Linear flexo-electro-optic effect in a hybrid aligned nematic liquid crystal cell

N.V. Madhusudana, G. Durand

To cite this version:

HAL Id: jpa-00232499
https://hal.archives-ouvertes.fr/jpa-00232499
Submitted on 1 Jan 1985

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
Linear flexo-electro-optic effect in a hybrid aligned nematic liquid crystal cell (*)

N. V. Madhusudana (+) and G. Durand
Laboratoire de Physique des Solides, Bât. 510, Université de Paris-Sud, 91405 Orsay Cedex, France

(Reçu le 9 novembre 1984, accepté le 14 janvier 1985)

Résumé. — Une cellule nématique en texture hybride présente une polarisation flexo-électrique de volume P. Le couplage de P avec un champ extérieur normal aux plaques produit une distorsion de texture, pour un ancrage faible sur les plaques. Nous observons la variation linéaire de birefringence électriquement contrôlée qui en résulte, dans le MBBA. L'énergie de surface ne varie pas en sinus carré de l'angle, pour les grands angles de surface. L'écration de l'effet aux faibles valeurs de potentiel à cause d'électrodes imparfaites est aussi discuté.

Abstract. — A hybrid aligned nematic liquid crystal sample presents a bulk flexo-electric polarization P. The coupling of P with an external electric field normal to the bounding plates allows, for weak anchoring, a texture distortion. We report a new linear flexo-electro-optic effect in such a cell with a weak anchoring, which presents an electrically controlled birefringence. The results also indicate that the \( \sin^2 \theta \) form of the surface anchoring energy is inadequate when the surface angle \( \theta \) is large. Low voltage screening by counterions is also discussed.

A hybrid aligned nematic (HAN) liquid crystal cell (see Fig. 1) has a splay-bend distortion of the director (\( \mathbf{n} \)) field and hence a flexo-electric polarization (\( \mathbf{P} \)). The latter is given by the expression [1, 2]

\[
\mathbf{P} = e_1 \mathbf{n}(\text{div} \, \mathbf{n}) + e_3 (\text{curl} \, \mathbf{n} \times \mathbf{n}).
\]

The difference between the flexo-coefficients \( (e_1 - e_3) \) has been measured [3] in a HAN cell by the application of a transverse field, perpendicular to the plane containing \( \mathbf{n} \). When instead the external electric field \( \mathbf{E} \) is applied between the bounding plates, so that the director distortion remains confined to a plane, the orientation of \( \mathbf{n} \) can be characterized by a single variable \( \theta(z) \) where \( \theta \) is the angle made by \( \mathbf{n} \) with the z-axis (Fig. 1). In such a case, the free energy per unit area due to the flexo-polarization, \( \int - (\mathbf{P} \cdot \mathbf{E}) \, dz \), is of the form \( \int f(\theta) (d\theta/dz) \, dz \); it integrates out as a surface term. If the anchoring energy \( (W) \) at the boundaries is « strong », the surface
energy does not depend on $E$ and the flexo-electric effect is not observed. As was pointed out by Helfrich [4], a weak anchoring surface is necessary to observe flexo-electric effects in such geometries. To get a measure of the strength of anchoring, we use the usually assumed form, viz., $W/2 \sin^2 \theta_s$ for the dependence of the anchoring energy on the surface tilt $\theta_s$, and define the « extrapolation length » $L = K/W$, where $K$ is a curvature elastic constant of the medium. The anchoring is « strong » if $L \ll d$ where $d$ is the sample thickness. On the other hand, if $L \sim d$, the anchoring is « weak ». We can expect the flexo effect to influence the orientation at the surface and hence $\theta(z)$ in the bulk. Though there have been calculations on polarity dependent electro-optic effects in a few geometries with uniformly aligned nematic samples [4, 5], so far only some surface domains have been experimentally observed [6], and they do not yield strong electro-optic effects. In the present Letter, we report the first « large » polarity dependent flexo-electro-optic effect.

We first outline a simplified model. When the flexo-electric contribution dominates, for small values of $E$, we can ignore the energy due to dielectric anisotropy ($\Delta \varepsilon$) which is quadratic in $E$. In any case, even when the linear effect is small, we can extract it from the data as we shall see later. We assume that the anchoring energy is infinitely strong at the planar orienting surface and weak at the homeotropic one. In the one elastic constant approximation, the free energy per unit area of the sample is given by

$$F_s = \int \left[ \frac{K}{2} \left( \frac{d\theta}{dz} \right)^2 + (e_1 + e_3) \sin \theta \cos \theta \frac{d\theta}{dz} E \right] dz + \frac{W}{2} \sin^2 \theta_s. \quad (2)$$

The Euler-Lagrange equation leads to the constant curvature condition:

$$\frac{d\theta}{dz} = \frac{\pi}{2} - \theta_s. \quad (3)$$

The torque balance equation at the homeotropic surface reads

$$K \left( \frac{d\theta}{dz} \right) + (e_1 + e_3) \sin \theta_s \cos \theta_s E = W \sin \theta_s \cos \theta_s. \quad (4)$$
which can be rearranged to yield

$$\frac{2\left(\frac{\pi}{2} - \theta_s\right)}{\sin 2 \theta_s} = \frac{d}{L} \left[ 1 - \frac{(e_1 + e_3)}{K} EL \right].$$  \hspace{1cm} (5)

Equation (5) gives the explicit dependence of $\theta_s$ on $E$ arising from finite values of $L$ and $(e_1 + e_3)$. The optical path difference is given by:

$$\Delta l = n_o \left[ \int_0^n \frac{1}{\sqrt{1 - R \sin^2 \theta}} \, dz - d \right] \approx \left( \frac{n_o}{4} \right) R d \left[ 1 + \left( \frac{L}{d} \right) \left\{ 1 + \left( \frac{L(e_1 + e_3)}{K} E \right) \right\} \right].$$ \hspace{1cm} (6)

where $R = (n_e^2 - n_o^2)/n_e^2$, $n_e$ and $n_o$ being the extraordinary and ordinary refractive indices. We have expanded $\sqrt{1 - R \sin^2 \theta}$ for small $R$ ($R \approx 0.2$) and assumed that the applied voltage (and hence $EL$) is small compared to $K/(e_1 + e_3) \sim 0.3$ V. The three terms in the brackets in equation (6) bring out the influence of different physical processes. The first of them gives the value of $\Delta l$ for strong anchoring ($L = 0$). The second one brings out the correction due to weak anchoring and the third term the linear electro-optic effect due to flexo-electricity in conjunction with weak anchoring.

It is interesting to compare the field induced distortion in the HAN cell and in the homeotropic cell of Helfrich experiment [4] with infinitely weak anchoring. In the HAN cell the surface angle variation $A0_s$ in presence of field is of the order $(e_1 + e_3) EL/K$, to be compared to $e_3 Ed/K$ in the homeotropic geometry. As $e_3 \sim e_1$, for weak anchoring ($L > d/2$) the HAN cell is more sensitive, and vice versa. In fact, the homeotropic plates of Helfrich cell must present also some anchoring strength, so that the sensitivity of the HAN cell must be generally larger. The main effect which increases the optical sensitivity of the HAN cell is the average tilt of the director compared to the plate normal $z$, which allows $\Delta l$ to be linear in $E$. Practically, it is also simpler to apply a large homogeneous field in the HAN cell normal to the transparent electrodes, rather than parallel to the plates in the homeotropic geometry.

In our experiments, a methoxybenzylidene butylaniline (MBBA) sample is contained between two transparent indium-tin oxide coated glass plates. The upper electrode has a silicon monoxide coating at an oblique angle to ensure a planar alignment with a strong anchoring [7]. The lower plate is coated with DMOAP silane for homeotropic treatment. We have found that the strength of anchoring due to this treatment depends on the composition of the solution used to coat the material. The optical path difference ($\Delta l$) between the extraordinary and ordinary rays is measured using a tilting compensator in conjunction with a Leitz polarizing microscope.

The $\Delta l(E)$ measurements on a cell for which $W$ is not too low are shown in figure 2. The sample thickness $d \approx 22 \mu m$ is fixed by mylar spacers. The MBBA sample was relatively pure ($T_{NI} \approx 42^\circ C$). The measurements were made at room temperature ($\approx 20^\circ C$). The planar oriented plate was connected to the source terminal of a DC power supply and the homeotropically aligned one to the common terminal. Hence in figures 2 and 3 a positive voltage means a negative field $E$ in the sample. The readings were taken after waiting for a few minutes after each change of voltage to ensure equilibrium conditions. $\Delta l$ is independent of the applied voltage up to about $\pm 1$ V. For higher voltages, $\Delta l$ increases irrespective of the sign of the voltage, i.e. the quadratic effect predominates. However the magnitude of the slope is much higher for positive voltages (i.e. negative fields) than for negative voltages (positive fields). The absence of any voltage dependence of $\Delta l$ up to $\sim \pm 1$ V indicates that we have a strong screening of the electric field by counterions near the electrodes, and in the bulk of the cell the voltage is practically negligible. Beyond the redox potential ($\sim \pm 1$ V) of MBBA molecules and the butylaniline moieties which are always present in somewhat impure MBBA [8], the screening is no longer complete. The dielectric anisotropy of
MBBA [9] is negative (−0.7 for the sample under consideration) and the dielectric torque tends to increase θ and hence Δl, which increases with voltage. However, the difference in the values of |d(Δl)/dV| for the ± signs of voltage shows that the linear flexo effect makes an important contribution. Using the known values of n₀ ≈ 1.57 and R = 0.23 [10] and the measured value of Δl for V = 0 we get L/d ≈ 0.088 from equation (6). Since K ≈ 0.7 × 10⁻⁶ dyne [11], we get from equation (5) ν ≈ 8°. By comparing Δl(V) at corresponding positive and negative voltages, we can extract both the quadratic and linear parts. The linear part has a slope d(Δl(linear))/dV = 55 μm/V. Using this in (6) we get (e₁ + e₃) ≈ −7.5 × 10⁻⁴ cgs units, in good agreement both in magnitude [12] and sign [13] with recent direct measurements.

The results on a second cell with a very low anchoring energy at the homeotropic boundary is shown in figure 3. It is clear that the linear effect predominates in this case, Δl actually decreasing for negative voltages up to −3.5 V. Beyond this negative voltage, Δl again increases due to the dielectric term. We also see the influence of the screening effect till ±1 V: the slope of the Δl(V) curve is about 5 times smaller within this voltage range than for higher voltages. The sample used in this cell was less pure (TNI = 35 °C) and the experiments were done at TNI − 10°. Using the corresponding values of n₀ = 1.55 and R ≈ 0.19 [10], with d ≈ 15 μm, from the measured value of Δl at V = 0, we estimate L/d ≈ 0.72 from equation (6), and ν ≈ 53°. Both values confirm that we have indeed a very weak anchoring in the present case. We again extract the linear part by using data at various pairs of positive and negative voltages of magnitudes greater than 1.5 V. This part has a slope of 380 μm/V. Using K ≈ 0.5 × 10⁻⁶ dyne, we estimate (e₁ + e₃) ≈ −1 × 10⁻⁴ cgs units. The sum of the flexo-coefficients is expected to decrease with temperature like the order parameter but the calculated value appears to be about three times too small compared to the expected one. More curiously, the slope d(Δl)/dV at say −2 V is larger than that at +2 V in figure 3. This means that the quadratic part which can be extracted from the data would imply a positive sign of Δε. These observations indicate that the assumed sin²θ_s form of the surface anchoring term is not adequate. The deviations from the equilibrium orientation are very large in this cell and as we have shown elsewhere [14], for large θ_s, higher order terms of the surface potential become important. If one assumes the surface torque to present a peak value at θ_s > 45° which can be got for example by adding a negative sin 4 φ term to the torque, both the value of (e₁ + e₃) and the quadratic part on the electro-optic response move in the right direction. A more detailed analysis would require independent measurements of the surface potential.

Finally, to support the interpretation that electrochemical screening of the field is responsible for the strong non-linearity at ±1 V seen in figure 3, we have studied the electrical conductivity of the cell to reversals of voltage. We have distinctly seen two time constants for the current...
Fig. 3. — Voltage dependence ($V$) of the optical path difference ($\Delta l$) in a HAN cell with a weak anchoring at the homeotropic plate.

decay, the faster one ($\sim 10^{-2}$ s) corresponding to the bulk liquid crystal dielectric relaxation. The slower decay gave a current decrease by a factor of $\sim 5$ over several seconds. This should correspond to the time constant of the thin screening counterions layers near the electrodes. This factor 5 is in good agreement with the observed ratio of the slopes $d(\Delta l)/dV$ above and below 1 V, i.e. to the screening of the bulk electric field by the surface counterions. Another problem is the possible existence of space charges in the bulk, related to the anisotropy of conductivity. These space charges would create a non-uniform field in the HAN cell. To estimate the effect, since $\text{div } J = 0$, where $J$ is the current density, we can write

$$E_z = J_z/\sigma_\parallel (1 - v \sin^2 \theta(z))$$

where $v$ is the anisotropy of conductivity $v = (\sigma_\parallel - \sigma_\perp)/\sigma_\parallel$, $\sigma_\parallel$ and $\sigma_\perp$ being conductivities parallel and perpendicular to the director, respectively; $v \approx 0.3$ for MBBA. Using (7) in equation (2) we find that $d(\Delta l)/dV$ is altered only by $\sim 15 \%$, which is not important in the present stage.

In conclusion, we have demonstrated a large linear flexo-electro-optic effect using a hybrid aligned nematic liquid crystal. This is made possible only by using an aligning plate with weak anchoring energy. In a few volt range the flexo-electric effect becomes dominant and results in an electrically controlled birefringence. The results also provide evidence for the inadequacy of the $\sin^2 \theta_s$ form of the surface potential when the surface angle $\theta_\parallel$ is large. The important influence of electrochemical effects in MBBA is also seen as a strong non-linearity near the redox potential. It must be possible to improve the linearity of the response by dissolving in the nematic, dopants with a small redox potential.
Acknowledgments.

We thank Ph. Martinot-Lagarde and J. F. Palierne for stimulating discussions.

References