



## Liquid Crystals

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## An experimental investigation of electromechanical coupling in cholesteric liquid crystals

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Cholesteric liquid crystals which have a helical arrangement of oriented chiral molecules are expected to show novel cross couplings between fluxes and forces. The most convincing demonstration of these couplings is through a dynamical effect on the structure. Though Lehmann found a rotation of the structure under a temperature gradient in 1900, there has been no subsequent experiment confirming the same. We argue that it is very difficult to obtain a sufficiently weak anchoring of the director at the solid-cholesteric interface which is a necessary condition for the occurrence of Lehmann rotation. In order to achieve a practically zero anchoring energy at the surface, we have devised a simple technique of floating essentially flat cholesteric drops in the isotropic phase. Using this configuration we study the electromechanical coupling which produces a rotation of the structure under the action of a DC electric field. Using measurements on samples with different values of the pitch the relevant electromechanical coefficient of the materials investigated is found to satisfy the relation  $v_E = -0.6 \times 10^{-12} (q/m^{-1}) J m^{-2}$ , where  $q$  is the wavevector of the helix, whose sign is positive (negative) for a right (left) handed structure, confirming that  $v_E$  is hydrodynamic in origin.

### 1. Introduction

In parity conserving systems, the Curie or Von Neumann principle [1, 2] requires that the symmetry of physical effects must be contained in the causes which give rise to them. In chiral systems like the cholesteric, the Curie principle predicts novel cross couplings between fluxes and forces [3]. The chiral interactions are cooperative in a cholesteric resulting in a macroscopic manifestation of the same. We can hence expect the cross couplings to be relatively large and to lead to clearly observable effects. Indeed Lehmann [4] found such an effect soon after the discovery of liquid crystals. Oseen [5] summarized his observations in the following manner: "He found that in certain cases a substance spread out between two glass surfaces would be put into motion, when influenced by a flow of heat coming from below, during which motion the different drops of liquid seemed to be in violent rotation. Further investigations convinced Lehmann that in this case it was not the drop itself, but the structure, that moved." The thermomechanical effect results in an angular momentum density of the director even though the applied force, viz., the temperature gradient, has the nature of a polar vector.

Leslie [6] and Lubensky [7] have developed hydrodynamic theories of the cholesteric phase which lead to solutions corresponding to Lehmann rotation. We summarize the theoretical results in the next section. As we shall see, the cholesteric is characterized by at least two thermomechanical coefficients. It is obviously very interesting to confirm experimentally the Lehmann rotation phenomenon which arises due to a particular combination of the coefficients. But, to our knowledge, the Lehmann experiment has never been reproduced, though there have been a few attempts including one in our laboratory [8] during the past two decades. Consequently,

alternative experimental geometries have been suggested in the literature for determining the thermomechanical coefficients [9–11].

The only claim of an experimental determination of a thermomechanical coupling coefficient is due to Eber and Janossy [12, 13]. They have used a mixture of two liquid crystals which shows a compensation temperature  $T_c$ . The medium has a left-handed helical structure below  $T_c$ . The pitch increases with increase of temperature to become  $\infty$  and reverse sign at  $T_c$ , i.e., the structure becomes right handed above  $T_c$ . In the experiment, a homeotropically aligned sample near  $T_c$  was subjected to a transverse temperature gradient ( $dT/dx$ ). This results in a tilting of the director. In a linear approximation the tilt angle is given by

$$\theta(z) = \frac{\lambda_{\text{eff}}}{K_{33}} \frac{L^2 - 4z^2}{8} \frac{dT}{dx} \quad (1)$$

where  $L$  is the thickness of the sample,  $K_{33}$  the bend elastic constant. Assuming that the thermomechanical coefficients are independent of the magnitude or sign of  $q$  ( $= 2\pi/P$ ) Eber and Janossy derived

$$\lambda_{\text{eff}} = \lambda_3 + K_{22} dq/dT \quad (2)$$

where  $\lambda_3$  is the thermomechanical coupling coefficient in the notation used in §2 and  $K_{22}$  the twist elastic constant. Eber and Janossy have measured the deformation by an optical technique, and estimated  $K_{22} dq/dT$  by an independent experiment. (1) and (2) can then be used to calculate  $\lambda_3$ . However, as we shall see in the next section, the cross coupling term which arises due to the helical arrangement of the cholesteric should vanish when  $q = 0$ , i.e. at the compensation temperature. (Indeed, the theory requires that  $\lambda_3 \propto q$  and should change sign with that of  $q$ .) Eber and Janossy assumed that  $\lambda_3$  is independent of  $q$  in deriving equations (1) and (2), i.e., they assume in effect that the thermomechanical coefficient is molecular rather than structural in origin (and in that case,  $\lambda_3$  can be expected to be very small). Indeed recently there is some controversy in the literature on the interpretation of the experiments of Eber and Janossy [14–16]. In any case our own results to be presented in §3 clearly show that  $\lambda_3 \propto q$ , confirming that the origin of  $\lambda_3$  is indeed hydrodynamic.

Janossy [17, 18] has conducted another interesting experiment in which a planar aligned cholesteric sample is taken between two discs, one of which is rotated with reference to the other. He then observed that small ( $1\text{--}2\text{ }\mu\text{m}$  thick) dust particles drifted radially. For one sense of rotation, the particles concentrated near the axis of rotation, while for the opposite sense, they drifted away from the axis. He has interpreted the result in terms of a ‘diffuso-mechanical’ coupling, in analogy with the thermomechanical coefficient  $\mu_8$  of §2. However, it has also been suggested that the shear flow induces a secondary flow of the material with the same characteristic features [19] and very careful experiments along with a detailed analysis are necessary to separate the two effects.

Thus, it would appear that apart from the Lehmann experiment, up to now there is no unequivocal demonstration of the cross coupling terms in cholesteric liquid crystals. The most convincing experiment would of course be one which leads to a dynamic instability in the structure rather than a static one. In the present paper, we report on such an experiment leading to a Lehmann rotation of carefully prepared cholesteric drops under the action of a force with a polar character. We have found it convenient to use an electric field rather than a temperature gradient, i.e., we

demonstrate an electromechanical coupling. A preliminary report of this investigation has been published recently [20].

We give a brief summary of the theoretical background in §2. The experimental details and the results are given in §3. We end with some concluding remarks in §4.

## 2. Theoretical background

Lehmann himself tried to explain his observation of the rotation phenomenon in the cholesteric drops [4]. He assumed that the molecules in the drops are arranged such that the structure looks like a turbine blade and noted that the upwelling liquid under the action of the temperature gradient would then set the drop into rotation.

A detailed hydrodynamic theory of cholesteric liquid crystals was developed by Leslie [6, 21]. For a cholesteric subjected to a temperature gradient the entropy production rate reads as

$$T \frac{dS}{dt} = \sigma'_{ji} A_{ij} - g'_i N_i - \frac{q_i T_{,i}}{T}, \quad (3)$$

where the suffixes indicate components in a suitable coordinate system, repeated indices indicating the usual summation convention.  $S$  is the entropy density,  $T$  the temperature,  $\sigma'_{ji}$  the hydrodynamic stress tensor,  $A_{ij}$  the symmetric part of the velocity gradient tensor,  $g'_i$  the intrinsic body force per unit volume,  $N$  the rate of change of director with reference to the background liquid,  $q_i$  the heat flux,  $T_{,i}$  the temperature gradient along the direction  $i$ . The prime indicates that we are considering only the non-equilibrium part. The entropy rate can be considered as a sum of products of fluxes and conjugate forces and as usual, one assumes that the fluxes linearly depend on all the forces. This leads to the following set of relations [21, 9]

$$\sigma'_{ji} = \sigma_{ji}^N + \mu_7 n_j e_{ipq} n_p T_{,q} + \mu_8 n_i e_{jpq} n_p T_{,q}, \quad (4)$$

$$g'_i = g_i^N + \lambda_3 e_{i\lambda k} n_\lambda T_{,k}, \quad (5)$$

$$q'_i = \left( k_1 T_{,i} + k_2 n_j n_i T_{,j} + k_3 e_{ijk} n_j N_k + \frac{k_4}{2} (e_{ijk} n_j n_p + e_{ijp} n_j n_k) A_{kp} \right), \quad (6)$$

where the superscript N stands for nematic, i.e. the corresponding expression is the same as in the nematic phase (Leslie [21]).  $k_1$  and  $k_2$  are the usual anisotropic conductivity coefficients present even in the nematic phase.  $\mu_7$ ,  $\mu_8$ ,  $\lambda_3$ ,  $k_3$  and  $k_4$  are the new coefficients which arise from the helical symmetry of the cholesteric phase. Conservation of angular momentum density requires

$$\lambda_3 = \mu_7 - \mu_8. \quad (7)$$

Note that we have ignored terms incorporating  $\alpha$  another material parameter introduced by Leslie [21] as it does not figure in the angular velocity of the director in the solution corresponding to the Lehmann rotation (see equation 12).

Prost [9] reduced the number of coefficients to two by using the Onsager reciprocal relations which in the present case yield

$$\frac{k_3}{T} = \lambda_3 \quad (8)$$

and

$$\frac{k_4}{T} = -(\mu_7 + \mu_8), \quad (9)$$

where the negative sign is a consequence of the different time reversal symmetries of  $\sigma'_{ji}$  and  $q'_i$ .

de Gennes [22] has pointed out that in general any transport current would lead to similar cross coupling terms and for example if the external field is electric in nature, we get electromechanical coupling coefficients in place of the thermo-mechanical coefficients introduced above. The electric field  $E$  would then correspond to  $-(\nabla T/T)$ . Hereafter the suffix  $E$  indicates electromechanical coefficients. For example, using de Gennes notation,

$$v_E = k_{3E}. \quad (10)$$

We can also note that all the terms in equations (4)–(6) containing the Levicivita tensor  $\epsilon_{ipq}$  change sign when the coordinate system is reflected in a mirror. Then the cross coupling coefficients  $k_3$  and  $k_4$  must also change sign, i.e. they can only have a non-zero value for chiral systems. Since the handedness of the helix changes sign under reflection, the coupling coefficients must be odd functions of  $q$ , and in the simplest approximation, particularly for small values of  $q$ , they are  $\propto q$ . This would of course mean that in a nematic or in a compensated cholesteric with  $q = 0$ , the cross coupling does not exist.

Lubensky [7] has developed a hydrodynamic theory of cholesterics valid for spatial distortions whose wavelength is  $\gg P$ , the pitch. He again gets cross coupling terms similar to those in the Leslie formulation. In his first paper Lubensky [7] dropped the  $k_3$  term, which would lead to a rotation of the cholesteric under the action of a temperature gradient, considering it as a highly improbable behaviour. But in his later paper [26], following the argument of Martin *et al.* [24], he recognized that such a rotation would crucially depend on the boundary conditions. Martin *et al.* [24] developed a general theory of hydrodynamics of layered systems applicable to cholesterics also, while the Lubensky formulation is specifically meant for cholesterics.

Very recently Brand and Pleiner [27] have pointed out that the thermomechanical Lehmann effect has a static contribution in addition to the hydrodynamic one discussed above. However, the former does not exist for the electromechanical case, and we shall not discuss the static contribution in the present paper.

Returning now to the Lehmann experiment, a solution corresponding to the rotation of the structure was derived by Leslie [6] on the basis of his hydrodynamic theory. Let us consider a cholesteric sample with its helical axis along the  $z$  direction sandwiched between two glass plates. If a temperature gradient ( $-\nabla T/T = E$ ) acts along the helical axis, the torque balance equation at any given point yields, using the notation of de Gennes [22]

$$\gamma_1 \frac{d\phi}{dt} = K_{22} \frac{d^2\phi}{dz^2} + v_E \quad (11)$$

where  $\gamma_1 = \alpha_3 - \alpha_2$  is the difference between two of the Leslie viscosity coefficients (note that the sign of  $\gamma_1$  is opposite to that used by Leslie [6]). The above equation can be integrated by using appropriate boundary conditions. An extremely interesting solution is obtained if the anchoring energy for azimuthal orientation is zero. In that case  $d\phi/dz = q_0 (= 2\pi/P)$  at both the boundaries and hence a constant in the sample. The solution is then of the form

$$\phi = q_0 z + \frac{v_E t}{\gamma_1} + C, \quad (12)$$

where  $C$  is a constant of integration. The director then obviously rotates with a constant angular velocity given by  $vE/\gamma_1$ . Leslie thus concluded that the rotation phenomenon observed by Lehmann was a consequence of the cross coupling term  $v$ .

It is obvious that the above analysis requires that (a) the angular velocity is proportional to the temperature gradient ( $E$ ), (b) the sense of rotation should change if  $E$  is reversed, and (c) since the coupling coefficient  $v \propto q$ , the sense of rotation should be opposite for left handed helices ( $q < 0$ ), compared to that for right handed helices ( $q > 0$ ). The phenomenological theory does not put any restrictions on the sign of  $v$  for a given sign of  $q$ . This should essentially depend on the chemical nature of the material used.

The most convincing demonstration of the cross coupling coefficient would be to reproduce the Lehmann rotation experiment and check that the sense of rotation changes with the signs of  $E$  and  $q$ , for the given system. However, as we noted in §1, to our knowledge, the Lehmann experiment has never been reproduced.

We believe the reason for this negative result is that in none of the attempts the anchoring energy at the bounding surfaces was sufficiently weak. We can indeed estimate the allowed upper limit for the anchoring energy if the cholesteric structure has to be set in rotation. To simplify the argument let us assume that the upper surface at  $z = D$  is free and has zero anchoring energy. The lower surface which rests on a glass plate is supposed to have an anchoring energy given by  $(W/2) \sin^2(\phi_0)$  where  $\phi_0$  is the angle made by the director with reference to the easy axis at  $z = 0$ . In this case the restoring torque on the director due to this anchoring energy at the  $z = 0$  surface is  $W \sin \phi_0 \cos \phi_0$  which has a maximum value when  $\phi_0 = \pi/4$ . We could then assume that the surface anchoring can be overcome if the angle  $\phi_0$  exceeds  $\pi/4$  due to the action of the thermomechanical coupling, and the structure could then rotate, though not freely. We can estimate the maximum allowed value of  $W$  by solving the problem in the static limit. The surface torque balance equation is

$$W \sin \phi_0 \cos \phi_0 = K_{22} \left[ \left( \frac{d\phi}{dz} \right)_0 - q_0 \right]. \quad (13)$$

The bulk torque balance equation in the static limit is

$$K_{22} \frac{d^2 \phi}{dz^2} = vE. \quad (14)$$

As we have free a surface at  $z = D$ ;  $(d\phi/dz)_{z=D} = q_0$ . The solution then reads

$$\phi = \frac{vE}{2K_{22}} z^2 + qz + \phi_0, \quad (15)$$

where

$$q = \left( q_0 - \frac{vE}{K_{22}} D \right). \quad (16)$$

Using this at  $z = 0$ , we get

$$W = - \frac{vED}{\sin \phi_0 \cos \phi_0}. \quad (17)$$

$v$  has the dimension of an energy/unit area and, as we noted earlier,  $v \propto q_0$ . A dimensional estimate gives

$$v = xK_{22}q_0, \quad (18)$$

where  $x \sim 1$ . This gives  $v \sim 10^{-6} \text{ J m}^{-2}$ .

If  $\phi_0 = \pi/4$ , we get  $W \sim 10^{-7} \text{ J}$ . This is equal to an extrapolation length [22]  $L = K_{22}/W \sim 10 \mu\text{m}$ . If the drops have to rotate the anchoring energy must be weaker than this. To our knowledge, measurements of the azimuthal anchoring energy are not available, but for the polar angle  $\theta$ , a silicon monoxide coated surface gives an extrapolation length of only  $\sim 0.1 \mu\text{m}$  [28, 29] and it is not easy to obtain surfaces with weak anchoring. We could hence conclude that the azimuthal anchoring with an extrapolation length  $> 10 \mu\text{m}$  is also difficult to achieve on a solid surface. If the cholesteric is in contact with solid surfaces on both sides, we can expect that the anchoring energy must be much weaker than the above estimate to obtain any rotation of the cholesteric. We believe that in none of the earlier attempts to reproduce Lehmann rotation the anchoring was sufficiently weak for the structure to rotate.

### 3. Experimental technique and results

It is clear from the discussion in the previous section that cholesteric drops which are in contact with glass plates are not suitable for observing the Lehmann rotation phenomenon. On the other hand, one can expect that the azimuthal anchoring energy at a cholesteric-isotropic interface to be essentially zero. With this idea in mind, we first tried to sandwich cholesteric drops between two glass plates coated with glycerine. But this resulted only in spherical drops which do not have a suitable geometry. We found that a convenient method of getting the required type of drops was to dissolve in the cholesteric a few per cent of Lixon, which is a non-mesomorphic epoxy compound. This results in a lowering of the cholesteric-isotropic transition temperature, and a broad two phase region. Moreover, one can form cholesteric drops which no longer have a spherical shape but have a flattened appearance. More interestingly, they are surrounded by the isotropic phase on all sides (figure 1). The reason for this configuration is that the epoxy compound has a strong affinity for glass, i.e. it wets the glass much more efficiently than the cholesteric compound. The cholesteric phase has a lower concentration of the epoxy compound than the isotropic phase, which would hence prefer to be close to the glass plates. We can also note that the interfacial tension between the cholesteric and its own isotropic phase is likely to be  $< 10^{-6} \text{ N/m}$  (see for example [30]) and hence the energy required to change the shape of the drop from the spherical shape is negligible. Further, the relative difference in density between the cholesteric and its own isotropic phase is also very small,

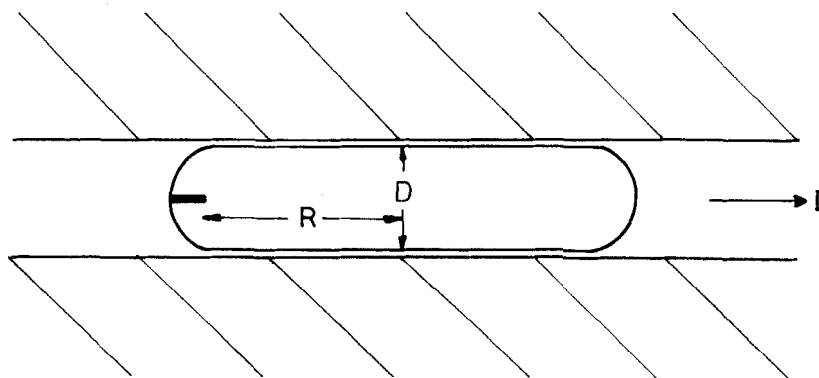


Figure 1. Vertical cross section of the flattened cholesteric droplet surrounded by the isotropic phase (I). The glass plates are shown by shaded area.

$\sim 10^{-3}$  which enables the cholesteric drop to be surrounded by the isotropic phase on all sides. This results in the configuration shown in figure 1. In thin cells, with the glass plates separated by  $\sim 8\ \mu\text{m}$ , we could easily form cholesteric drops with a lateral diameter of  $20\text{--}50\ \mu\text{m}$  using this technique. It is clear that the gap between the plates is too small for having a steady temperature gradient between them. However, as we mentioned earlier, cross couplings can be expected for any transport current. It is extremely convenient to apply a DC electric field to such thin cells. Hence we chose to look for the electromechanical rather than the thermomechanical coupling. If  $v_E$  is the relevant coupling coefficient, one can write, on the basis of dimensional arguments

$$v_E = nK_{22}q/V, \quad (19)$$

where  $n$  is a small number  $\sim 1$  and  $V$  is a characteristic voltage of the material. Identifying it with the redox potential of the mesogen,  $V \sim 1$  volt and using  $K_{22} \sim 10^{-12}\ \text{N}$  [31], we get  $|v_E/q| \sim 10^{-12}\ \text{J m}^{-1}$ . However, we must note that the director also couples to an external electric field through the dielectric anisotropy. Since the field has to be applied along the helical axis in the present experiment, we have chosen materials with negative dielectric anisotropy to avoid a change in the orientation of the director due to this coupling. We made a binary mixture of alkoxy phenyl *trans* alkyl cyclohexyl carboxylates (obtained from the Merck Co.) to get a room temperature nematic with a dielectric anisotropy  $\simeq -1$ . Materials with negative dielectric anisotropy and with ionic impurities can exhibit however electrohydrodynamic instabilities under the action of a DC field beyond a threshold voltage [32]. We checked that our material did not exhibit such instabilities up to about 8 volts. By dissolving a small percentage of Lixon, we could get a nematic–isotropic two phase region at room temperature. The nematic drops had the bipolar configuration [33, 34] characteristic of a tangential boundary condition at the interface. In order to get cholesteric drops, we added suitable chiral compounds to the mixture. Left and right handed helical arrangements were obtained by dissolving cholesteryl chloride and methyl butyl benzoyloxy heptyloxy cinnamate respectively in the sample. The handedness of the helical arrangement was determined using a wedge shaped sample of the chiralised material (without Lixon). The relative movement of the dark brushes near the edge of the wedge in relation to that of the analyser can then be used to find the sense of the helix [35]. The pitch could be varied by changing the concentration of the chiral dopant, and was measured using the Cano Wedge technique.

In thick cells, we can see spherical cholesteric drops with a characteristic radial  $\chi$  line of strength  $+2$  (figure 2(a)) [36]. The director pattern in cholesteric drops shown in figure 3 was described originally by Pryce and Frank and quoted by Robinson *et al.* [37]. As the thickness of the cell is reduced, the drops get flattened and then it is clear from figure 2(b) that the  $\chi$  line extends only in the lateral curved region of the drop. In thin cells, with  $D \simeq 8\ \mu\text{m}$ , cholesteric drops with a lateral diameter of  $20\text{--}50\ \mu\text{m}$  can be formed. These are highly flattened versions of the spherical drops described earlier. The director configuration is stretched such that the  $\chi$  line is now confined to the small lateral curved region, and has a length  $\sim D/2$ . The drops are surrounded by the isotropic phase on all sides, and since the director has a tangential alignment at the interface, the central ‘flat’ region of the drop has an essentially planar texture with the helical axis perpendicular to the flat surface. However, in each horizontal layer, the director has a splay-bend distortion arising from the presence of the  $\chi$  line at the edge and we can see 4 of the 8 dark brushes emanating from the  $\chi$  line for suitable settings of the polarizer and analyser which are set at suitable angles to get



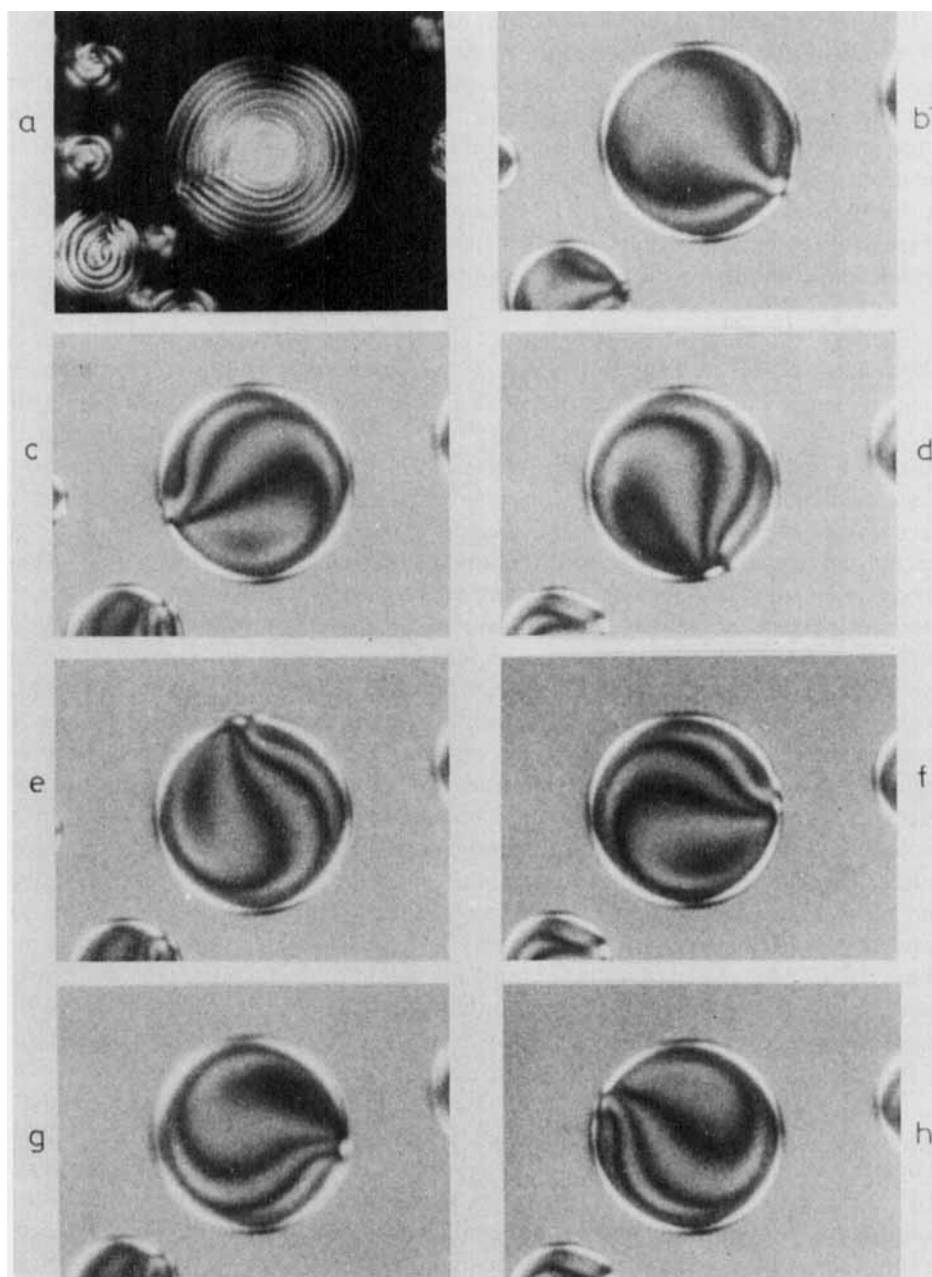


Figure 2. (a) Photograph of the cholesteric droplet showing the  $\chi$  defect line in a nearly spherical drop in a sample of thickness  $\approx 150\ \mu\text{m}$ , crossed polarizers ( $\times 1300$ ). (Note that the line defect extends only in the curved region of the droplet.) (b) Flat droplet of a left handed cholesteric when  $E = 0$ . Analyser rotated by  $\sim 20^\circ$  from the crossed position in relation to the polarizer. The dark brushes emanate from the short  $\chi$  line near the edge of the droplet ( $\times 1500$ ). (c)–(h) Photographs of the drops shown in (b) at different times after the application of a voltage to the cell. (c), (e), (g) correspond to  $+2\ \text{V}$  and (d), (f), (h) to  $-2\ \text{V}$ .

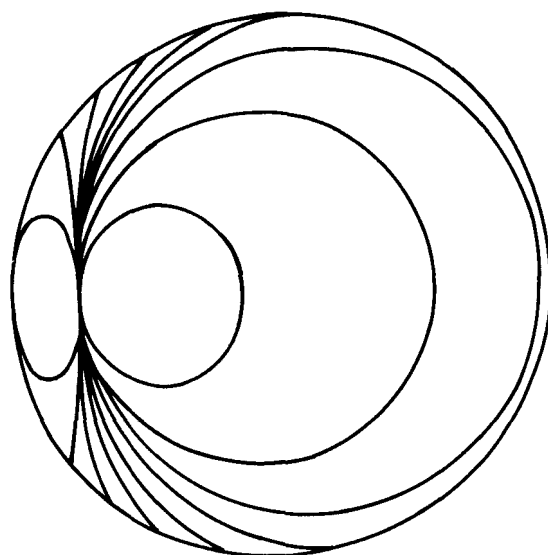


Figure 3. Director orientation on the concentric spherical surfaces of a cholesteric drop described by a family of circles passing through a singular point (after Robinson *et al.* [37]).

the dark brushes (figure 2(b)). We may also note that the splay-bend distortion in the layers can result in a flexoelectric polarization [38] which can in turn give rise to only a static distortion of the director field of the cholesteric drops under the action of the external DC field.

In the experiment, such 'flat' drops are formed in cells which are made with electrically conducting glass plates. A DC electric field is applied to the electrodes, but nothing happens as the voltage is increased gradually from 0 to 2 V. At  $\sim 2$  V the dark brushes get curved such that all of them have the same curvature, unlike in the field free case (figure 2(b)). This results from a rearrangement in the director configuration and indicates that the reorientation of the director is easier if it is far away from the  $\chi$  line. Then the whole structure starts rotating apparently without any further deformation of the director field (figure 2(c), (e), (g) and figure 2(d), (f), (h)). These rotating structures are indeed reminiscent of the diagrams sketched by Lehmann [4] (see also Chandrasekhar [39]). Detailed observations on drops of a given pitch lying in the range  $4\text{--}10\text{ }\mu\text{m}$  lead to the following results: (a) all the drops in a given sample rotate in the same direction for a given sense of the field: the right handed helix has an anticlockwise rotation when viewed along the field direction. When the voltage is reversed, the curvature of the dark brushes and the sense of rotation of the structure reverse, (b) the angular velocity increases linearly with applied voltage up to  $\sim 3.5$  V beyond which the structure of the drop changes and the rotational velocity becomes a nonlinear function of the applied voltage (figure 4), (c) nematic drops do not rotate under the action of  $\mathbf{E}$ , (d) when the handedness of the helix is reversed, the angular velocity also reverses sign for any given sense of the field  $\mathbf{E}$ , (e) the angular velocity does not depend on the radius of the drop, showing that we have a rotation of the structure rather than that of the rigid body of the drop, (f) though the angular velocity is roughly similar in all drops, some drops which touch dust particles rotate with a lower velocity (figure 4), (g) the extrapolated angular velocity tends to zero for

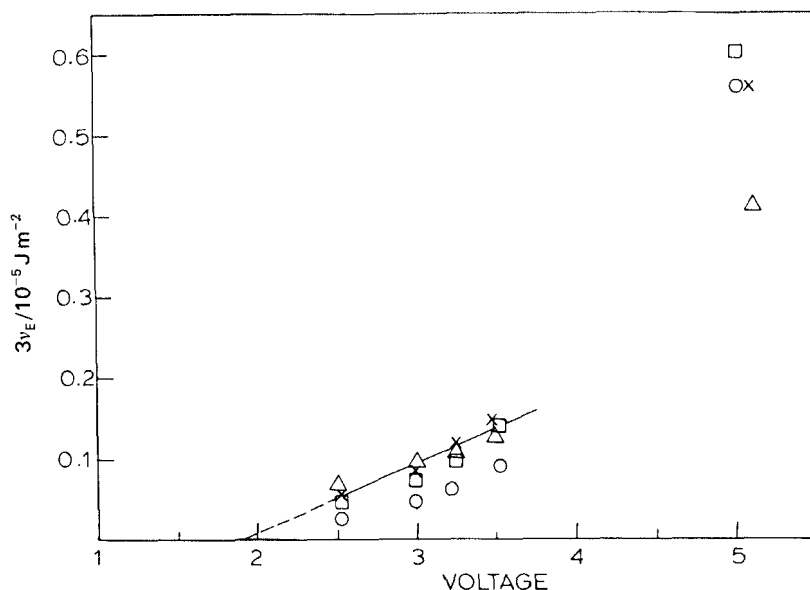


Figure 4. Plot of the rotational velocity of the structure as a function of applied voltage. Different symbols correspond to different drops. The slope of the straight line which corresponds to the drops with the fastest angular velocity is used in the calculation of  $v_E$ . Between 3.5 V and 5 V there is a visible disturbance in the structure of the drops and measurements were not possible.

$V \sim 1.9$  V (figure 4). The last point indicates that the DC field is totally screened up to  $\sim 1.9$  V and that the redox potential of at least one of the components in the mixture is  $\sim 1.9$  V.

Measurements could be made on drops with different values of pitch in the range  $4\text{--}10\text{ }\mu\text{m}$ . The coefficient  $v_E$  (calculated using equation (23)) varies linearly with  $q$  (figure 5). The line passes through the origin, i.e.  $v_E \propto q$  as required by the hydrodynamic theory. This also means that  $v_E$  is of structural origin, with practically no measurable contribution from the molecular chirality. If  $P$  is  $\lesssim 4\text{ }\mu\text{m}$ , for samples of  $D = 8\text{ }\mu\text{m}$ , there would be two pitches at the lateral curved edge of the drops and in this case the structure of the drop itself changed periodically under the action of the field: the central 'flat' part winds up to a finger print configuration with spirals from the centre (figure 6) and then unwinds to give the 'Planar' texture once in a while. If the pitch is too large ( $\gtrsim 12\text{ }\mu\text{m}$ ) the  $\chi$  defect in the drop moved to the centre and this configuration is not suitable for the experiment.

For a defect free planar undistorted cholesteric sample, the solution corresponding to the Lehmann rotation is, from equation (12)

$$d\phi/dt = v_E E/\gamma_1, \quad (20)$$

where  $v_E (=k_{3E})$  is the electromechanical coefficient,  $E$  being the electric field. However, we must also note that near the mid plane of the curved periphery of the drop,  $\mathbf{q}$  lies in the horizontal plane, i.e. perpendicular to  $E$ . In this case, as we mentioned in §2, the  $\mu_{8E}$  coefficient alone produces a linear velocity of the molecules along  $E \times \mathbf{q}$  [21]. This could produce a rotation of the drop, but our observation that the reorientation of the director starts near the central 'planar' part of the drop, and the fact that the period of rotation is independent of the diameter of the drop shows that the  $\mu_{8E}$  effect is indeed small and we have neglected it in further analysis.

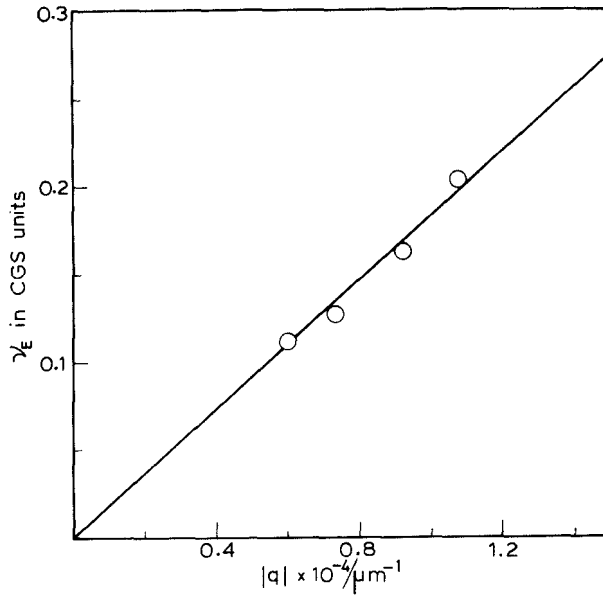


Figure 5. Plot of  $v_E$  as a function of  $q$  ( $= 2\pi/p$ ).

However, we may add that in drops with a low value of the pitch, whose structure changes periodically, the  $\mu_{8E}$  effect may make an important contribution. In the present paper, we confine our attention to the drops with a larger pitch in which the effect of the  $\mu_{8E}$  coefficient is unimportant.

It is clear that our observations (a) to (e) are consistent with the predictions of equation (20) and the requirement that  $v_E$  should change sign with that of  $q$ . However, before we use the data to determine  $v_E$ , we have to note that the line defect which is confined to the periphery of the drop also rotates with the structure. The deformation of the director field is extremely large near the  $\chi$  line and the reorientation implied by the motion of the defect requires a considerable energy. On the other hand, the electromechanical coupling which drives the rotation of the director is confined to the 'planar' oriented flat part of the drop. The effective friction coefficient  $\zeta$  (per unit length) for a slow motion of a nematic line singularity with a core has been estimated by Imura and Okano [40] and de Gennes [41]. They have shown that  $\zeta \propto s^2$ , where  $s$  is the strength of the defect line. It is very likely that the  $\chi$  line of strength 2 would be coreless because of the collapse of the director in the third dimension [22]. It is easy to extend the formalism of [40] to this case and the result is

$$\zeta = 2\gamma_1 |s|, \quad (21)$$

which is a linear function of  $|s|$ .

The entropy generation arises due to the rotational motion of the director ( $= \gamma_1 \dot{n}^2$ ) and the linear motion of the defect around the periphery of the drop, which is  $\zeta u^2$  per unit length of the defect line where  $u$  is the velocity of the defect. Assuming that the defect has a length  $= D/2$ , we can now write an approximate energy rate balance equation for the entire drop:

$$2\pi\gamma_1 |s| u^2 D/2 + \pi r^2 D \gamma_1 (d\phi/dt)^2 \simeq \pi r^2 D v_E E \frac{d\phi}{dt}, \quad (22)$$

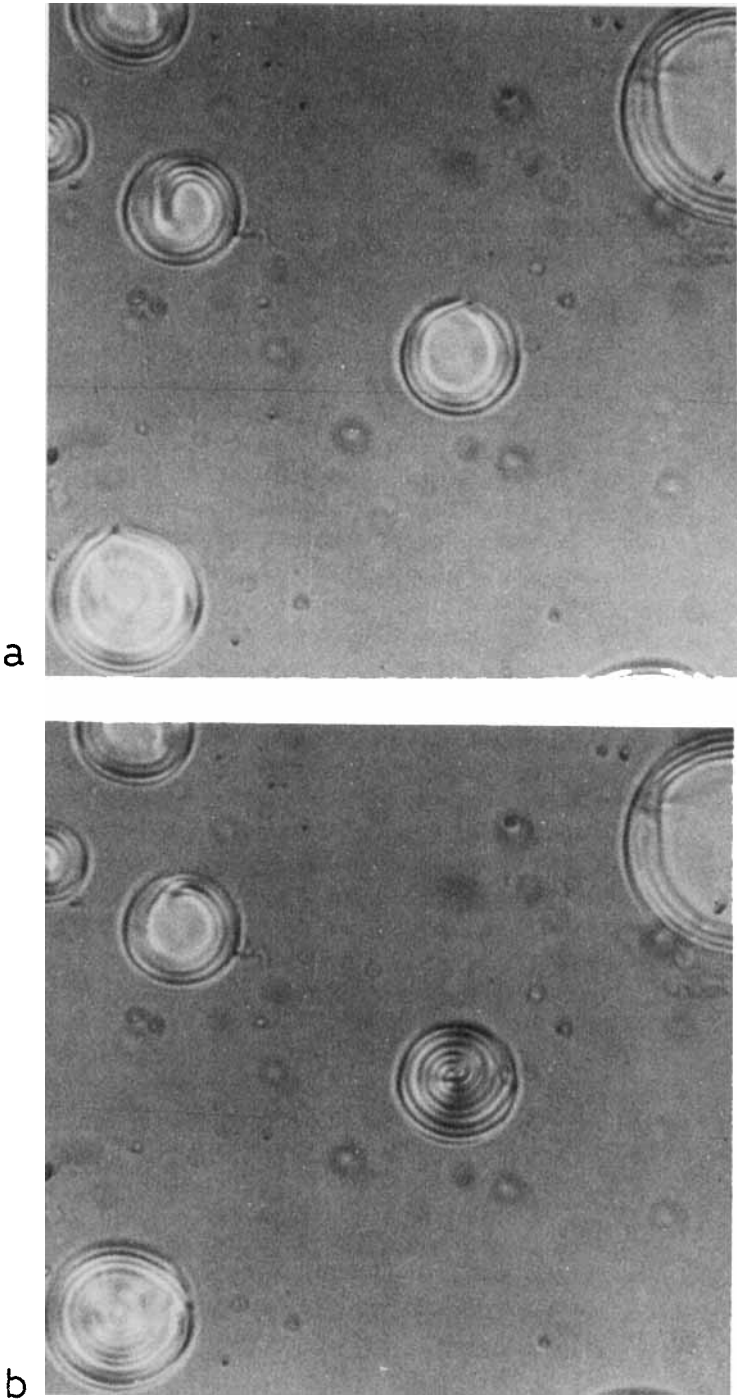


Figure 6. Flat droplets of a left handed cholesteric with pitch  $p = 3.6 \mu\text{m}$  with an applied voltage of 2.54 V. The central region of the droplets periodically change over from a 'planar' configuration to a helically wound one. The two configurations can be clearly seen for a droplet just to the right of the centre of the photographs in (a) and (b) respectively ( $\times 1000$ ).

where  $r$  is the radius of the flat drop, and we can take  $u = r d\phi/dt$  for the velocity of the defect and its strength  $s = 2$ . We recover equation (20) in the absence of the line defect. But in the presence of the line defect rotating with the structure, we get

$$d\phi/dt = v_E E/3\gamma_1. \quad (23)$$

Using the slope of the linear part of the  $d\phi/dt$  vs.  $E$  curve (figure 4), and  $\gamma_1 \simeq 0.7P$  typical of a room temperature nematic, we have calculated  $v_E$  for samples with a few different values of the pitch  $P$ . The variation of  $v_E$  with  $q$  is shown in figure 5. It is clear that  $v_E$  is a linear function of  $q$  and moreover, the extrapolated line passes through the origin.  $v_E/q$  is found to be equal to  $-0.6 \times 10^{-12} \text{ J m}^{-2}$ , which agrees with the dimensional estimate made earlier.

#### 4. Conclusion

The cholesteric liquid crystal has a macroscopic helical arrangement of oriented molecules. One could then expect that the cross coupling coefficients between fluxes and forces allowed by the Curie principle in such chiral systems would be quite large and have observable effects. The rotation of the cholesteric structure observed by Lehmann in 1900 has been attributed to this cross coupling. Since then, however, there has been no convincing demonstration of such a cross coupling term. A truly couple stress free boundary condition is essential to observe this phenomenon. We have devised a simple technique of realizing the same. For the sake of convenience, we have looked for an electromechanical coupling, and have been able to reproduce the 'Lehmann rotation' of cholesteric drops under an electric field. Indeed such an observable cross coupling appears to be unique to chiral liquid crystals, viz. cholesterics and chiral smectics as has been recently discussed by Brand and Pleiner [27]. We have checked that the phenomenon satisfies all the required symmetry properties: the rotational velocity linearly depends on  $E$ , and  $v_E$  the coupling coefficient changes sign with that of  $q$ . Indeed  $v_E \propto q$  demonstrating that  $v_E$  has a structural origin. The cholesteric drops that we have studied do not have the ideal defect free planar configuration assumed in deriving the theoretical results, but the presence of the line defect and the associated deformation of the director field actually helps in visualizing the rotation of the structure. We have allowed for the entropy production involved in the motion of the defect line in an approximate manner, and this allows us to estimate the coupling coefficient  $v_E$ . In the system studied,  $v_E/q$  has a negative sign, and it is gratifying that its value, viz.  $-0.6 \times 10^{-12} \text{ J m}^{-2}$  is quite close to the estimate made by using dimensional arguments.

We are extending these studies to exactly compensated cholesteric mixtures and to other chiral liquid crystal systems. We are also planning to determine the thermomechanical coefficients.

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