Vol. 25 No. 1-2 KEXUE TONGBAO January 1980

NONLINEAR LIQUID CRYSTALS

LIU JIAGANG (刘家冈) AND LIN LEI (林 磊)*
(*Institute of Physics, Academia Sinica*)

Received February 19, 1979.

I. INTRODUCTION

Nonlinear effect in liquid crystals is an important subject[1,2] which has been widely investigated (see Ref. [1] and references therein). Generally speaking, nonlinear effects in liquid crystals may be divided into three classes[1]: (i) The material of liquid crystal itself is nonlinear (i.e. the dissipation function is higher than a second order polynomial; see Refs. [1] and [3]). (ii) Nonlinear optical properties. (iii) The effects due to higher order terms left over after the linearization of the relevant equations. In this paper we are concerned with nonlinearities of class (i).

In simple liquid, nonlinear terms in the stress tensor can lead to observable effects. For example, due to the action of the nonlinear terms shear flow may give rise to normal stresses, as is shown in the case with the Weissenberg effect[4]. As for liquid crystals, Moritz and Franklin have discussed the nonlinear terms in the stress tensor for incompressible nematics[5]. However, their work is unsatisfactory (see Sec. III below).

Recently, dissipation functions have been introduced by Lam to treat the irreversible thermodynamics of molecular liquids and molecular solids with internal structures[6]. In addition to the capability of systematic derivation of the conservation laws of momentum, energy and angular momentum, this method includes in a natural way the important consequences of angular momentum conservation and the Onsager reciprocal relations. When applied to liquid crystals, the theory of dissipation functions in the linear regime agrees with that of Ericksen-Leslie[1].

Although there are still some ambiguities in the theoretical application of the Onsager reciprocal relations[7] for liquid crystals[8] and many other materials, there exist quite a number of experimental verifications already. In the nonlinear regime, the existence of dissipation functions (related to the generalization of the Onsager reciprocal relations) has been established at least in the case of reaction kinetics of chemical processes[9]. Therefore, the use of dissipation functions in describing nonlinear liquid crystals and an investigation of the resulting consequences are not unjustified. (For discussions of some basic questions on dissipation functions in the nonlinear regime, see Ref. [10]).

II. THEORY

For isothermal processes, the dissipation function of nematic liquid crystals is given by $D = D(d_{ij}, N_i)$, where d_{ij} is the symmetric tensor of velocity gradient and N_i is the vector related to the velocity of the director (see Ref. [1] for detailed definitions). In nematics, the material is invariant under rotation about the director $\boldsymbol{n}$ and there is a reflection symmetry with respect to a plane containing $\boldsymbol{n}$. Consequently, there are only ten basic invariants that can be formed from d_{ij} and N_i up to the third order (see Ref. [11], Table IV):

$$\begin{aligned}&d_{33},\ d_{\alpha\alpha},\ d_{3\alpha}d_{\alpha3},\ d_{\alpha\beta}d_{\beta\alpha},\ d_{3\alpha}d_{\alpha\beta}d_{\beta3},\\&N_3,\ N_\alpha N_\alpha,\\&N_\alpha d_{\alpha3},\ N_\alpha d_{\alpha\beta}d_{\beta3},\ N_\alpha d_{\alpha\beta}N_\beta,\end{aligned}\tag{1}$$

where "3" is the direction of $\boldsymbol{n}$, and $\alpha, \beta = 1, 2$ are the two directions perpendicular to $\boldsymbol{n}$. The convention of summing over repeated subscripts in the same term is adopted. In (1), N_3 should be understood as $N_i n_i$, and $d_{3\alpha} = n_i d_{i\alpha}$, etc., where $i = x, y, z$. Obviously, $d_{\alpha\alpha}$ may be replaced by d_{ii}.

Because of the constraints $n_i n_i = 1$ and $d_{ii} = 0$ due to incompressibility, d_{ii} and N_3 in (1) are identically zero. We are left with only eight terms. From these eight basic invariants we may construct a third order polynomial for D such that

$$D = D^{(2)} + D^{(3)},\tag{2}$$

where $D^{(2)}$ is the second order part with five terms given by Eq. (5.1) of Ref. [1] (the a_1—a_5 terms). $D^{(3)}$ is the third order part which is the linear sum of the following eight terms:

$$\begin{aligned}&(d_{33})^3,\ d_{33}d_{3\alpha}d_{\alpha3},\ d_{33}d_{\alpha\beta}d_{\beta\alpha},\ d_{3\alpha}d_{\alpha\beta}d_{\beta3},\\&d_{33}N_\alpha d_{\alpha3},\ N_\alpha d_{\alpha\beta}d_{\beta3},\ d_{33}N_\alpha N_\alpha,\ N_\alpha d_{\alpha\beta}N_\beta.\end{aligned}\tag{3}$$

It is easy to show that (using the identity $d_{\alpha\beta}d_{\beta\gamma}d_{\gamma\alpha} = 0$), through a linear transformation, the eight invariants in (3) are equivalent to the following set:

$$\begin{aligned}\rho D^{(3)} = {}& e_1(d_{ij}[ij])^3 + e_2 d_{ij}d_{jp}d_{pi} + e_3[ij]d_{ij}d_{pk}d_{kp}\\&+ e_4[ij]d_{ip}d_{pk}d_{kj} + e_5[ijk]d_{ij}N_p d_{pk}\\&+ e_6[i]N_j d_{jp}d_{pi} + e_7[ij]d_{ij}N_k N_k\\&+ e_8 N_i d_{ij}N_j,\end{aligned}\tag{4}$$

where e_1—e_8 are constants, $[ij\cdots] \equiv n_i n_j\cdots$. Note that $D^{(3)}$ satisfies the symmetry requirement $D^{(3)}(\boldsymbol{n}) = D^{(3)}(-\boldsymbol{n})$. Using the relations[1]

$$\left.\begin{aligned}\sigma'_{ij} &= \sigma'^0_{ij} + g'_{[i}n_{j]},\\\sigma'^0_{ij} &= \frac{\rho}{2}(\partial D/\partial d_{ij} + \partial D/\partial d_{ji}),\\g'_i &= -\rho\partial D/\partial N_i,\end{aligned}\right\}\tag{5}$$

we find $\sigma'_{ij} = \sigma'^{(2)}_{ij} + \sigma'^{(3)}_{ij}$, $g'_i = g'^{(2)}_i + g'^{(3)}_i$, where $\sigma'^{(2)}_{ij}$ and $g'^{(2)}_i$ are the linear parts given by the a_1—a_5 terms in Eqs. (5.5) and (5.3) of Ref. [1]. The bilinear parts are obtained by (4) and (5) as follows:

$$g_i'^{(3)} = -2e_7d_{kp}[kp]N_i - e_5d_{kp}d_{ij}[kpj] - e_6'd_{ij}d_{jp}[p] - 2e_8d_{ij}N_j, \tag{6}$$

$$\sigma_{ij}'^{(3)} = 3e_1[ijpqrs]d_{pq}d_{rs} + \left(\frac{e_5}{2} + e_7\right)[ipq]N_jd_{pq} + \left(\frac{e_5}{2} - e_7\right)[jpq]N_idpq + 2e_3[pq]d_{ij}d_{pq} + \frac{e_5}{2}[ipqr]d_{jp}d_{qr} - \frac{e_5}{2}[jpqr]d_{ip}d_{qr} + e_4[pq]d_{ip}d_{jq} + 3e_2d_{ip}d_{jp} + e_8N_iN_j + \frac{e_6}{2}[p]d_{ip}N_j + \frac{e_6}{2}[p]d_{jp}N_i + \left(\frac{e_6}{2} + e_8\right)[i]d_{jp}N_p + \left(\frac{e_6}{2} + e_4\right)[iq]d_{jp}d_{pq} + \left(\frac{e_6}{2} - e_8\right)[j]d_{ip}N_p + \left(-\frac{e_6}{2} + e_4\right)[jq]d_{ip}d_{pq} + e_3[ij]d_{pq}d_{qp} + e_7[ij]N_kN_k + e_5[ijp]N_kd_{kp}. \tag{7}$$

In (6) and (7) there are only eight independent coefficients. The mutual connection between the coefficients in $g_i'^{(3)}$ and $\sigma_{ij}'^{(3)}$ is the consequence of the angular momentum conservation[1], while the connection between the different coefficients in $\sigma_{ij}'^{(3)}$ results from the assumption of the generalized Onsager reciprocal relations (through the assumed existence of a dissipation function). In our theory these interrelations are hidden in (4) and are derived directly through the dissipation function.

III. Discussion

Comparing with Ref. [5] (called MF below) we obtained not only the expression for σ'_{ij} but also the nonlinear part of g'_i in (6). In Eq. (11) of MF, there are only fifteen terms in σ'_{ij} and the last three terms in (7) of ours are missing. In fact, $[ij]d_{pq}d_{qp}$ does appear in Eq. (7) of MF (the η_6 term) but is absent in the final result in Eq. (11) of MF. It appears there is an error in the calculations in between. On the other hand, the two terms $[ij]N_kN_k$ and $[ijp]N_kd_{kp}$ are completely absent in MF. These are the shortcomings and unsatisfactory aspects of MF. Obviously, one of the advantages of the dissipation function method is that once the invariants are written down, the stress tensor and other quantities can be derived directly and immediately. As a result, no terms will be left out even in relatively complicated cases.

Comparing Eq. (7) with Eq. (11) of MF, we obtain seven independent relations among the fifteen dissipation coefficients ξ_1—ξ_{15} of MF, viz.:

$$\xi_2 + \xi_3 = 2\xi_5, \quad \xi_5 + \xi_6 = 0, \quad \xi_{10} = \xi_{11},$$
$$\xi_{12} + \xi_{14} = 2\xi_{11}, \quad \xi_{12} - \xi_{14} = 2\xi_9,$$
$$\xi_{13} + \xi_{15} = 2\xi_7, \quad \xi_{13} - \xi_{15} = 2\xi_{11}. \tag{8}$$

The nonlinear terms discussed in this paper should be observable in the relaxation and propagation of sound in liquid crystals. The identities in (8) can be verified experimentally.

We thank Shen Juelian (沈觉琏) and other colleagues for helpful discussions.

References

[1] Lam, L., *Z. Physik*, **B 27** (1977), 349.
[2] Liquid Crystal Group of Institute of Physics (ed.), *Guide to Literatures on Liquid Crystals*, published by Institute of Physics, Academia Sinica, November, 1978.
[3] Lam L. & Lax, M., *Phys. Fluids*, **21** (1978), 9.
[4] McLennan, J. A., *Phys. Rev.*, **A 8** (1973), 1479.
[5] Moritz, E. & Franklin, W., *Phys. Rev.*, **A 14** (1976), 2334.
[6] Lam, L., *Z. Physik*, **B 27** (1977), 101.
[7] ———, *ibid*, **B 27** (1977), 273.
[8] De Jeu, W. H., *Phys. Lett.*, **69A** (1978), 122.
[9] Edelen, D. G. B., *Int. J. Engng. Sci.*, **12** (1974), 397.
[10] Lavenda, B. H., *Thermodynamics of Irreversible Processes*, McMillan Press, New York, (1978).
[11] Spencer, A. J. M., Theory of Invariants, in A. C. Eringen (ed.), *Continuum Physics*, Academia Press, 1 (1971), 239—353.