J. Physique **41** (1980) 437-442 MAI **1980**, PAGE 437

Classification
Physics Abstracts
61.30 — 47.20

Shear flow induced propagating domains in cholesterics

I. Jánossy

Central Research Institute for Physics, H-1525 Budapest, P.O.B. 49, Hungary

(*Reçu le 10 septembre 1979, révisé les 26 octobre et 13 décembre 1979, accepté le 21 janvier 1980*)

Résumé. — Il est montré, qu'au-dessus du seuil du taux de cisaillement, un écoulement déstabilise la texture planaire des cholestériques dont le pas est inférieur à l'épaisseur de l'échantillon. Les perturbations de la configuration planaire se propagent à une vitesse proportionnelle au taux de cisaillement. Dans cet article, nous présentons une théorie simplifiée de ces phénomènes qui est comparée aux résultats expérimentaux.

Abstract. — It is shown, that shear flow destabilizes the planar texture of cholesterics of pitch small compared to sample thickness above a threshold shear rate. In addition, in the presence of the shear, perturbations of the planar configuration propagate with a velocity proportional to the shear rate. In this paper we present a simplified theory of these effects and compare our results with experimental data.

1. **Introduction.** — The theory of the distortion of a planar cholesteric structure by a shear flow (direction of flow perpendicular to the helical axis) has been discussed by different authors. Leslie gave the fundamental equations of the problem [1]; Prost considered the linearized equations and derived the flow induced flexoelectric polarization [2]; Kini calculated the effective viscosity as a function of the shear rate [3]. In all these considerations *homogeneous* distortions were investigated, i.e. it has been supposed that the director remains constant in the planes perpendicular to the unperturbed helical axis; it changes only along this axis.

As is well known, in liquid crystals, external forces often induce *periodic* structures in oriented samples. E.g. in nematics periodic domains can be induced by electric field, temperature gradient or by shear flow [4]. In cholesterics the application of a magnetic or an electric field parallel to the helical axis also produces a periodic distortion [5, 6]. The question arises whether a shear flow can induce a periodic deformation in cholesterics. The discussion of this problem is the main purpose of the present paper.

As it will be shown, the answer to our question is : yes. We shall investigate small deformations of the helical structure which are periodic along the direction perpendicular to the flow and to the helical axis. It turns out, that at a critical shear rate the planar texture becomes unstable with respect to such perturbations. Furthermore, these perturbations propagate with a certain velocity along the direction of the periodicity. The velocity of propagation is essentially proportional to the shear rate and reverses its direction when the flow direction is reversed.

In section 2 a somewhat simplified calculation of these effects are presented. We have extended Hurault's method, which he used for describing field induced deformations [6]. Simple expressions are derived for the critical shear rate and the propagation velocity of the periodic deformations.

In section 3 we compare our theoretical predictions with some experimental data. It is demonstrated, that the instability, discussed here may correspond to the textural instability observed previously in cholesteryl oleyl carbonate [7]. The observed instability however was found to be discontinuous and no regular structure could be observed above the transition. Nevertheless, propagating domains can be observed by superposing a shear flow on an electric field induced periodic structure. We carried out such measurements on MBBA, doped with a small amount of cholesteric. The sign and order of magnitude of the measured propagation velocity agrees with the calculated one.

2. **Theory of the instability.** — We investigate a planar cholesteric structure of pitch P and thickness L, sandwiched between two glass plates. The lower plate moves with a velocity V in the y direction. The helical axis is along z, i.e. perpendicular to the plates.

In the undistorted helix the director is

$$n_x = \cos \psi_0 , \quad n_y = \sin \psi_0 , \quad n_z = 0$$

with

$$\psi_0 = t_0 z\,, \quad t_0 = 2\pi/P\,.$$

A small perturbation can be characterized by two variables, n_z and φ, such that

$$n_x = \cos(\psi_0 + \varphi) \approx \cos\psi_0 - \varphi\sin\psi_0$$

$$n_y = \sin(\psi_0 + \varphi) \approx \sin\psi_0 + \varphi\cos\psi_0\,.$$

First let us consider the viscous torque, which is in general given as [8]

$$\underline{\Gamma}^{(v)} = \underline{n} \times \left(\gamma_1\left\{\frac{d\underline{n}}{dt} - \underline{v} \times \underline{n}\right\} + \gamma_2 \underline{An}\right)$$

with

$$\underline{v} = \frac{1}{2}\,\mathrm{rot}\,\underline{v}\,; \quad A_{ij} = \frac{1}{2}\left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i}\right);$$

$$\gamma_1 = \alpha_3 - \alpha_2\,; \quad \gamma_2 = \alpha_3 + \alpha_2\,.$$

The following two simplifications are made in the calculation of $\underline{\Gamma}^{(v)}$.

1. Since generally $|\alpha_2| \gg |\alpha_3|$, we assume that $\alpha_3 = 0$, i.e. $\gamma_2 = -\gamma_1 = \alpha_2$. As can be seen e.g. from ref. [2], if $\alpha_3 = 0$, in our geometry the twisted structure has no homogeneous deformation. However, periodic deformations can still occur.

2. We take for the velocity

$$v_x = v_z = 0\,; \quad v_y = sz \quad \text{with} \quad s = V/L\,.$$

As pointed out by Leslie [1] the real situation is that there is a transverse flow ($v_x \neq 0$) and the shear rate dv_y/dz depends on z even for an undistorted helix. In the present paper we disregard these effects which would make the calculations much more complicated.

Using these assumptions the components of $\underline{\Gamma}^{(v)}$ parallel and perpendicular to the helical axis for small perturbations are respectively :

$$\Gamma_z^{(v)} = \alpha_2\left(sn_z\cos\psi_0 - \frac{d\varphi}{dt}\right);$$

$$\Gamma_\perp^{(v)} = -\alpha_2\frac{dn_z}{dt}\,.$$

In the following, as in ref. [6], periodic tilts of the cholesteric planes will be investigated, with periodicity along x. In this case n_z can be expressed as $n_z = \theta\cos\psi_0$. Using the θ variable :

$$\Gamma_z^{(v)} = \alpha_2\left(s\cos^2\psi_0\,\theta - \frac{d\varphi}{dt}\right), \tag{1}$$

$$\Gamma_\perp^{(v)} = -\alpha_2\cos\psi_0\frac{d\theta}{dt}\,. \tag{2}$$

The elastic torques were derived by Hurault [6]. In the case $K_1 = K_3$ they are

$$\Gamma_z^{(e)} = -K_3\left(\frac{\partial^2\varphi}{\partial x^2} + t_0\frac{\partial\theta}{\partial x}\right) - K_2\left(\frac{\partial^2\varphi}{\partial z^2} + t_0\cos 2\psi_0\frac{\partial\theta}{\partial x}\right) + \frac{1}{2}(K_3 - K_2)\sin 2\psi_0\frac{\partial^2\theta}{\partial x\,\partial z}\,; \tag{3}$$

$$\Gamma_\perp^{(e)}\cos\psi_0 = 2K_3\cos^2\psi_0\,t_0\left(t_0\theta + \frac{\partial\varphi}{\partial x}\right) - \frac{1}{2}(K_2 - K_3)\sin 2\psi_0\frac{\partial^2\varphi}{\partial x\,\partial z} - \frac{K_2}{4}\sin^2 2\psi_0\frac{\partial^2\theta}{\partial x^2} + K_3\left(t_0\sin 2\psi_0\frac{\partial\theta}{\partial z} - \cos^2\psi_0\frac{\partial^2\theta}{\partial z^2} - \cos^4\psi_0\frac{\partial^2\theta}{\partial x^2}\right). \tag{4}$$

The equation of motion of the director is

$$\underline{\Gamma}^{(v)} + \underline{\Gamma}^{(e)} = 0\,. \tag{5}$$

First of all we show that in the presence of a shear flow, perturbations are propagating. For this purpose let us consider a small perturbation which has the form at $t = 0$,

$$\theta = \theta_0(z)\sin kx\,. \tag{6}$$

From the z component of the equation of motion, $\Gamma_z^{(v)} + \Gamma_z^{(e)} = 0$ we have

$$\alpha_2\frac{d\varphi}{dt} = \alpha_2 s\theta\cos^2\psi_0 + \Gamma_z^{(e)}\,. \tag{7}$$

The first term on the right hand side of eq. (7) is proportional to $\sin kx$, while the second one contains terms like $\partial\theta/\partial x$, which are proportional to $\cos kx$. As a consequence φ must have in general the form

$$\varphi = \varphi_c(z)\cos kx + \varphi_s(z)\sin kx\,. \tag{8}$$

From the equation $\Gamma_\perp^{(v)} + \Gamma_\perp^{(e)} = 0$ we get using the above relation

$$-\alpha_2\cos^2\psi_0\frac{d\theta}{dt} = \Gamma_\perp^{(e)}\cos\psi_0 = a(z)\sin kx + b(z)\cos kx\,. \tag{9}$$

Integrating eq. (9) by t it follows that for $t > 0$, θ will have the form

$$\theta(t) = \theta_s(z, t)\sin kx + \theta_c(z, t)\cos kx = \theta_0(z, t)\sin(kx + \delta(t))\,.$$

This means that the spatial phase of perturbation (6) will be shifted in time, i.e. the deformation propagates in the x direction.

In view of the above considerations we investigate perturbations of the form

$$\theta = \theta_0 \sin(kx - \omega t) \cos q_0 z$$
$$\varphi = (\varphi_c \cos(kx - \omega t) + \varphi_s \sin(kx - \omega t)) \cos q_0 z \tag{10}$$

with $q_0 = \pi/L$.

As the elastic torques always stabilize the planar configuration, at small shear rates this perturbation decays, i.e. θ_0, φ_c, φ_s relax to zero. At higher shear rates the viscous torque may destabilize the planar texture, and θ_0, φ_c, φ_s may increase in time. The threshold of the instability can be calculated from the assumption that θ_0, φ_c, φ_s have stationary values.

In the following (as in the theory of field induced deformations) we restrict ourselves to the limit

$$q_0 \ll k \ll t_0 \tag{11}$$

furthermore θ_0 and $\Gamma_\perp \cos\psi_0$ will be replaced by their averages over one period of the helix.

In this case the last term in $\Gamma_z^{(e)}$, which is proportional to $kq_0\theta_0$ can be neglected. The equation $\Gamma_z^{(v)} + \Gamma_z^{(e)} = 0$ gives

$$\varphi_c = \varphi_c^{(0)} + \varphi_c^{(1)} \cos 2\psi_0 \,; \qquad \varphi_s = \varphi_s^{(0)} + \varphi_s^{(1)} \cos 2\psi_0$$

with

$$\varphi_c^{(0)} = \lambda \frac{1 - AB}{1 + B^2}\theta_0 \,; \qquad \varphi_c^{(1)} = \eta \frac{1 - CD}{1 + D^2}\theta_0 \,;$$
$$\varphi_s^{(0)} = -\lambda \frac{A + B}{1 + B^2}\theta_0 \,; \qquad \varphi_s^{(1)} = -\eta \frac{C + D}{1 + D^2}\theta_0 \,, \tag{12}$$

where

$$\lambda = \frac{t_0/k}{1 + K_2 q_0^2/K_3 k^2} \,; \qquad \eta = \frac{k/4\,t_0}{1 + K_3 k^2/4\,K_2 t_0^2} \,;$$

$$A = \frac{\alpha_2 s}{2K_3 t_0 k} \,; \quad B = -\frac{\alpha_2 \omega}{K_3 k^2 + K_2 q_0^2} \,; \quad C = \frac{\alpha_2 s}{2K_2 t_0 k} \,; \quad D = -\frac{\alpha_2 \omega}{4t_0^2 K_2 + k^2 K_3} \,.$$

Now let us consider the equation

$$\Gamma_\perp^{(v)} + \Gamma_\perp^{(e)} = 0 \,. \tag{13}$$

Multiplying (13) by $\cos\psi_0$ and taking the average over one period of the helix, the terms proportional to $\cos kx$ and $\sin kx$ give respectively :

$$\frac{\alpha_2 \omega}{2}\theta_0 + t_0 k K_3 \varphi_s^{(0)} + \frac{t_0 k}{2} K_2 \varphi_s^{(1)} = 0 \,; \tag{14}$$

$$K_3(t_0^2\theta_0 - kt_0\varphi_c^{(0)}) + \frac{1}{8}(3K_3 + K_2) k^2 \theta_0 - \frac{t_0 k}{2} K_2 \varphi_c^{(1)} = 0 \,. \tag{15}$$

With the help of eq. (12) $\varphi_s^{(0)}$ and $\varphi_s^{(1)}$ can be eliminated from eq. (14). Thus we obtain a relation between s and ω :

$$\frac{B}{2}(K_3 k^2 + K_2 q_0^2) + t_0 k K_3 \lambda \frac{A + B}{1 + B^2} + \frac{t_0 k K_2}{2}\eta \frac{C + D}{1 + D^2} = 0 \,.$$

Assuming that ω is not extremely large (see later) we have $D^2 \ll 1$. Using inequality (11) we get

$$A \approx -B\left(1 + \frac{3}{8}\frac{k^2}{t_0^2}(1 + B^2)\right). \tag{16}$$

In the limit $q_0 \ll k \ll t_0$ the relation between ω and s becomes linear :

$$\omega = \frac{1}{2} s \frac{k}{t_0} \,, \tag{17}$$

or for the propagation velocity

$$v_d = \omega/k = \frac{1}{2}\frac{s}{t_0} \,.$$

Eliminating $\varphi_c^{(0)}$ and $\varphi_c^{(1)}$ from eq. (15) and using eq. (16), we get a condition for s, i.e. for the threshold shear rate. In the same limit as above the result is

$$s(k) = \pm 2\sqrt{\frac{8}{3}\frac{K_3}{\alpha_2}} t_0 \left(\frac{K_2}{K_3}\frac{t_0^2 q_0^2}{k^2} + \frac{3}{8}k^2\right)^{1/2} . \tag{18}$$

$s(k)$ is minimum for

$$k_m^2 = \sqrt{\frac{8 K_2}{3 K_3}} \mid t_0 \mid q_0 .$$

This is the same result as that obtained for field effects. Using this result, the threshold is

$$s_{th} = \pm 4 \frac{\sqrt{K_2 K_3}}{\alpha_2} t_0 \sqrt{\mid t_0 \mid q_0} . \tag{19}$$

The main results of this section are expressed by eqs. (17) and (19), which provide the propagation velocity and the threshold of the instability respectively.

The above theory is readily extended to the case when a magnetic or electric field parallel to the helical axis and the shear flow act simultaneously. As the field exerts a torque proportional to θ it is only necessary to modify eq. (15) which now becomes (for a magnetic field) :

$$K_3(t_0^2 \theta_0 - k t_0 \varphi_c^{(0)}) + \frac{1}{8}(3 K_3 + K_2) k^2 \theta_0 - \frac{t_0 k}{2} K_2 \varphi_c^{(1)} - \frac{1}{2} \chi_a H^2 \theta_0 = 0 . \tag{20}$$

As eq. (15) has not been used to derive the connection between ω and s, eq. (16) is unchanged. In place of eq. (18) we obtain :

$$s(k, H) = \pm 2 \sqrt{\frac{8}{3} \frac{K_3}{\alpha_2}} t_0 \left(\frac{K_2}{K_3} \frac{t_0^2 q_0^2}{k^2} + \frac{3}{8} k^2 - \frac{1}{2} \frac{\chi_a H^2}{K_3} \right)^{1/2} .$$

The minimum is again for

$$k_m^2 = \sqrt{\frac{8 K_2}{3 K_3}} \mid t_0 \mid q_0 .$$

The threshold as a function of the magnetic field is

$$s_{th}(H) = s_{th}(0) (1 - H^2/H_{th}^2)^{1/2} \tag{21}$$

where H_{th} is the threshold for the magnetic field induced instability :

$$H_{th}^2 = \frac{1}{\chi_a} \sqrt{6 K_2 K_3}\, q_0 \mid t_0 \mid ,$$

$s_{th}(0)$ is given by eq. (19).

A similar equation can be derived for an electric field :

$$s_{th}(U) = s_{th}(0) (1 - U^2/U_{th}^2)^{1/2} \tag{22}$$

where U_{th} is the threshold voltage for the electric field induced instability, given in ref. [6].

From eq. (22) it follows, that the planar structure becomes unstable if

$$\alpha(U, s) = \frac{U^2}{U_{th}^2} + \frac{s^2}{s_{th}^2} > 1$$

where U and s denote the applied voltage and the shear rate respectively. The threshold is given by $\alpha(U, s) = 1$.

We note that eq. (17) is — strictly speaking — only valid for the threshold $\alpha = 1$. On the other hand, measurements of ω can be carried out only slightly above the threshold ($0 < \alpha - 1 \ll 1$). To find an exact relation between ω and s above the threshold, terms nonlinear in the deformation components must be considered also. However for a continuous transition, where just above the threshold the deformation remains small, the nonlinear terms can be neglected when deriving the relation between ω and s. Hence eq. (17) will be a good approximation slightly above the threshold too.

Now let us discuss briefly the influence of the approximations we made when deriving these formulae.

If $\alpha_3 \neq 0$, there will be a homogeneous distortion also. This deformation has no threshold, its amplitude is in the linear approximation [2]

$$n_z = \theta_h \sin \psi_0$$

with

$$\theta_h = \frac{\alpha_3}{1 + (\alpha_6 + \alpha_3)/\alpha_4} \frac{s}{t_0^2(K_3 + K_1)} .$$

At the threshold for periodic instability given by eq. (19), θ_h is of the order of $\alpha_3/\alpha_2 . \sqrt{q_0/t_0}$. In our limit θ_h becomes very small and the homogeneous deformation is linearly superposed on the periodic distortion.

The influence of the secondary flow may be more serious. It is known in the case of hydrodynamic instabilities in nematics that transverse flow effects modify the threshold considerably [9, 10]. The situation is similar in the present case. The precise treatment of the velocity field should lead to additional linear terms in the viscous torques. However these terms will be comparable or smaller than those one which we considered. As a consequence the order of magnitude of ω and s_{th} should not change.

Finally, it was assumed that

$$D^2 = \left(\frac{\alpha_2 \omega}{4 t_0^2 K_2 + k^2 K_3} \right)^2 \ll 1 .$$

Using eqs. (17) and (19) it can be seen, that at the threshold D^2 is of the order of $(k/t_0)^4 \ll 1$.

3. Comparison with experiments. — As reported previously [7], in cholesteryl oleyl carbonate, with pitch $P \approx 0.3$ μm, sample thickness 50 μm, an instability of the planar texture was observed at

$s_{cr} = 1.5 \times 10^3\ s^{-1}$. Taking for $\sqrt{K_2 K_3} = 5 \times 10^{-7}$ dyn., $\alpha_2 = -1$ poise (typical values for nematics), eq. (19) yields

$$s_{th} = \frac{\sqrt{2}\, 8\, \pi^2}{P \sqrt{L\,|P|}} \frac{\sqrt{K_2 K_3}}{\alpha_2} \approx 5 \times 10^3\ s^{-1} .$$

The order of magnitude is the same for the calculated and measured critical shear rates, however there is a considerable difference between them. This difference may be explained by several facts : the value of material constants may be incorrect ; effect of secondary flow, etc.

There is also another reason for the discrepancy. The calculated threshold refers to a continuous transition, where the amplitude of the distortion remains small just above the threshold. The validity of this assumption cannot be verified from the linearized equations. If this assumption does not hold eq. (19) gives only an upper limit for the threshold.

As a matter of fact in the reported experiment the transitions was found to be discontinuous ; at s_{cr} the planar texture is strongly deformed immediately. This fact may also contribute to the deviation of s_{cr} and s_{th}.

As already mentioned, due to the discontinuous character of the instability, no regular domains could be observed above the threshold. However propagating domains were observed in another experiment by superposing an electric field and shear flow. In this case, with sufficiently low shear rates the distortion increases continuously above the threshold.

In the experiment the same set-up was used as reported in [7]. The material (MBBA doped with 1 % cholesteryl nonanoate) was sandwiched between two disks, and the lower disk was rotated. At the boundaries the director was oriented tangentially. The electric field induced a square pattern. However on switching on the flow the domains became linear resembling the well known Williams domains. The periodicity was along the radial direction. The propagation velocity of the domains was determined by measuring the time in which a number of bright lines intersected the middle point of the cross-hairs in a microscope.

In figure 1 a typical experimental result is shown.

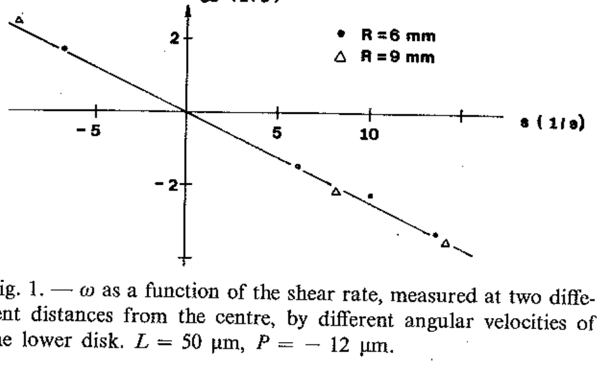

Fig. 1. — ω as a function of the shear rate, measured at two different distances from the centre, by different angular velocities of the lower disk. $L = 50$ μm, $P = -12$ μm.

The precision of the measurements is somewhat limited by the fact that after a short time disinclinations form in the sample, which make further observations impossible. Usually the intersection of 3-4 lines could be registered before the structure became irregular. The points shown on the figure are the average of several measurements. From the points on figure 1, we get

$$\omega / s = -0.25 .$$

As noted in the theoretical part of the paper, the relation between s and ω, given by eq. (17) does not depend on how the periodic structure is induced ; it is valid in the presence of an external field too.

The pitch of the material is $P = -12$ μm (left handed). At $L = 50$ μm the periodicity of the domains was found to be $2\pi/k = 39$ μm. With these data, from eq. (17) the theoretical value is

$$\omega / s = \tfrac{1}{2} k / t_0 = -0.15 .$$

In this case we suggest that the deviation of the theoretical and experimental values is first of all due to secondary flow effects.

From the theory presented in section 2, it follows that the threshold voltage should depend on the presence of the shear. From eq. (22) we get

$$V_{th}(s) = V_{th}(0)\,(1 - s^2/s_{th}^2)^{1/2} . \qquad (23)$$

With the data of MBBA ($\sqrt{K_2 K_3} = 4.5 \times 10^{-7}$ dyn., $\alpha_2 = -0.8$ poise) and $P = -12$ μm, $L = 50$ μm, eq. (19) yields $s_{th} = 21\ s^{-1}$. However we were unable to prove eq. (23) even approximately as the time from switching on the flow till the formation of the disclinations was too short to make any reasonable measurements of the decrease of the threshold voltage.

4. **Conclusion.** — In the paper we have shown that shear flow, above a threshold, destabilizes the planar structure of cholesterics. The threshold is of the order of $K/\alpha_2\, t_0 \sqrt{t_0 q_0}$. This conclusion is supported by experiments.

In the mechanism of the instability there is an important difference compared with other instabilities. In other cases the coupling between a perturbation and the external force leads to a destabilizing torque, which has the same spatial phase as the perturbation itself. In the present case the destabilizing torque is shifted with respect to the perturbation by a quarter-period. As a consequence, perturbations propagate. The velocity of propagation is of the order of s/t_0. This result is also verified experimentally.

The mechanism described here is only possible for optically active media. In other phases (nematic, smectic A), the destabilizing torque or force has for symmetry-reasons the same phase as the perturbation. This is also reflected by the fact, that the propagation velocity is proportional to the pitch (sign included). If a left handed cholesteric were replaced by its right handed isomer the domains would propagate in the opposite direction.

References

[1] LESLIE, F. M., *Mol. Cryst. Liq. Cryst.* **7** (1969) 407.
[2] PROST, J., *J. Physique* **39** (1978) 639.
[3] KINI, U. D., *J. Physique Colloq.* **40** (1979) C3-62.
[4] e.g. DUBOIS-VIOLETTE, E. *et al.* in the *Supplement to Solid State Phys.* (1979).
[5] HELFRICH, W., *J. Chem. Phys.* **55** (1971) 839.
[6] HURAULT, J. P., *J. Chem. Phys.* **59** (1973) 2068.
[7] JÁNOSSY, I., *Ann. Phys.* **3** (1978) 345.
For earlier observations see : POCHAN, J. M., MARSH, D. G., *J. Chem. Phys.* **57** (1972) 1193.
[8] PARODI, O., *J. Physique* **31** (1970) 581.
[9] MANNEVILLE, P. and DUBOIS-VIOLETTE, E., *J. Physique* **37** (1976) 285.
[10] LESLIE, F. M., *J. Phys. D* **9** (1976) 925.