LE JOURNAL DE PHYSIQUE-LETTRES

J. Physique — *LETTRES* 42 (1981) L- 327- L-330

15 JUILLET 1981, PAGE L-327

Classification
Physics Abstracts
47.30 — 61.30

Hydrodynamics of cholesteric liquid crystals with impurities

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(Reçu le 9 mars 1981, révisé le 1^{er} juin, accepté le 2 juin 1981)

Résumé. — Nous présentons les équations hydrodynamiques pour les cristaux liquides cholestériques et nous proposons une interprétation de l'expérience de Janossy à l'aide des équations hydrodynamiques.

Il apparaît que les termes réversibles qui couplent le vecteur déplacement et la densité d'impulsion (qui existent déjà sans les impuretés) sont à l'origine des observations expérimentales.

Abstract. — We present the hydrodynamic equations for cholesteric liquid crystals with impurities and we propose an explanation for the experiment of Janossy in terms of the hydrodynamic equations.

As it turns out the reversible coupling terms between the displacement vector and the density of linear momentum (which are already present without impurities) are responsible for the experimental observations.

1. Introduction. — In two recent publications [1, 2] we derived the nonlinear reversible hydrodynamic equations for various types of liquid crystals : nematics, smectics A, cholesterics and smectics C. In discussing the hydrodynamic equations for cholesteric liquid crystals we have pointed out that there exist various reversible couplings due to the fact that there is no mirror symmetry in this type of liquid crystals. This property leads to the possibility to introduce a pseudo scalar quantity q_0 (i.e. a quantity which is odd under spatial inversion and even under time reversal) and which is connected to the pitch p_0 via $q_0 = 2 \pi p_0^{-1}$.

The purpose of the present letter is the generalization of the results given in [1, 2] by including the hydrodynamic effects of impurities and to explain very recent experimental results of Janossy [3] within the hydrodynamic equations presented. Especially it will be pointed out that the reversible couplings which produce probably the effects observed in the experiments are already present in cholesterics without impurities and are not due to a diffusomechanical effect. Furthermore, we propose additional experiments to determine all the phenomenological para-

meters involved. Throughout the paper we assume that the impurities have approximately the same size as the pitch.

As has already been discussed in reference [3], the hydrodynamic properties of particles which have a size very large compared to the pitch resemble those of particles in a smectic A liquid crystal. In the case of impurities whose size is much smaller than the pitch, the surrounding of the particles can be visualized approximately as a nematic phase. We study exclusively bulk effects.

2. Nonlinear hydrodynamic equations for cholesteric liquid crystals with impurities. — In addition to the conserved quantities already present in cholesterics without impurities, namely energy density ε (or entropy density σ), density ρ and density of linear momentum $\mathbf{g}$ we have as additional conserved quantity the concentration c of the particles which are immersed in the cholesteric liquid crystal. For the broken symmetries there is no change compared to usual cholesterics : the only hydrodynamic variable characterizing a spontaneously broken symmetry, is

the component of the displacement vector $\mathbf{R}$ parallel to the helical (or pitch) axis $\hat{\mathbf{p}}$.

The Gibbs relation for the hydrodynamic variables takes the form

$$d\varepsilon = T d\sigma + \mu d\rho + \mathbf{v} \cdot d\mathbf{g} + \varphi_{ij} d\nabla_j R_i + \psi_{ijk} d\nabla_k \nabla_j R_i + \\ + Z dc + \tau_i d\nabla_i \rho + \pi_i d\nabla_i \sigma + \Lambda_i d\nabla_i c \quad (2.1)$$

and for the pressure we have

$$p = -\varepsilon + T\sigma + \mu\rho + \mathbf{v} \cdot \mathbf{g} + Zc + \tau_i \nabla_i \rho + \\ + \pi_i \nabla_i \sigma + \Lambda_i \nabla_i c. \quad (2.2)$$

The corresponding conservation and quasi conservation laws read

$$\begin{aligned} \frac{d\rho}{dt} + \rho \nabla_i v_i &= 0 \\ \frac{d\sigma}{dt} + \sigma \nabla_i v_i + \nabla_i \left(\frac{q_i}{T} \right) &= \frac{R}{T} \\ \frac{dg_i}{dt} + g_i \nabla_j v_j + \nabla_j \sigma_{ij} + \nabla_i p &= 0 \\ \frac{d\varepsilon}{dt} + (\varepsilon + p) \nabla_i v_i + \nabla_j (\sigma_{ij} v_i) + \nabla_i j_i^e + v_i \nabla_i p &= 0 \\ \frac{dc}{dt} + c \nabla_i v_i + \nabla_i j_i^c &= 0 \\ \frac{dR_i}{dt} - p_i p_j v_j + Z_i &= 0. \end{aligned} \quad (2.3)$$

For the equations of state which express the thermodynamic forces in terms of the variables we find

$$\begin{aligned} Z_i^R &= Z_i^R(A) - \frac{p_i p_k}{2 q_0} (\text{curl } \mathbf{v})_k + g_{kj} q_0 p_i \nabla_k v_j \\ j_i^e &= q_0 \tilde{\lambda}_{imk} \nabla_m v_k \\ \sigma_{ij}^R &= \sigma_{ij}^R(A) - \left(\frac{1}{q_0} \varepsilon_{ijk} + p_k q_0 g_{ji} \right) (\nabla_l \varphi_{kl} - \nabla_l \nabla_p \psi_{kpi}) + \beta \varepsilon_{ijk} p_s R_m (\nabla_k R_m) \nabla_l \varphi_{si} - q_0 \lambda_{kji} \nabla_k \tilde{T} - q_0 \tilde{\lambda}_{kji} \nabla_k \tilde{Z} \end{aligned} \quad (2.6)$$

where $\sigma_{ij}^R(A)$, $Z_i^R(A)$ are the reversible parts of the stress tensor and the current of the quasi conservation law for smectics A (cf. Ref. [1]). The tensors λ_{ijk} , $\tilde{\lambda}_{ijk}$ are of the form

$$\xi_{ijk} = \xi_1 p_i p_j p_k + \xi_2 (p_i \delta_{jk}^r + p_j \delta_{ik}^r) + \xi_3 p_k \delta_{ij}^r + \xi_4 \frac{1}{2} [p_i p_l \varepsilon_{ljk} + p_j p_l \varepsilon_{lik}].$$

From inspection of the first equation (2.6) we see immediately that there exist reversible couplings in cholesterics which are due to the lack of mirror symmetry. Like in any hydrodynamic system there exist further terms if we allow for terms which involve even more gradients; for Z_i we obtain e.g.

$$Z_i = p_i \psi_{jk} \nabla_j \Omega_k + p_i \chi_{jkl} \nabla_j A_{kl} \quad (2.7)$$

$$\begin{aligned} v_i &= \frac{\partial F}{\partial g_i} \Big|_{\dots} \\ \tilde{\mu} &= \mu - \nabla_i \tau_i = \frac{\partial F}{\partial \rho} \Big|_{\dots} - \nabla_i \frac{\partial F}{\partial \nabla_i \rho} \Big|_{\dots} \\ \tilde{T} &= T - \nabla_i \pi_i = \frac{\partial F}{\partial \sigma} \Big|_{\dots} - \nabla_i \frac{\partial F}{\partial \nabla_i \sigma} \Big|_{\dots} \\ \tilde{Z} &= Z - \nabla_i \Lambda_i = \frac{\partial F}{\partial c} \Big|_{\dots} - \nabla_i \frac{\partial F}{\partial \nabla_i c} \Big|_{\dots} \\ \varphi_{ij} &= p_i p_k \frac{\partial F}{\partial \nabla_j R_k} \Big|_{\dots} \\ \psi_{ijk} &= p_i p_p \delta_{jq}^r \delta_{rk}^r \frac{\partial F}{\partial \nabla_q \nabla_r R_p} \Big|_{\dots} \end{aligned} \quad (2.4)$$

where the energy density is given by

$$\begin{aligned} F &= F(\delta c = 0) + \zeta_1 (\delta \sigma) (\delta c) + \zeta_2 (\delta \rho) (\delta c) + \zeta_3 (\delta c)^2 + \\ &+ \gamma_3 (p_i p_j \nabla_i R_j - \frac{1}{2} p_m p_j (\nabla_i^r R_m) (\nabla_i^r R_j)) \delta c \\ &+ \chi_3 q_0 p_i p_j p_k (\nabla_i R_k) (\nabla_j c) \end{aligned} \quad (2.5)$$

and where

$$\delta_{ij}^r = \delta_{ij} - p_i p_j.$$

In equation (2.5) $F(\delta c = 0)$ has been given in references [1, 2]; the terms involving ζ_1 , ζ_2 , ζ_3 , γ_3 and χ_3 are novel.

To close the system of our hydrodynamic equations we have to relate the currents in the conservation and quasi conservation laws to the thermodynamic conjugate quantities.

For the reversible currents which have to satisfy the requirement of vanishing entropy production ($R \equiv 0$) we have

where

$$\begin{aligned} \psi_{jk} &= \psi_1 p_j p_k + \psi_2 p_m \varepsilon_{mjk} \\ \chi_{jkl} &= \chi_1 p_j p_k p_l + \chi_2 p_j \delta_{kl}^r + \chi_3 (p_k \delta_{jl}^r + p_l \delta_{jk}^r) + \\ &+ \chi_4 (p_k p_m \varepsilon_{mjl} + p_l p_m \varepsilon_{mjk}) \end{aligned}$$

and

$$\begin{aligned} A_{kl} &= \frac{1}{2} (\nabla_k v_l + \nabla_l v_k) \\ \Omega_k &= \frac{1}{2} (\text{curl } \mathbf{v})_k. \end{aligned}$$

Contrary to the terms involving g_{kl} the contributions presented in (2.7) exist in smectics A as well as in cholesterics (of course, the tensors χ_{jkl} and ψ_{jk} give rise to corresponding expressions in the stress tensor!).

For the irreversible currents we find

$$\begin{aligned} Z_i^D &= \xi \nabla_j \varphi_{ij} + p_i p_j (\tilde{\xi} \nabla_j \tilde{Z} + \xi \nabla_j \tilde{T}) \\ \sigma_{ij}^D &= \eta_{ijkl} A_{kl} \\ j_i^c &= \chi'_\perp \delta_{ij}^r \nabla_j \tilde{Z} + \chi'_\parallel p_i p_j \nabla_j \tilde{Z} + p_i p_j \tilde{\xi} \nabla_k \varphi_{jk} + \\ &\quad + \xi_\perp \delta_{ij}^r \nabla_j \tilde{T} + \xi_\parallel p_i p_j \nabla_j \tilde{T} \\ j_i^{\sigma D} &= \kappa_\perp \delta_{ij}^r \nabla_j \tilde{T} + \kappa_\parallel p_i p_j \nabla_j \tilde{T} + p_i p_j \tilde{\xi} \nabla_k \varphi_{jk} + \\ &\quad + \xi_\perp \delta_{ij}^r \nabla_j \tilde{Z} + \xi_\parallel p_i p_j \nabla_j \tilde{Z}. \end{aligned} \quad (2.8)$$

The equations (2.8) present a slight generalization of the results previously given by P. C. Martin, O. Parodi and P. S. Pershan [4] (cf. also de Gennes' book [5]). We have included the concentration c in our description which brings along five new dissipative contributions $\tilde{\xi}$, $\xi_\parallel$, $\xi_\perp$, $\chi'_\perp$ and $\chi'_\parallel$. In addition we have expanded the dissipative currents (as well as the reactive ones in equations (2.6)) into the «renormalized» thermodynamics forces given in (2.4). Concerning the irreversible coefficients we restrict our considerations to the linear domain since we are mainly interested in the reversible effects in the present paper.

Thus we have derived the hydrodynamic equations for cholesteric liquid crystals with impurities and we are therefore prepared for the application to specific examples. In the following we discuss as an example which is unique to cholesterics the experiments of I. Janossy.

3. Interpretation of Janossy's experiment in the framework of hydrodynamics. — Janossy considered a planar texture of cholesterics in a torsional shear apparatus where the shear was produced by rotating one of the horizontal discs while keeping the other one fixed. The axis of rotation was parallel to the axis of the cholesteric helices, $\hat{\mathbf{p}}$.

He observed that for one sense of rotation the impurities (small glass spheres) were accumulated near the horizontal boundaries of the apparatus whereas for the other sense of rotation the particles accumulated near the rotation axis. In addition Janossy observed that the decrease of the concentration in time was homogeneous in space [cf. Fig. 1 of Ref. [3]].

I. Janossy explained his experiment by means of a diffusomechanical effect. By inspection of the second equation of (2.6), however, we find that a rotation about $\hat{\mathbf{p}}$ (described by $\frac{1}{2} p_k (\text{curl } \mathbf{v})_k$) produces only a concentration current parallel to $\hat{\mathbf{p}}$ (proportional to the parameter $\tilde{\lambda}_4$). But such an upward or downward concentration current cannot explain the accumulation of impurities in horizontal directions.

Therefore we propose a different explanation of the experiment which runs as follows.

We take the normal to the discs to be the z -direction which is thus parallel to the axis of rotation. Therefore we have for the angular velocity

$$\boldsymbol{\Omega} = \frac{1}{2} (\text{curl } \mathbf{v}) = \frac{1}{2} \begin{pmatrix} 0 \\ 0 \\ \partial_x v_y - \partial_y v_x \end{pmatrix}. \quad (3.1)$$

As has been noticed by Janossy the derivative of the angular velocity in z -direction is held fixed during the experiment

$$\frac{\partial \Omega_z}{\partial z} = \text{const}. \quad (3.2)$$

Furthermore we assume in what follows $v_z \equiv 0$, $R_i \equiv 0$ and we look for a solution of equations (2.3)-(2.7) which meets these requirements.

Then we find from (2.6), (2.7)

$$(\partial_x v_x + \partial_y v_y) g_1 q_0 = \frac{\Omega_z}{2 q_0} + \psi_1 \frac{\partial \Omega_z}{\partial z}. \quad (3.3)$$

Equation (3.3) shows that a rotation (either homogeneous or inhomogeneous) can give rise to a transversal divergence of the velocity and this effect is solely due to the existence of the pseudo scalar q_0 in cholesterics (with or without impurities) and not to a «diffusomechanical coupling». From the last equation of (2.3) we find

$$\begin{aligned} \frac{\partial c}{\partial t} &= - \text{div} (c \mathbf{v}) - \nabla_i j_i^c = - c \text{div } \mathbf{v} - \mathbf{v} \cdot \text{grad } c - \\ &\quad - q_0 \tilde{\lambda}_{imk} \nabla_i \nabla_m v_k. \end{aligned} \quad (3.4)$$

We refrain from a discussion of the other dynamic equations because the experiments yield only information about the initial time evolution of the concentration and this behaviour can be extracted from the equation for c .

The second term on the right hand side of equation (3.4) vanishes according to the results of Janossy's experiment (cf. Fig. 1 of Ref. [3]). Thus we end up with

$$\frac{\partial c}{\partial t} = - c \text{div } \mathbf{v} - q_0 \tilde{\lambda}_{imk} \nabla_i \nabla_m v_k. \quad (3.5)$$

Combining (3.5) and (3.3) we have ($v_z \equiv 0$)

$$\frac{\partial c}{\partial t} = - \left(\frac{\Omega_z}{2 q_0^2 g_1} + \frac{\psi_1}{g_1 q_0} \frac{\partial \Omega_z}{\partial z} \right) c - \tilde{\lambda}_4 \frac{1}{g_1 q_0} \frac{\partial \Omega_z}{\partial z}. \quad (3.6)$$

As has already been discussed by I. Janossy his experiment is accessibly to a continuum type description only for short times whereas for long times the dynamics of this experiment is dominated by defects.

Therefore we obtain from (3.6) by comparison with the notation used by I. Janossy

$$\left(c(t) = c(0) \exp \left[- \frac{2 \Omega}{L} \lambda t \right] \right),$$

$$\frac{\Omega}{L} 2 \lambda = - \left(\frac{\Omega_z}{2 q_0^2 g_1} + \frac{\psi_1}{g_1 q_0} \frac{\partial \Omega_z}{\partial z} \right). \quad (3.7)$$

To arrive at equation (3.7) one should keep in mind that Ω/L in Janossy's notation is identical to $\partial \Omega_z / \partial z$ which is constant in the experiment whereas the expression involving Ω_z has to be averaged over the sample.

Keeping this in mind we get from (3.7)

$$\lambda = - \frac{\langle \Omega_z \rangle}{4 q_0^2 g_1} \frac{L}{\Omega} - \frac{\psi_1}{2 g_1 q_0}. \quad (3.8)$$

By inspection of equation (3.8) we see that the second term on the right hand side changes sign if the sign of the pitch is reversed, an effect observed by Janossy.

The first term on the r.h.s., however, is even under reversal of the pitch and such a term has been omitted in the Ansatz of Janossy (cf. Eq. (2) of [3]). If the first term is small it may be masked by the second. Nevertheless, it is highly desirable to repeat the experiments

for $\Omega_z = \text{const.}$, $\partial \Omega_z / \partial z = 0$ because in this case the hydrodynamic theory would give an effect which is independent of the sign of the pitch and which would allow the direct determination of the reversible coefficient g_1 . (The coefficients g_i are already involved in the convective instabilities of cholesterics [6, 7], though in a more intricate manner.) In addition it should be possible to carry out an experiment with $\langle \Omega_z \rangle \equiv 0$ (e.g. both discs rotating with an angular velocity which is of equal magnitude but of different sign) thus yielding ψ_1/g_1 .

Furthermore it should be stressed (cf. Eqs. (3.6), (3.7)) that a change of sign of Ω_z (or $\partial \Omega_z / \partial z$) leads to a change of $\partial c / \partial t$ and that therefore a change of the flow direction of the particles (the transverse drift of the particles) occurs because $\nabla_i v_i$ changes sign too in this case.

To summarize we have presented the hydrodynamic equations for cholesteric liquid crystals with impurities (which are assumed to be of approximately the same size as the pitch) and we propose an interpretation of a recent experiment of I. Janossy in the framework of the hydrodynamic equations showing in an impressive manner the unique properties of cholesterics due to the lack of mirror symmetry and thus the simultaneous existence of a pseudo scalar quantity.

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