappa_{ij}(\nabla_i T)(\nabla_j T) + \eta_{ijkl}(\nabla_i v_j)(\nabla_k v_l) + \xi_{ij} h_i h_j \;, \tag{2.1}$$

where η_{ijkl} contains five phenomenological parameters, κ_{ij} two thermal conductivities, and ξ_{ij} brings along one parameter characterizing the self-viscosity of the director motion.

After having carried out the straightforward generalizations from the linear to the nonlinear domain discussed in Sec. I, there arises naturally the question whether nonlinearities exist in the entropy production which shows a new structure. For quadratic nonlinearities in the stress tensor, this point has been discussed in detail by Moritz and Franklin,[25] who considered especially terms in the stress tensor which are quadratic in the symmetrized velocity gradients.

We examine here as a first step all terms which are allowed by symmetry considerations and which are cubic in the thermodynamic forces. We find that the following terms are possible:

$$R^{(3)} = \zeta_{ijk} h_i (\nabla_j T)(\nabla_k T) + \eta_{ijklm} h_i (\nabla_j v_k)(\nabla_l v_m) \;, \tag{2.2}$$

where

$$\zeta_{ijk} = \zeta[(\delta_{ik} - n_i n_k) n_j + (\delta_{jk} - n_j n_k) n_i] \;,$$

$$\begin{aligned} \eta_{ijklm} = {} & \eta_1 (n_i \delta^{\mathrm{tr}}_{kl} \delta^{\mathrm{tr}}_{jm} + n_j \delta^{\mathrm{tr}}_{kl} \delta^{\mathrm{tr}}_{im} + n_k \delta^{\mathrm{tr}}_{ij} \delta^{\mathrm{tr}}_{lm} + n_l \delta^{\mathrm{tr}}_{ij} \delta^{\mathrm{tr}}_{km}) \\ & + \eta_2 (n_j n_k n_l \delta^{\mathrm{tr}}_{im} + n_i n_k n_l \delta^{\mathrm{tr}}_{jm} + n_i n_j n_l \delta^{\mathrm{tr}}_{km} + n_i n_j n_k \delta^{\mathrm{tr}}_{lm}) \\ & + \eta_3 (n_i \delta^{\mathrm{tr}}_{jk} \delta^{\mathrm{tr}}_{ml} + n_j \delta^{\mathrm{tr}}_{ik} \delta^{\mathrm{tr}}_{ml} + n_i \delta^{\mathrm{tr}}_{jl} \delta^{\mathrm{tr}}_{mk} + \delta^{\mathrm{tr}}_{il} \delta^{\mathrm{tr}}_{mk} n_j) \;, \end{aligned} \tag{2.3}$$

and where $\delta^{\text{tr}}_{ij}=\delta_{ij}-n_i n_j$.

For example, we find four new parameters. In addition it seems worth to notice that none of the terms presented by Moritz and Franklin are dissipative ones. Instead they are reversible; this follows from their behavior under time reversal. As reversible terms they must lead to vanishing entropy production; this is, however, not the case. Therefore these terms do not exist.

An experiment to measure the coefficient ζ presented in Eqs. (2.2) and (2.3) will be discussed in Sec. III. From inspection of the contributions (2.2) and (2.3) we arrive at the general conclusion that the bending of the director field induces nonlinear irreversible couplings to the temperature field and the symmetrized velocity gradients.

However, we wish to point out that general symmetry considerations allow further terms which are quadratic in the thermodynamic conjugates and which contain in addition the hydrodynamic variables. These terms contain the same number of gradients (or even one less) than the contributions given in Eqs. (2.2) and (2.3) and there seems to exist no argument to discard such terms. For uniaxial nematics we find

$$\begin{aligned}\widetilde{R}^{(3)}=\;&\psi_{mqij}(\nabla_i n_j)(\nabla_q T)h_m\\&+\eta_{ijklmnqp}(\nabla_i n_j)(\nabla_k n_l)(\nabla_m v_n)(\nabla_q v_p)\\&+\tau_{ijklmn}(\nabla_i n_j)(\nabla_k n_l)(\nabla_m T)(\nabla_n T)\\&+\lambda_{ijklmn}(\nabla_i n_j)(\nabla_k n_l)h_m h_n\ ,\end{aligned}\tag{2.4}$$

where, e.g.,

$$\psi_{mqij}=\psi_1\delta^{\text{tr}}_{ij}\delta^{\text{tr}}_{mq}+\psi_2\delta^{\text{tr}}_{mi}\delta^{\text{tr}}_{qj}\ .\tag{2.5}$$

All these terms lead to further dissipative couplings between the gradients of the director field and the other variables. The terms $\sim\eta_{ijklmnqp}$, e.g., can be interpreted as a generalization of the terms η_{ijkl} of Eqs. (2.1) by allowing for a dependence of the parameter on the gradients of $\vec{n}$.

B. Biaxial nematics

In biaxial nematics, which have attracted considerable attention recently, we have for $R^{(2)}$ (Ref. 22)

$$\begin{aligned}R^{(2)}=\;&\eta_{ijkl}(\nabla_i v_j)(\nabla_k v_l)+\kappa_{ij}(\nabla_i T)(\nabla_j T)\\&+\xi^m_{ij}h^m_i h^m_j+\xi^n_{ij}h^n_i h^n_j\ ,\end{aligned}\tag{2.6}$$

where η_{ijkl} contains nine parameters, κ_{ij} three different thermal conductivities, and ξ^m_{ij} and ξ^n_{ij} are given by

$$\begin{aligned}&\xi^m_{ij}=\xi_3\delta^3_{ij}\ ,\\&\xi^n_{ij}=\xi_1\delta^3_{ij}+\xi_2 m_i m_k\ .\end{aligned}\tag{2.7}$$

Due to the biaxiality the number of coefficients involved increases, unfortunately, in a rather drastic manner. Therefore we confine ourselves in the following on the display of the structure of the novel contributions and refrain from writing out the corresponding tensors in all detail.

For the terms analogous to $R^{(3)}$ of uniaxial nematics, we have

$$\begin{aligned}R^{(3)}=\;&\tau^m_{kij}h^m_k(\nabla_i T)(\nabla_j T)+\tau^n_{kij}h^n_k(\nabla_i T)(\nabla_j T)\\&+\eta^n_{ijklm}h^n_i(\nabla_j v_k)(\nabla_l v_m)\\&+\eta^m_{ijklm}h^m_i(\nabla_j v_k)(\nabla_l v_m)\ ,\end{aligned}\tag{2.8}$$

and for $\widetilde{R}^{(3)}$ we find (introducing as in Ref. 22 the vector $\vec{\Theta}\equiv\{\vec{m}\cdot\delta\vec{n},(\vec{n}\times\vec{m})\cdot\delta\vec{n},(\vec{m}\times\vec{n})\cdot\delta\vec{m}\}$ and corresponding notations for the thermodynamic forces)

$$\begin{aligned}\widetilde{R}^{(3)}=\;&\Sigma_{ijklmn}(\nabla_i\Theta_j)(\nabla_k\Theta_l)h^\theta_n h^\theta_m\\&+\Xi_{ijklmn}(\nabla_i\Theta_j)(\nabla_k\Theta_l)(\nabla_m T)(\nabla_n T)\\&+\tau_{ijklmnqp}(\nabla_i\Theta_j)(\nabla_k\Theta_l)(\nabla_m v_n)(\nabla_q v_p)\\&+\Lambda_{ijkl}(\nabla_i\Theta_j)h^\theta_k(\nabla_l T)\ .\end{aligned}\tag{2.9}$$

C. Smectics *A* and smectics *C*

For the quadratic terms we have for smectics *A* and smectics *C* (Ref. 5)

$$\begin{aligned}R^{(2)}=\;&\eta_{ijkl}(\nabla_i v_j)(\nabla_k v_l)+\kappa_{ij}(\nabla_i T)(\nabla_j T)\\&+\zeta_{ij}(\nabla_i\Phi)(\nabla_j\Phi)+\zeta_{ij}(\nabla_i T)(\nabla_j\Phi)\\&+\tau hh\ ,\end{aligned}\tag{2.10}$$

where Φ and h are the thermodynamic conjugates to the variable R (layer displacement) and to the variable δn (rotation of the averaged molecule axis about the layer normal p_i), respectively.

Of course, τ exists only in smectics *C*. For the number of coefficients involved in the tensors in (2.10) for smectics *A* and smectics *C*, respectively, we refer to Ref. 5. As it turns out there exist for smectics *A* no terms like those contained in $R^{(3)}$ and $\widetilde{R}^{(3)}$ for nematics and only the dependence of the transport parameters on the scalar quantity $(\nabla_i R_j p_i p_j)$ survives.

For smectics *C* the situation is completely different due to the fact that there are two different kinds of spontaneously broken continuous symmetries: a broken translational symmetry and a broken rotational symmetry. We find for the terms cubic in the forces

$$R^{(3)}=\tilde{\zeta}_{jk}h(\nabla_jT)(\nabla_kT)+\tilde{\xi}_{jklm}h(\nabla_jv_k)(\nabla_lv_m)$$
$$+\psi_{jk}h(\nabla_jT)(\nabla_k\phi)\ , \tag{2.11}$$

and for the terms which are quadratic in the forces and which contain the variables, we have

$$\tilde{R}^{(3)}=\Sigma_{ikmnqp}(\nabla_in)(\nabla_kn)(\nabla_mv_n)(\nabla_qv_p)$$
$$+\Xi_{ikmn}(\nabla_in)(\nabla_kn)(\nabla_mT)(\nabla_nT)$$
$$+\Theta_{ijkl}(\nabla_in)(\nabla_jn)(\nabla_k\Phi)(\nabla_l\Phi)$$
$$+\Omega_{ijkl}(\nabla_in)(\nabla_jn)(\nabla_k\Phi)(\nabla_lT)\ . \tag{2.12}$$

We refrain from writing down the details of the tensors, we note in passing, however, that, eg., ψ_{jk} brings along only one additional phenomenological parameter.

D. Cholesterics

Contrary to all other types of liquid crystals, cholesterics allow for the existence of a pseudoscalar quantity whose existence has drastic consequences on the hydrodynamic behavior of cholesterics.[4,6–8,11–15,20,29] We obtain for the terms cubic in the forces

$$R^{(3)}=\kappa_{ijk}(\nabla_iT)(\nabla_jT)(\nabla_kT)$$
$$+\tilde{\eta}_{ijklm}(\nabla_iv_k)(\nabla_jv_l)(\nabla_mT)$$
$$+\psi_{ijkl}(\nabla_iv_j)(\nabla_kv_l)(\nabla_m\Phi)p_m$$
$$+\chi(\nabla_i\Phi)(\nabla_j\Phi)(\nabla_k\Phi)p_kp_jp_i$$
$$+\omega_k(\nabla_kT)(\nabla_i\Phi)(\nabla_j\Phi)p_ip_j$$
$$+\tau_{jk}(\nabla_jT)(\nabla_kT)(\nabla_i\Phi)p_i\ , \tag{2.13}$$

whereas no terms like those of $\tilde{R}^{(3)}$ in nematics and smectics *C* occur in cholesterics. Due to the existence of the pitch, dynamical viscous couplings of the temperature field (and the quantity $\nabla\phi$), on the one hand, and the symmetrized velocity gradients, on the other hand, become possible in cholesterics.

III. PROPOSED EXPERIMENTS FOR UNIAXIAL NEMATICS

We consider two different experimental configurations to detect the coefficient ζ in uniaxial nematics representing a viscous coupling between the temperature field and the thermodynamic conjugate force of the director field.

First we discuss a texture of uniaxial nematics where the director is forced by special surface treatment of the sidewalls to make a *constant* angle Θ ($\Theta\neq0°,90°$) with the *z* axis (cf. Fig. 3.7 of Ref. 2). In addition we assume that a heat gradient (well below threshold for the onset of any thermoconvective instability) parallel to the *z* axis is applied.

In general we have from Eqs. (2.1)–(2.3) for the entropy current and the quasicurrent X_i

$$j_i^{\sigma}=\zeta_{ijl}(\nabla_jT)h_l\ , \tag{3.1}$$

$$X_i=\zeta_{lji}(\nabla_lT)(\nabla_jT)\ . \tag{3.2}$$

Thus stationary solutions of the hydrodynamic equations must satisfy the conditions

$$\nabla_i(\kappa_{ij}\nabla_jT)=\nabla_i(\zeta_{ijl}h_l\nabla_jT)$$
$$-\xi h_i+\zeta_{lji}(\nabla_lT)(\nabla_jT)=0\ . \tag{3.3}$$

If we assume

$$\nabla_iT=\delta_{iz}g(z)\ , \tag{3.4}$$

we have

$$\nabla_i[\kappa_{iz}g(z)]=\nabla_i[\zeta_{ijl}h_l\delta_{jz}g(z)] \tag{3.5}$$

and

$$\xi h_i=g^2(z)\delta_{lz}\delta_{jz}\zeta_{lji}$$
$$=g^2(z)\zeta[\,(\delta_{li}-n_ln_i)n_j$$
$$+\,(\delta_{ji}-n_in_j)n_l\,]\delta_{lz}\delta_{jz} \tag{3.6}$$

with

$$h_i=\frac{\partial F^g}{\partial n_i}-\nabla_j\frac{\partial F^g}{\partial\nabla_jn_i}\ ,$$

where F^g is the gradient free energy of uniaxial nematics.

It is now easy to check that Eqs. (3.5) and (3.6) do not allow for a solution $\vec{n}=$const., $\nabla_jT=\delta_{jz}$const (except for $\Theta=0°,90°$).

Thus, the proposed experiment provides a decisive test for the existence and for the order of magnitude of the coefficient ζ: If an external temperature gradient is applied, making an angle Θ with the initial (constant) orientation of $\vec{n}$, there exist exactly two configurations ($\Theta=0°,90°$) in which no distortion of the constant director field occurs, whereas for all other initial orientations of

$\vec{n}$ ($\Theta\neq 0°,90°$), the director field $\vec{n}$ will become inhomogeneous (and the temperature distribution nonlinear in z). The strength of the resulting inhomogeneity is proportional to the magnitude of ζ (the nonlinearity of $T(z)\sim\zeta^2$).

A second possibility to achieve in the bulk an angle Θ between the director orientation $\vec{n}$ and the applied temperature gradient $\vec{\nabla}T$, is the application of an external static magnetic field. Again, for $\Theta\neq 0°,90°$ there occurs a distortion of $\vec{n}$ proportional to ζ. The formal considerations are as above; in Eq. (3.6) the gradient free energy F^g now contains also the orientational energy of an uniaxial nematic in an external magnetic field.[2]

Equations (3.5) and (3.6) have been derived for bulk hydrodynamics. In realistic experiments, therefore, one has to minimize or to subtract the influence of boundary layers (in which $\vec{n}$ does not fit the bulk behavior) from the evaluation of ζ.

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