Electromechanical Effect in Cholesteric Liquid Crystals with Fixed Boundary Conditions

N. V. Madhusudana [a] , R. Pratibha [a] & H. P. Padmini [a]

[a] Raman Research Institute, Bangalore, 560080, India
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Electromechanical Effect in Cholesteric Liquid Crystals with Fixed Boundary Conditions

N. V. MADHUSUDANA, R. PRATIBHA and H. P. PADMINI

Raman Research Institute, Bangalore 560080, India

We describe a number of experiments to demonstrate the electromechanical effect in cholesteric liquid crystals. The Lehmann rotation phenomenon of drops with stress free boundary conditions is the most convincing effect produced by this coupling. In samples with fixed boundary conditions, the cross coupling produces a distortion of the profile of the azimuthal angles of the director. We have used a conoscopic technique to demonstrate this distortion in thick samples containing negative dielectric anisotropy materials. A strong *linear* electrooptic effect of electromechanical origin is produced in materials with a weak positive dielectric anisotropy. We provide an evidence that a twisted nematic cell without any chiral molecules also exhibits the electromechanical effect, due to the macroscopic chirality of the medium in such a cell.

Keywords: electromechanical effect, cholesteric LCs

1. INTRODUCTION

Chiral systems can sustain novel cross couplings between fluxes and forces. A classic example of such an effect is the 'Lehmann rotation' phenomenon, which was indeed one of the earliest physical experiments to be reported on thermotropic liquid crystals.[1,2] A temperature gradient applied along the helical axis of a cholesteric drop resulted in a continuous rotation of the structure. It is interesting to note that there has been no other report in the literature of repeating this unique experiment on chiral liquid crystals. Leslie[3] developed a general theoretical model of the hydrodynamics of cholesteric liquid crystals and obtained solutions corresponding to the Lehmann rotation. Lubensky[4,5] developed a hydrodynamic theory of cholesterics valid for spatial distortions whose wavelength is >>P, the pitch of the helix. A general model for the hydrodynamics of layered systems has been given by Martin *et al.*[6] Recently Brand and Pleiner[7] have argued that the thermomechanical Lehmann rotation has a possible static contribution in addition to the hydrodynamic one.

From the experimental point of view, the only claim about a measurement of the thermomechanical coefficient is due to Eber and Janossy.[8,9] They used a mixture

of two liquid crystals which shows a compensation temperature T_c at which the pitch is infinity. Further, they assumed that the thermomechanical coefficient is independent of the pitch, i.e., they effectively assumed that it is molecular in origin. On the other hand, since the theories are valid in the hydrodynamic regime, one would expect that the coupling constants actually depend on the macroscopic chirality associated with the helical arrangement of the director rather than that of the molecule.[10,11] It would hence appear that there is no unequivocal measurement of the thermomechanical coefficient in cholesteric liquid crystals.

de Gennes[10] has pointed out that in general any transport current would lead to cross coupling effects in cholesterics. We recently demonstrated[12,13] the electro-mechanical coupling which arises due to the transport of ions in a cholesteric sample. In that experiment, a dynamic Lehmann rotation phenomenon was found by the action of a DC electric field on carefully chosen cholesteric droplets with the requisite structure floating in the isotropic phase. We report in this paper the observation of electromechanical effects in more general geometries of the sample.

We should point out here that Li et al.[14,15] have recently found evidence for an 'electroclinic' effect in some long pitch cholesteric mixtures. The electroclinic effect is well known in smectic A liquid crystals made of chiral molecules,[16] in which a tilt angle is produced in a plane orthogonal to the one containing E the electric field and the layer normal. Li et al. have found a weak electroclinic effect in the cholesteric phase of a system which shows a smectic A-cholesteric transition. The experiments clearly indicate that the electroclinic coefficient strongly depends on the smectic A like short range order, pointing to the possibility that the tilting of the molecules is taking place in the layers. Li et al. have argued that a biaxial chiral molecule with a transverse permanent dipole could in principle give an electroclinic effect. In any case, since the electroclinic effect is essentially molecular in origin, it is practically insensitive to the frequency of the applied field in the range of 250 Hz-10 KHz. On the other hand the electromechanical effect which is hydrodynamic in origin should decrease quite rapidly with the frequency of the applied field.

We give a brief summary of the relevant theoretical background on electromechanical coupling in Section 2. The experimental techniques and results are described in Section 3. We conclude the paper with some remarks in Section 4.

2. THEORETICAL BACKGROUND

If the electric field acts along the helical axis of a sample contained between two conducting glass plates, the torque balance equation takes a very simple form:[3,10,17]

$$\gamma_1 \frac{\partial \phi}{\partial t} = k_{22} \frac{\partial^2 \phi}{\partial z^2} + \nu E \tag{1}$$

where $\gamma_1 = \alpha_3 - \alpha_2$ is the difference between two Leslie viscosity coefficients. k_{22} is the twist elastic constant, ν the electromechanical coefficient and ϕ the azimuthal angle. The above equation can be integrated to obtain a solution for ϕ by using appropriate boundary conditions. An extremely interesting solution is obtained if

the anchoring energy for azimuthal orientation is zero on both surfaces. The boundary conditions then correspond to $d\phi/dz = q_0(=2\pi/P)$ at the boundaries and hence is a constant in the sample. The solution is then of the form[3,10]

$$\phi = q_0 z + \frac{\nu E}{\gamma_1} t + C \qquad (2)$$

where C is a constant of integration. It corresponds to a continuous rotation of the structure with time, i.e., to 'Lehmann rotation.' The angular velocity is given by $\nu E/\gamma_1$. The cholesteric drops which are found to follow this dynamical behaviour contain a χ-line defect and the above Equation has to be modified to take into account the entropy production due to the motion of this defect.

Experimentally it is very convenient to prepare samples in which the director is fixed at both the boundaries $z = 0$ and D. Under the action of an applied DC field the medium has a static deformation, and the solution to Equation (1) reads as

$$\phi(z) = \frac{\Phi_D z}{D} + \frac{\nu E z}{2k_{22}} (D - z) \qquad (3)$$

where $\phi = 0$ and Φ_D at $z = 0$ and D respectively. In the absence of the applied field, the director has a uniform twist in the sample. When the field is applied, the variation of ϕ across the sample thickness becomes non-uniform (Figure 1) such that the thickness averaged value $\bar\phi$ will be greater than or lower than that in the field free case depending on the sign E (and that of ν).

It is usually preferable to apply AC rather than DC electric fields to liquid crystals to avoid electrolytic processes. If the applied field is given by $E = E_0 e^{i\omega t}$, one may expect in the linear regime that ϕ also oscillates with the frequency ω. The field-dependent part can be written as

$$\delta\phi(z, t) = \hat\phi_0(z)e^{i\omega t} \qquad (4)$$

where $\hat\phi_0(z)$ is a complex amplitude. Substituting this in Equation (1), the general solution for the amplitude is of the form

$$\hat\phi_0(z) = a \cosh p(1 + i)z + b \sinh p(1 + i)z - \frac{i\nu E_0}{\omega\gamma_1} \qquad (5)$$

where

$$p = \sqrt{\omega\gamma_1/(2k_{22})}.$$

With the boundary conditions
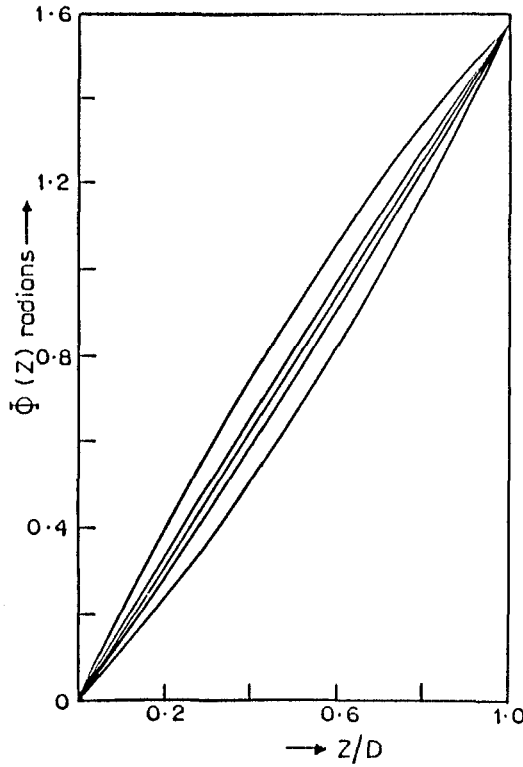
$$\hat\phi_0(O) = 0 \text{ and } \hat\phi_0(D) = 0$$

FIGURE 1   Theoretical ϕ-profiles (from Equation 3) of the director in a $\pi/2$-twisted cholesteric cell subjected to a DC electric field. $\nu = 0.05$ cgs units, $k_{22} = 5 \times 10^{-7}$ dynes, $D = 3$ μm. The curves from top to bottom correspond to V $= +10$ V, $+3$ V, 0 V, -3 V and -10 V respectively.

the explicit expression for the amplitude is given by

$$\hat{\phi}_0(z) = \left[-\left(\frac{\nu E_0}{\omega\gamma_1}\right) (\sin pz \sinh pz - (\cos pD + \cosh pD)^{-1} \right.$$

$$\left. \times (\sinh pD \sin pz \cosh pz + \sin pD \cos pz \sinh pz)) \right]$$

$$+ i \left[\left(\frac{\nu E_0}{\omega\gamma_1}\right) (\cos pz \cosh pz - (\cos pD + \cosh pD)^{-1} \right.$$

$$\left. \times (\sinh pD \cos pz \sinh pz - \sin pD \sin pz \cosh pz) - 1) \right] \qquad (6)$$

We have shown the real and imaginary parts of $\phi(z)$ for two different frequencies in Figure 2. The lower frequency corresponds to $\nu E_0/\omega\gamma_1 \simeq 0.86$ and the higher
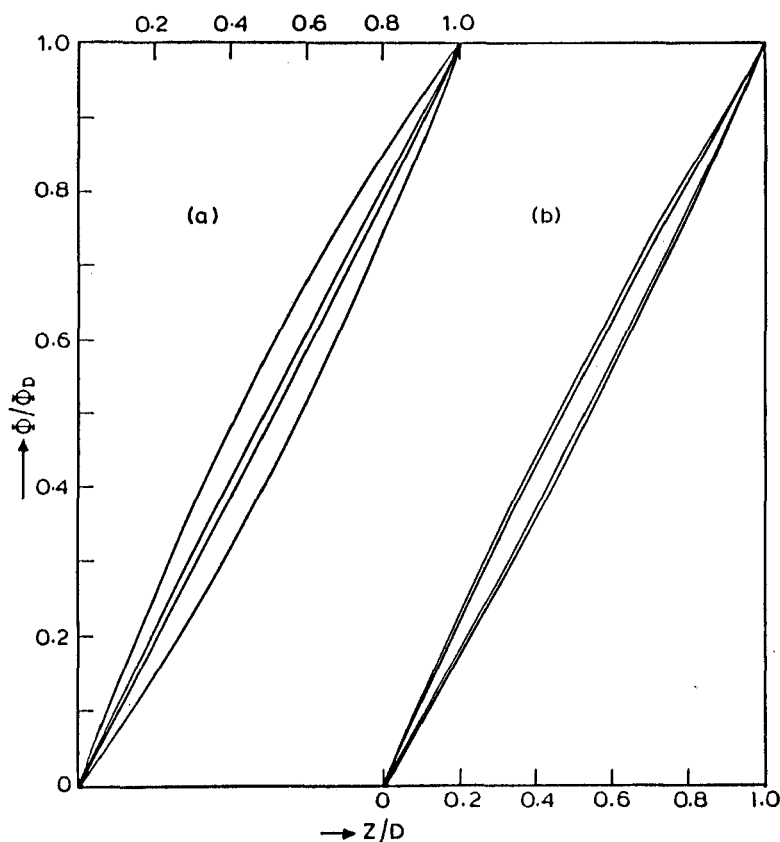
FIGURE 2   Theoretical curves showing the real and imaginary components of the field dependent amplitude $\hat{\phi}_0/\Phi_d$ (from Equation 6) superposed on the uniform field free azimuthal profile. $D = 3$ μm, $k_{22} = 5 \times 10^{-7}$ dyne.
a) $\omega\gamma_1 = 7$ cgs units. The first and third curves from the top (bottom) correspond to the real and imaginary parts of $\hat{\phi}/\Phi_d$ for $vE_0 = +6$ (-6) cgs units respectively.
b) $\omega\gamma_1 = 70$ cgs units. The first and third curves from the top (bottom) correspond to the imaginary and real parts of $\hat{\phi}/\Phi_d$ for $vE_0 = -6$ ($+6$) cgs units respectively.

one to 0.086. It is clear that as the frequency increases, the amplitude decreases while the phase lag with respect to the applied field increases.

In deriving the above equations we have ignored various other mechanisms through which the director couples to the applied electric field. If the dielectric anistropy is negative the corresponding torque due to the field will favour the director to be perpendicular to the field. In this case the equations derived earlier will be applicable, provided however that the applied voltage is lower than the threshold for electrohydrodynamic instability. The most convenient method of detecting the distortion in the director field is through an optical technique. With the usual materials and typical sample thicknesses, the phase difference of the sample is much larger than the angle of twist. In such a case, the polarization follows the director (Mauguin criterion) and the deformation in ϕ-profile cannot be detected.

This problem can be overcome by using materials with a positive dielectric anisotropy. At voltages above the Freedericksz threshold, a tilt-deformation is produced in the director field thus reducing the effective birefringence of the sample. It then becomes possible to detect the changes in the $\phi(z)$ profile. It is necessary however to select a sample thickness $D < P/4$, where P is the pitch of the cholesteric helix. For larger thicknesses the helical axis rotates to the XY plane above a threshold voltage[19] giving rise to a striped domain pattern. Also, if $\Delta\varepsilon$ is very large, the Freedericksz transition occurs at a very low voltage. As the voltage is increased above the threshold, the average orientation of the director in the sample tilts towards the field and the electromechanical coupling becomes weaker. Since the torque due to this coupling is linear in E (see Equation 1) it is better to apply higher voltages to the cell to improve the possibility of detecting the corresponding signal. We cannot however make $\Delta\varepsilon$ too small as the sample becomes unstable to a flexoelectric periodic distortion if $\Delta\varepsilon \simeq 0$.[20]

The above discussion makes it clear that one may hope to look for an electro-optic signal due to the electromechanical coupling if a material with weakly positive dielectric anisotropy is chosen and the pitch and sample thickness are properly adjusted. However, the theoretical analysis of the problem becomes quite complicated once we have both θ and ϕ distortions in the sample. Firstly the electric field becomes non-local,[21,22] whose spatial profile depends on either the conductivity anisotropy or the dielectric anisotropy depending on the ratio of the frequency of the applied field to that of charge relaxation. Secondly velocities are produced in the material under AC fields, which have to be properly taken into account. We shall not take up a solution of this problem in the present paper. We describe our experimental results in the following section.

3. EXPERIMENTAL RESULTS AND DISCUSSION

First we shall briefly describe the results on the 'Lehmann rotation' of drops with zero surface anchoring energy and later those on samples with fixed boundary conditions.

a) Samples with Stress Free Boundary Conditions[12,13]

The material consisted of a binary mixture of alkoxy phenyl *trans* alkyl cyclohexyl carboxylates (obtained from the Merck Co.) to get a room temperature nematic with $\Delta\varepsilon \simeq -1$. The threshold voltage for EHD instabilities was about 8 V. Cholesteryl chloride and methyl butyl benzoyloxy heptyloxy cinnamate were added to get cholesteric materials with left- and right-handed helical arrangements respectively. The experimental details concerning the method of preparation of the samples and the basic observations have been given in our earlier papers.[12,13] By allowing for the entropy generated in the movement of the χ-line defect present in the drops, it can be shown that[13]

$$d\phi/dt = \nu E/3\gamma_1. \tag{7}$$

We have calculated v using the slopes of $d\phi/dt$ vs. E curves for samples with different values of the pitch. In the materials studied, the compensated mixture (with $q_0 = 0$) has the highest amount of chiral additives, viz., 10%. Thus the basic chemical constituents can be considered to be similar in all the mixtures studied to a good approximation. We find that $v \propto q_0$ (Figure 3). In particular, we find that a compensated cholesteric mixture which contains both cholesteryl chloride (~3%) and of methyl butyl benzoyloxy heptyloxy cinnamate (~7%) does *not* show the rotation phenomenon, clearly demonstrating that the electromechanical effect is essentially of structural origin. From the slope of the line we get $v/q_0 \simeq -2 \times 10^{-5}$ cgs units.

While the Lehmann rotation phenomenon is perhaps the most convincing evidence for the cross coupling term in the cholesteric phase, experimentally it requires a very special combination of chemicals to get the required type of drops. In the following we describe some experiments on samples with fixed boundary conditions.

b) Samples with Fixed Boundary Conditions

In these experiments, a strong anchoring of the director on the indium-tin oxide coated glass plates was obtained by oblique evaporation of silicon monoxide, at a grazing angle of ~35°. As we discussed in the previous section, the spatial variation of the azimuthal angle of a cholesteric material can be expected to become non-uniform under the action of a DC electric field, due to the electromechanical effect. In a material with negative $\Delta\varepsilon$, only the electromechanical coupling is effective if
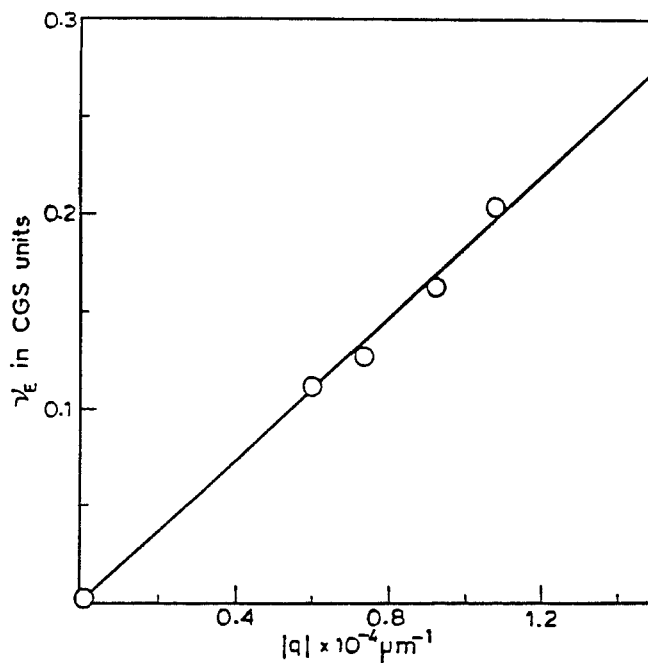

FIGURE 3   Plot of v_E (electromechanical coupling coefficient) as a function of q_0 ($=2\pi/P$).

the voltage is below the threshold for EHD instability. The average value of ϕ can be derived from Equation (3) for DC excitation and is given by

$$\bar{\phi} = \frac{1}{D} \int \phi(z)dz = \frac{\Phi_D}{2} + \frac{\nu D^2}{12k_{22}} E \tag{8}$$

$\bar{\phi}$ is greater than or less than that of the field-free value of $\Phi_D/2$ depending on the sign of E. In view of the difficulty of detecting this change in $\bar{\phi}$ by using an optical beam incident normally on the sample as we discussed earlier, we tried to measure it by using small cylindrical rods of ~ 5 μm diameter and ~ 25 μm length as microprobes of the director orientation. The density of the rods which were made of an epoxy material matched with that of the liquid crystal and they floated in the sample. However, we found that the rods were always negatively charged and they moved from one electrode to the other as the field direction was reversed (Figure 4). The rods of course rotate in the process to align along the local direction of the director. This experiment actually serves to bring out the physical mechanism of the electromechanical coupling: the permeative helical motion of the charged particles produces a torque on the medium.

The average azimuthal angle of the distorted director field can be sensed by a conoscopic technique.[10] We need relatively thick (~ 50 μm) samples to see the conoscopic figure characteristic of an optically negative uniaxial medium along the optic axis. We chose a sample with P = 3.5 μm and $\Delta\varepsilon \simeq -1$ for this observation. It was a mixture with about 90% of 2-cyano-4-heptylphenyl-4'-pentyl-4-biphenyl carboxylate (7P(2CN)5BC) and 10% of the optically active 4-cyano-4-(2-methyl) isobutoxy biphenyl (OA). As a low frequency (6 Hz) square wave electric potential of ~ 210 V is applied, the conoscopic Figure clearly rotates back and forth showing that the average value of $\bar{\phi}$ is oscillating at the frequency of the applied field (Figure 5). The applied voltage is above the restabilisation voltage of EHD instabilities.[2]

Our more detailed observations were made on samples with a weak positive dielectric anisotropy. For this purpose, we used mixtures of 82% 7P(2CN)5BC and 18% of 4'-n-heptyl-4-cyanobiphenyl (7CB), with a very wide temperature range of the nematic phase (~ 45–90°C) and with a dielectric anisotropy of ~ 0.2 and birefringence $\Delta n \simeq 0.18$ at 50°C.[23] Left handed cholesterics with the appropriate values of pitch were obtained by adding a few per cent of OA such that the mixture always had 82 mol % of 7P(2CN)5BC. The pitch was measured using the Cano wedge technique. Typically the pitch is ~ 10 μm and the sample thickness ~ 3–4 μm so that the director had a rotation of π radians in the sample. At low enough frequencies, of about a few Hz, the medium had a very clear *linear* electroptic effect when kept between a pair of crossed polarizers and oriented such that the surface alignment made an angle ψ of $\pi/8$ radians with the polarizer. Indeed as Figure 6 shows, the electrooptic signal recorded using a photodiode and a digital storage oscilloscope varies at the frequency of the applied field in this case. The electrooptic signal oscillates at twice the frequency of the applied field when ψ is changed to $\pi/4$ radians. This change over can easily be understood as follows. The
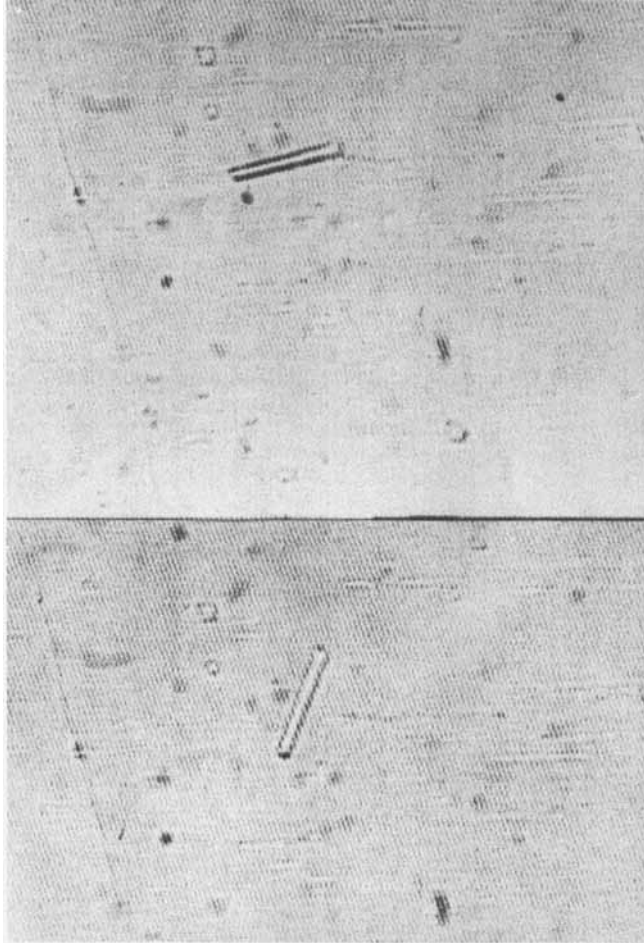
FIGURE 4   Print from a video recording showing the orientation of an epoxy rod in a cholesteric sample with $\Delta\varepsilon \simeq -1$, a twist of π radians and $D \simeq 20$ μm, as it settles close to the positive electrode. Top: +5V, Bottom: −5V. The vertical motion of the rod causes the change in the focus (× 500).

coefficient of transmitted intensity of a light beam through a homogeneously aligned sample kept between two crossed polarizers is given by

$$T_I = \frac{\sin^2 2\psi}{2}(1 - \cos \Delta\Phi) \tag{9}$$

where ψ is the angle made by the optic axis with the polarizer and $\Delta\Phi$ is the phase difference between the extraordinary and ordinary rays passing through the sample. If the medium has a small change $\delta\psi$ in the azimuthal angle, the change in the transmitted intensity is given by

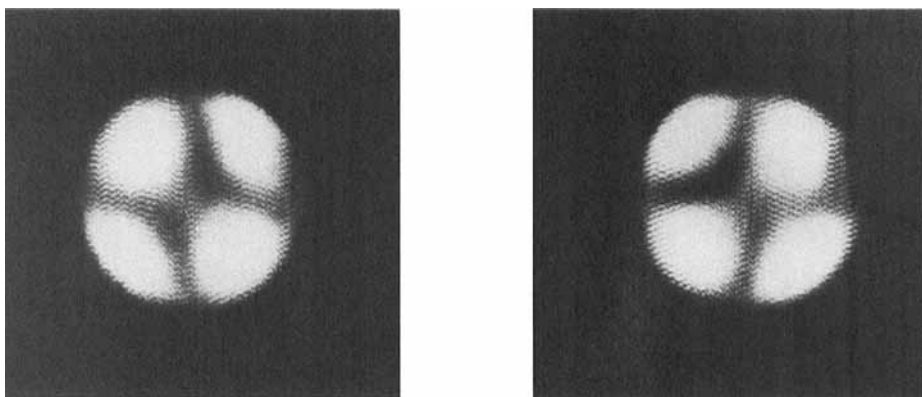$$\delta T_{I(\delta\psi)} = \sin 4\psi(1 - \cos \Delta\Phi)\delta\psi \tag{10}$$

FIGURE 5   Print from a video recording showing the rotation of the conoscopic figures of a 50 μm thick negative Δε material when the voltage oscillates between +210 V (left) and −210 (right) at 6 Hz.
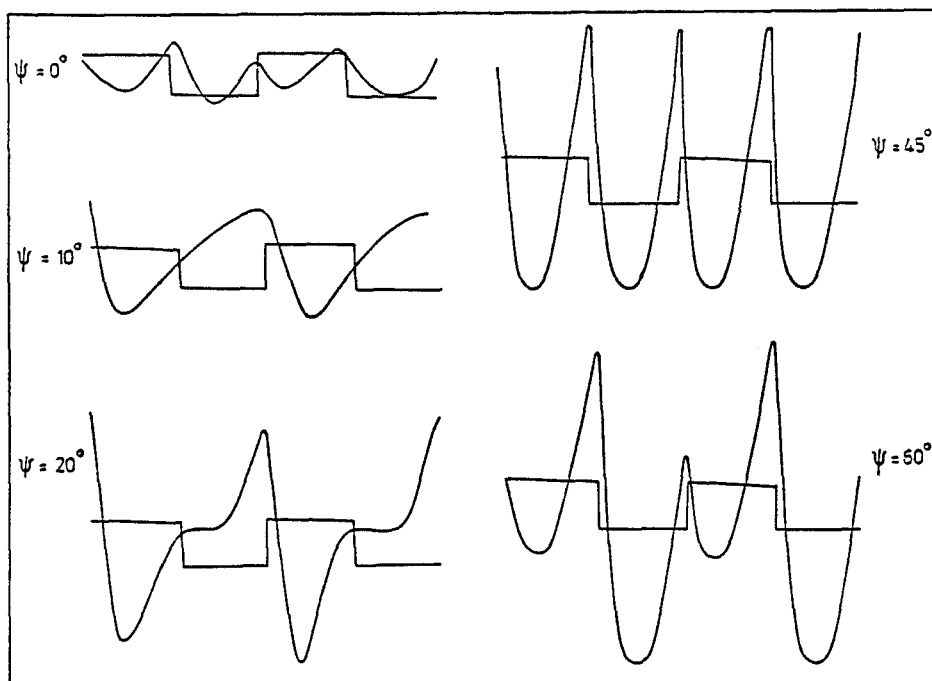

FIGURE 6   Electrooptic signal from a cholesteric with weak positive Δε and subjected to a square wave voltage (of frequency 20 Hz and rms value 3V), recorded from a digital storage oscilloscope after smoothening. ψ, the angle between the director on the bottom electrode and the polarizer is varied between 0 and 60°. Notice the predominance of the '*f*' signal at ψ = 20° and that of '2*f*' signal at ψ = 45°.

The φ oscillations due to the electromechanical coupling effectively gives rise to ψ oscillations and the maximum in $\delta T_{I(\delta\psi)}$ occurs at ψ = π/8 radians. On the other hand, if there are θ variations, ΔΦ varies and

$$\delta T_{I(\delta\theta)} = \frac{\sin^2 2\psi}{2} \sin \Delta\Phi\delta(\Delta\Phi) \tag{11}$$

or $\delta T_{I(\delta\theta)}$ has its maximum value for $\psi = \pi/4$ radians. The θ oscillations occur due to the dielectric anisotropy of the sample and are quadratic in E, as seen in Figure 6.

The electromechanical effect which is linear in E has no threshold. But as we discussed in Section 2, the ϕ distortions can be made visible optically only when there is a sufficient θ distortion, i.e., somewhat above the Freedericksz threshold. This can be clearly seen by monitoring the optical signal on a spectrum analyzer (Anritsu model No. MS420K). If f is the frequency of the applied sine wave electric field, as seen in Figure 7, with a gradual increase of the voltage initially only the $2f$ signal develops, signifying a θ-distortion. At a slightly higher voltage the f signal shows up. At even higher voltages the spectrum consists of many higher harmonics indicating that the system has become non-linear.

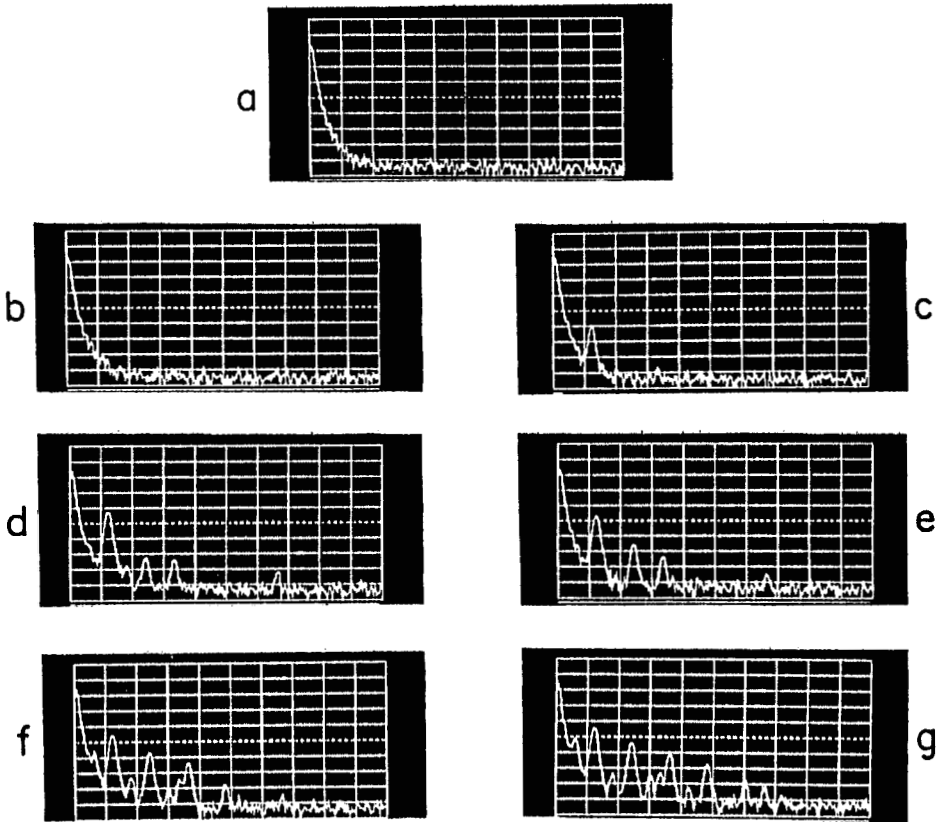We have made some detailed measurements of the optical signals at f and $2f$

FIGURE 7   The spectrum of the electrooptical response of a chiral sample with $\Delta\varepsilon \simeq +8$ with a π radian twist subjected to an 18 Hz sine wave electric field. $\psi \simeq 5°$. The rms values of the voltages are: (a) 0 V, (b) 1 V, (c) 1.5 V, (d) 1.8 V, (e) 2 V, (f) 3 V and (g) 4 V, respectively.

using a lock-in-amplifier (PAR model 5301A). The block-diagram of the experimental set up is shown in Figure 8. The measurements made on a sample with d = 3.2 μm and P = 18 μm and $\psi \simeq \pi/8$ radians are shown in Figure 9. The '2f' signal shows the characteristic oscillations corresponding to a continuous decrease of the optical phase difference as the voltage is increased. The 'f' signal does not show such oscillations. Further it decreases continuously as the frequency is raised, showing that it arises from the electromechanical rather than the electroclinic effect. Further, as the frequency is increased, the f and 2f signals become measurable at voltages which are increasingly larger than the Freedericksz threshold. The above sample is likely to be 'untwisted', as its thickness is less than P/4. In view of the linear dependence of v on q_0 that we found in our experiments on the drops the present result indicates that v depends on the natural wavevector (2π/P) of the samples. We have carried out a similar experiment on a 4.5 μm thick cell which contains the same mixture between two glass plates twisted by ~70°. The electrooptic signals are similar to those shown in Figure 9, except that both the f and 2f signals are much stronger in this case.

Very recently we found that an *unchiralised* nematic sample with a twist angle of ~20° shows the ψ angle dependence of the f and 2f signals as shown in Figure 10. The f signal again has a maximum at $\psi \simeq \pi/8$ radians and the 2f signal at $\psi \simeq \pi/4$ radians. The medium did not have striped domain pattern characteristic of the
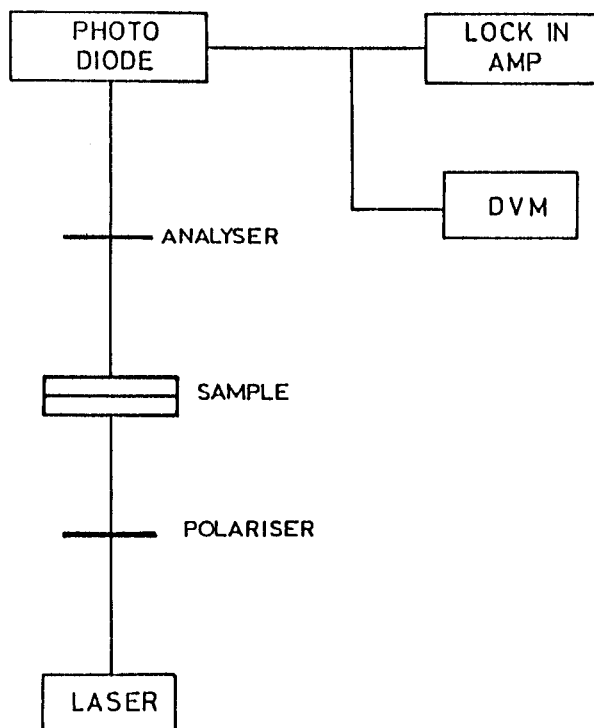

FIGURE 8   Block diagram of the experimental set up to study f and 2f components of the electrooptic signal. The sample sits on the rotating stage of a microscope.
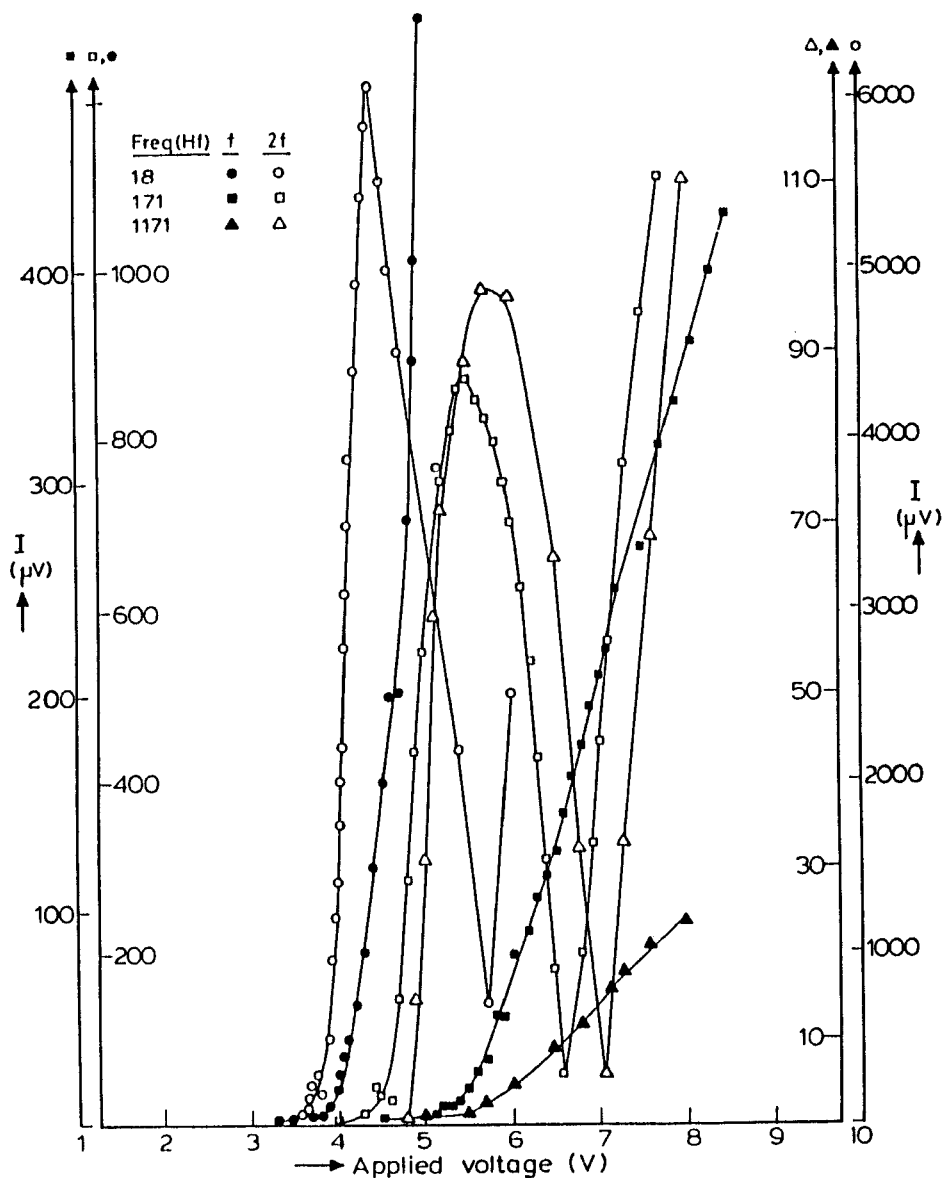
FIGURE 9   The f and $2f$ components of the electroptic signal from a 3.2 μm thick cholesteric sample with P ≃ 18 μm, at ψ = π/8 radians. The f signal does not exhibit maxima and minima unlike the $2f$ signal with increase of voltage. Both f and $2f$ signals decrease with increase of frequency.

flexoelectric distortions[20] which can occur with fixed boundary conditions. If the surface anchoring was not strong enough for the tilt angle θ, one would have got flexoelectric tilt distortions which are linear in E. However, this process would have produced a peak of the f signal at ψ = π/4 radians. It would appear that there are φ-oscillations in the sample and we believe that they are caused by electromechanical coupling. When we twist a nematic we produce a macroscopic
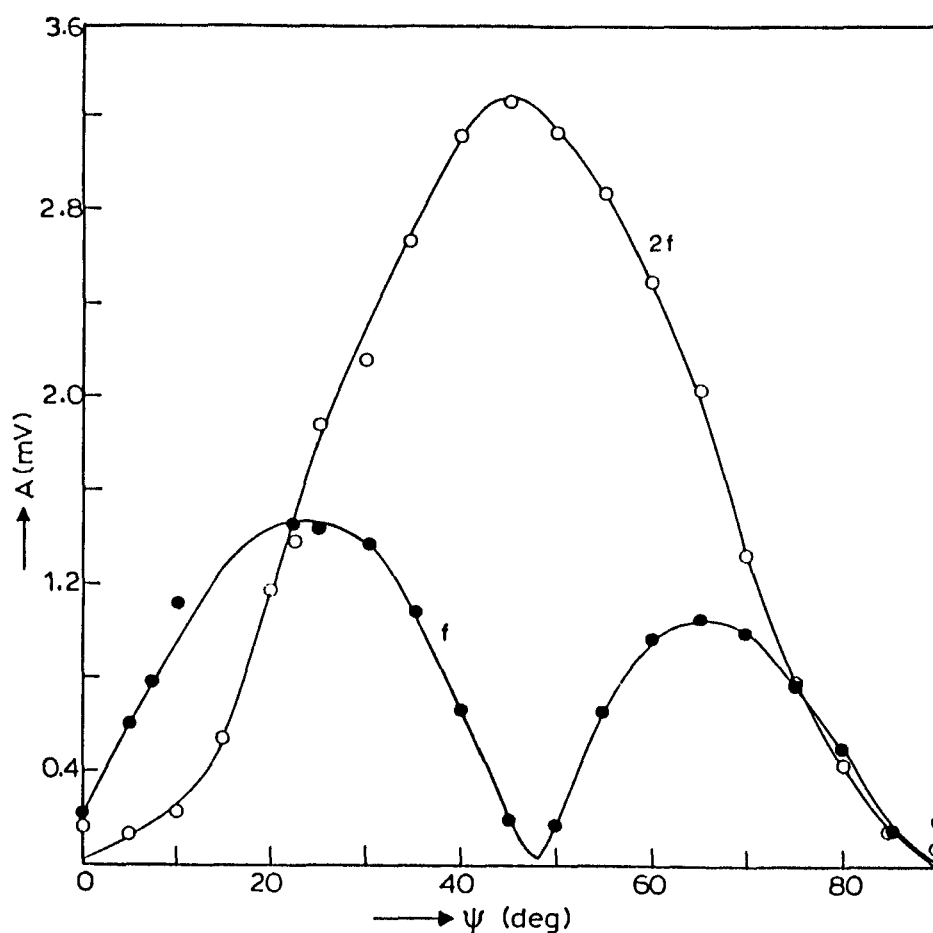
FIGURE 10   The ψ angle dependence of the f and $2f$ signals from a twisted nematic sample with weak positive dielectric anisotropy. Twist angle $\simeq 20°$. $f = 18$ Hz. The signals were amplified through the transformer input of the lock-in-amplifier.

chirality in the sample and our observations again shows that the electromechanical coupling has a contribution from the macroscopic helical arrangement of the director.

CONCLUDING REMARKS

We have described in the paper several experiments which demonstrate the phenomenon of electromechanical coupling in chiral nematics. The 'Lehmann rotation' phenomenon of drops with stress free boundary conditions is perhaps the most fascinating consequence of this coupling. We have used a conoscopic technique to demonstrate the oscillations of the azimuthal profile in a material with the negative dielectric anisotropy and subjected to a low frequency AC field. We have also

obtained an easily observable linear electrooptic effect by choosing a material with a small positive dielectric anisotropy. The measured frequency dependence of the optical signals is quite large and this rules out any significant contribution from the electroclinic effect in this system. Our preliminary measurements on a twisted nematic without any chiral additives serves to demonstrate that the electromechanical coupling has a significant contribution from the macroscopic twist of the director. Our results on untwisted chiralised, twisted chiralised and twisted nematic samples appear to indicate that the electromechanical coupling has a contribution which depends on q_0 the natural wavevector, and a contribution which depends on the macroscopic twist. It appears that the smectic C^* type crystals should also have observable consequences due to the electromechanical effect in appropriate geometries.[24]

We are at present solving the full electrooptic equations for materials with a positive dielectric anisotropy. We are also searching for appropriate materials to optimise the linear electrooptic effect, and to measure the electromechanical effect in materials with negative dielectric anisotropy. We are also carrying out detailed experiments to separate the q_0-dependent and twist contributions and finally to look for a possible molecular contribution to the electromechanical coefficient.

Acknowledgments

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