Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

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To cite this Article Saunders, F. C. , Hughes, J. R. , Pedlingham, H. A. and Towler, M. J.(1989) 'Electro-optic pulse response of ferroelectric liquid crystals', Liquid Crystals, 6: 3, 341 — 347
To link to this Article: DOI: 10.1080/02678298908029085
URL: http://dx.doi.org/10.1080/02678298908029085

LIQUID CRYSTALS, 1989, VOL. 6, NO. 3, 341–347

Electro-optic pulse response of ferroelectric liquid crystals

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(*Received 3 January 1989; accepted 9 May 1989*)

The electro-optic response of ferroelectric smectic C* liquid crystals has been studied. Anomalous switching behaviour of such materials which possess a negative dielectric anisotropy has been reported. These materials show a minimum in response time at a sufficiently high field. We present results showing the dependency of this minimum upon spontaneous polarisation and the effect of AC bias. Calculations based upon the equation of motion of the director around the cone are presented which describe this effect and its dependence on the relative magnitudes of the spontaneous polarization and dielectric anisotropy of the material. Good agreement with the experimental results is found.

1. Introduction

The switching response time of a ferroelectric smectic C (S_C*) liquid crystal to an applied pulse voltage usually decreases with increasing voltage [1] but recently anomalous behaviour has been reported [2, 3] in materials with negative dielectric anisotropy. This takes the form of a minimum in the response time (t_{min}) at a field E_{min}. If the field is further increased the response time increases rapidly as the dielectric torque becomes dominant with respect to the ferroelectric torque. Calculations using the equation of motion of the director around the cone have been made [4] and are in qualitatively good agreement with observed experimental behaviour [2, 3]. From these calculations we present expressions for E_{min} and t_{min}. We have studied the minimum in the response time and measurements are presented showing the effect of spontaneous polarization on this minimum. The effect of the AC bias on the minimum is also reported and related to its effect in inducing a structure which more closely resembles that for which the expressions are derived. This effect is termed AC stabilization. The results provide general support for the theory but a more rigorous analysis reveals discrepancies and we do not believe that the uniform director configuration, upon which the calculations are based, is valid.

2. Calculations

A simplified representation of a ferroelectric smectic C liquid crystal sample, in the so called bookshelf geometry, is shown in figure 1. Assuming that the majority of the sample reacts as a uniform slab to any applied fields the switching process can be described as a simple rotation of the director around the cone. Thus it is the azimuthal angle φ, as shown in figure 1, which is the characteristic variable of the reorientation process and the response time is dependent on the rotational torque induced by the applied field.

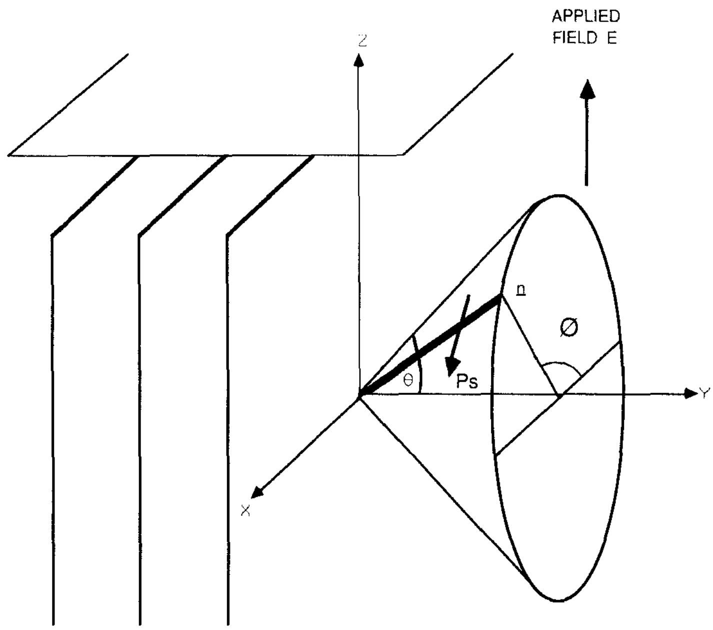

Figure 1. Simplifed representation of a ferroelectric smectic C liquid crystal aligned in the bookshelf geometry.

The equation of motion is written [4] as

$$\eta\phi_t = P_s E \sin\varphi + \varepsilon_0 \Delta\varepsilon E^2 \sin^2\theta \sin\varphi\cos\varphi. \quad (1)$$

Let

$$\alpha = \varepsilon_0 \Delta\varepsilon E \sin^2\theta / P_s \quad (2)$$

then equation (1) becomes

$$(\varepsilon_0 \Delta\varepsilon \sin^2\theta / P_s)\eta\phi_t = \alpha \sin\varphi(1 + \alpha\cos\varphi). \quad (3)$$

This function is illustrated in figure 2. It can be seen that the initial rate of change of torque with φ goes to zero as α approaches -1. We therefore expect a divergence in the switching time versus applied field when

$$\left|\frac{\partial}{\partial\varphi}(\eta\varphi_t)\right|_{\varphi=0} = 0 \quad (4)$$

Hence for the condition of reorientation from $\varphi = 0$ to $\varphi = \pi$

$$\left|\frac{E_{\text{div}}}{0 \to \pi}\right| = \left|\frac{P_s}{\varepsilon_0 \Delta\varepsilon \sin^2\theta}\right|. \quad (5)$$

To investigate where a minimum in the switching time versus the applied field is expected we first integrate equation (1) [4] to give

$$\frac{P_s^2 t}{\varepsilon_0 \Delta\varepsilon \sin^2\theta\eta} = \frac{1}{(1-\alpha^2)}\left[\frac{1}{\alpha}\ln\left|\frac{\tan\varphi_1/2}{\tan\varphi_0/2}\right| + \ln\left|\frac{(1+\alpha\cos\varphi_1)\sin\varphi_0}{(1+\alpha\cos\varphi_0)\sin\varphi_1}\right|\right], \quad (6)$$

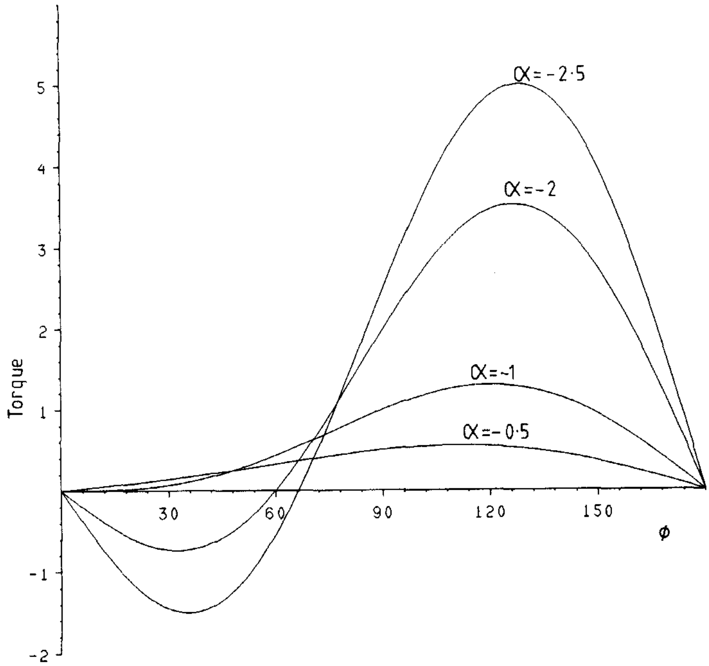

Figure 2. Reorientational torque, $(\varepsilon_0 \Delta\varepsilon \sin^2\theta/P_s)\eta\phi_t$, as a function of the azimuthal angle at different applied fields.

where φ_0 and φ_1 are the initial and final positions on the cone, respectively. Now minimize equation (6) with respect to α, which gives

$$\frac{(3\alpha^2 - 1)}{\alpha^2(1 - \alpha^2)} \ln\left|\frac{\tan\varphi_1/2}{\tan\varphi_0/2}\right| + \frac{2\alpha}{(1 - \alpha^2)} \ln\left|\frac{(1 + \alpha\cos\varphi_1)\sin\varphi_0}{(1 + \alpha\cos\varphi_0)\sin\varphi_1}\right| + \frac{(\cos\varphi_1 - \cos\varphi_0)}{(1 + \alpha\cos\varphi_0)(1 + \alpha\cos\varphi_1)} = 0. \tag{7}$$

For a given φ_0, φ_1 we can find α_{min} such that equation (7) is satisfied and this is shown in figure 3. Generally then

$$\left|\begin{matrix} E_{min} \\ \varphi_0 \rightarrow \varphi_1 \end{matrix}\right| = \left|\frac{\alpha_{min} P_s}{\varepsilon_0 \Delta\varepsilon \sin^2\theta}\right|. \tag{8}$$

Hence, we expect E_{min} to decrease and t_{min} to increase (see equation (6)) as $\varphi_0 \rightarrow 0$. In the limiting case of switching completely around the cone we set $\varphi_0 + \varphi_1 = \pi$ and let $\varphi_1 \rightarrow \pi$, thus

$$\left|\begin{matrix} E_{min} \\ 0 \rightarrow \pi \end{matrix}\right| = \left|\frac{P_s}{\sqrt{3}\varepsilon_0 \Delta\varepsilon \sin^2\theta}\right| = \frac{1}{\sqrt{3}}\left|\begin{matrix} E_{div} \\ 0 \rightarrow \pi \end{matrix}\right|. \tag{9}$$

This is a simplistic argument and it would be more correct to solve a complete equation of motion including elastic terms and, due to the resulting non-uniformity, the displacment field. Unfortunately until we are sure of an initial configuration this is not possible. However, assuming that complete switching around the cone occurs at some point through the sample and that it dominates the reorientation process, then

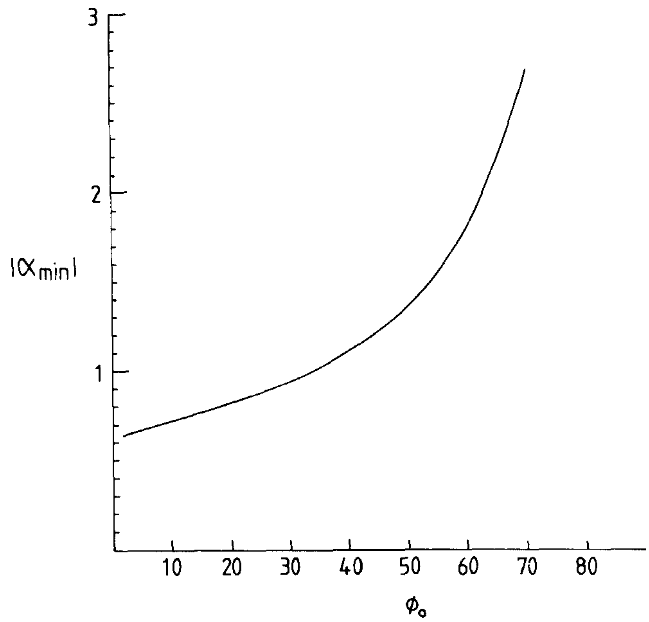

Figure 3. $|\alpha_{min}| = |\varepsilon_0 \Delta\varepsilon E_{min} \sin^2\theta / P_s|$ as a function φ_0 for $\varphi_1 = 179°$.

equation (9) should give a good approximation to the minimum in switching time versus applied field. Further, on AC stabilization, the approximation should improve. Equation (9) also highlights the importance of the parameter $P_s/(\Delta\varepsilon \sin^2\theta)$. In high P_s and low $|\Delta\varepsilon|$ materials the minimum might not be observed as it would occur at very high fields.

3. Experimental details

The materials used in these experiments were mixtures of fluorinated biphenyl esters (MBF esters) [5, 6] supplied by BDH Ltd. Two versions of these mixtures were avialable, one of which contained the racemate of the chiral dopant. By combining various proportions of the racemic and normally doped materials it was possible to make mixtures where P_s changed systematically while maintaining the same phase transitions, dielectric properties and total quantity of chiral material, and hence viscosity.

The sample cells were about 2 μm in thickness and the material was aligned on rubbed polyimide boundary layers, the rubbing directions being assembled anti-parallel. The actual sample thickness was estimated from its interference colour. By cooling the sample slowly from the cholesteric phase, which had a sufficiently long pitch for the material to be pseudo-nematic under the influence of the alignment layers, through the S_A to S_C^* phase it was possible to prepare well-aligned samples [5]. The signals applied to the cells were alternate positive and negative pulses, of height V and duration t, with a gap between the pulses providing a 1 : 100 mark to space ratio. A separate AC signal could be gated with these switching pulses to effect AC stabilization of the two states and hence investigate the effect of such conditions on the magnitude of E_{min}. The experiments were undertaken in a temperature controlled stage at 30°C under continual microscopic observation. The criterion of 'visibly clean switching' was applied to the repetetive switching between the two states and used to judge the required pulse duration, *t*, for each voltage, *V*. For pulses shorter than this the cell was observed to switch incompletely, with many small domains failing to reorient fully, giving a speckled appearance. It was therefore a very sensitive measure of switching performance and did not suffer from the problems of estimating switching times between arbitrarily set transmission levels.

4. Pulse response results

A summary of the properties of the mixtures measured is given in the table together with calculated and measured values of E_{min}. The detailed results for the relationship between response time and voltage for the 25/75*R* mixture are shown plotted in figure 4. From these graphs it can be seen that increasing the magnitude

Properties of the mixtures determined at 30°C

Mixture	P_s/nC cm^{-2}	θ/deg	$\Delta\varepsilon$	E_{min}/V μm^{-1} calc	E_{min}/V μm^{-1} meas	V_{AC}/V rms
100/0*R*	27·1	22	−1·8	70	–	
50/50*R*	15·1	22	−1·8	39	35	10
35/65*R*	10·2	20	−1·8	31	36	0
					36	5
					32	7·5
					28	10
25/75*R*	6·1	19	−1·8	21	32	0
					28	5
					24	10

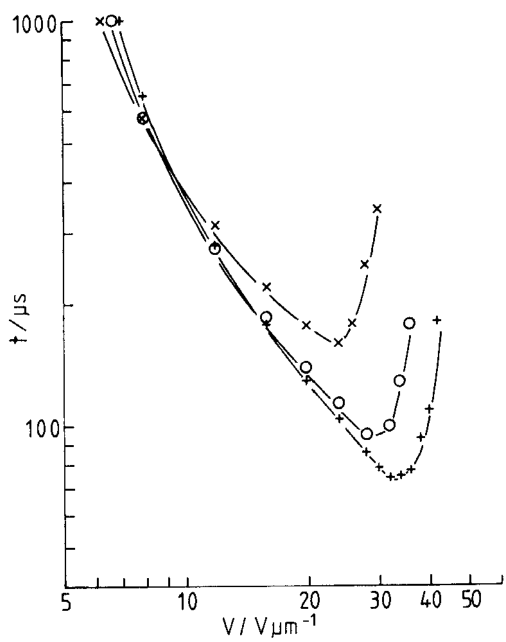

Figure 4. Response time as a function of pulse voltage at different superimposed RMS AC bias voltages for the material containing 75 per cent racemic component. +, 0 V AC; o, 5 V AC; x, 10 V AC.

of the applied AC signal produce two significant effects. Firstly the minimum response time occurs at a lower field and secondly this minimum response time increases. Both these effects can be understood by considering that the AC works to stabilise the $\varphi = 0, \pi$ configurations during the interval between the switching pulses. This is confirmed in the experiments as it results in a reduction in the decay of the optical response, which is normally observed after removal of the switching pulse [8]. Therefore the values of E_{min} measured which should give the best agreement with those calculated from equation (9) are those at the highest values of applied AC. Considering the experimental errors associated with measuring cell thickness accurately, the results in the table show good agreement between these measured and calculated values.

The effect of changing P_s can be seen from figure 5. Here the response time–voltage characteristics for four mixtures with varying P_s are shown. The materials with the high values of P_s fail to show a clear minimum over the voltage range measured, as the calculations predict. The figure also shows the expected decrease in response time with increasing P_s. We have studied other mixtures based upon the same and other host materials [9] and generally find the same behaviour of the response time–voltage curve. The P_s of the mixture has to be tailored in light of the $|\Delta\varepsilon|$ value according to equation (9) to produce an E_{min} within the measurement range of our equipment. Within the tolerances of cell thickness and cone angle we find acceptable agreement between the calculated and measured E_{min}. This behaviour does not always hold however, and some materials were found to produce a response time–voltage curve in which the response time decreased monotonically with increasing voltage.

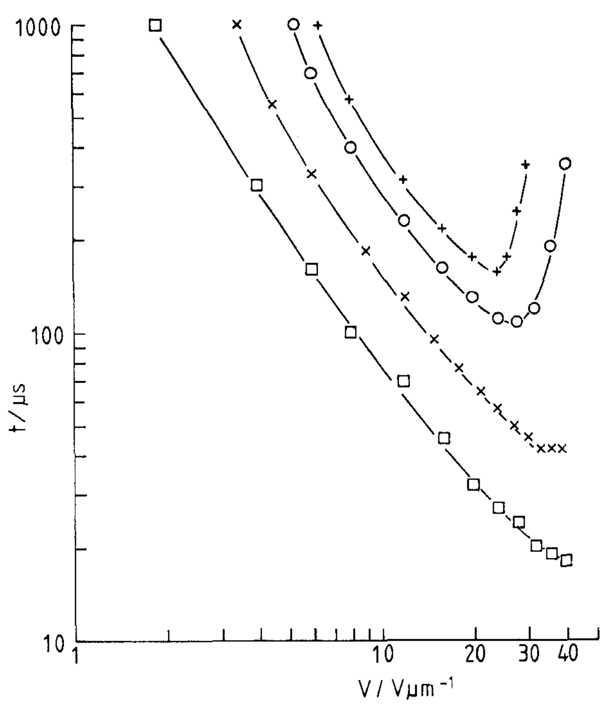

Figure 5. Response time as a function of pulse voltage at 10 V RMS AC bias for materials containing different proportions of racemic component. The mixtures studied were □, 100/0*R* (1·25 µm); x, 50/50*R* (1·66 µm); o, 35/65*R* (1·25 µm); +, 25/75*R* (1·25 µm) where the number in parenthesis is the sample thickness.

5. Discussion

We have demonstrated that anomalous electro-optic switching behaviour can occur under conditions of repetetive switching, that this is in agreement with simple theory and might therefore more properly be termed normal electro-optic switching behaviour. However in our experimental cells we observed an apparent cone angle of between 6° and 10° when no field is applied. If a uniform bookshelf is assumed, together with a true cone angle of 20°, then this would imply that the initial angle φ is of the order of 60°. This in turn leads to the prediction of values of $|\alpha_{min}|$ and therefore E_{min} that are much higher than measured. This can be rationalized if we accept that the initial director configuration is non-uniform through the cell. The apparent cone angle we observe is then the interference extinction angle associated with this non-uniform birefringent structure, which may, in addition, have some residual guiding properties. It is therefore dangerous to use this apparent angle as a direct indication of out of plane tilt and hence φ in the centre of the sample.

The effect of AC bias on the magnitude of E_{min} can be explained in terms of stabilisation of the $\varphi = 0, \pi$ configurations by a dielectric torque. This would be expected to induce a more nearly uniform director configuration throughout the bulk of the cell. We believe the structure to be important to the response time–voltage behaviour and that this is influenced by the material as well as by the aligning layer properties. Our results on materials which do not show an E_{min} support this. The possiblity of more complex structures and particularly tilted layers would alter significantly the expressions derived here and are a topic of continuing study.

6. Conclusion

We have confirmed the anomalous electro-optic switching behaviour in which a minimum response time occurs with increasing field. The values of field for which this occurs are in agreement with calculations derived from a simple model. Materials which do not show an E_{min} indicate that the structure is an important parameter and we see the modelling described here as a first step to understanding the reorientation process of the ferroelectric device under pulse voltage drive conditions.

This work was carried out under a JOERS/Alvey collaborative programme. The authors would like to acknowledge valuable discussions with colleagues at RSRE and within the collaboration.

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