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Liquid Crystals

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To cite this Article Essid, S. , Manai, M. , Gharbi, A. , Marcerou, J. P. and Rouillon, J. C.(2005) 'Electro-optical switching properties for measuring the parameters of a ferroelectric liquid crystal', *Liquid Crystals*, 32: 3, 307 – 314

To link to this Article: DOI: 10.1080/02678290500033596

URL: <http://dx.doi.org/10.1080/02678290500033596>

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Electro-optical switching properties for measuring the parameters of a ferroelectric liquid crystal

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(Received 16 January 2004; in final form 6 October 2004; accepted 14 October 2004)

We present a description of the reorientation of the macroscopic polarization \mathbf{P}_S under a switching field in a ferroelectric liquid crystal assuming the direction of \mathbf{P}_S is spatially uniform but time-dependent throughout the sample. The driving torques are then the linear ferroelectric coupling and the quadratic dielectric torques. If the theory is correct, some of the important parameters of a FLC may be determined in a single experiment, such as the spontaneous polarization \mathbf{P}_S , the switching delay t_m , the switching time τ , the orientational viscosity η and the dielectric anisotropy ϵ_a . The measurements are performed using new compound, as a function of temperature with different field strengths and sample thicknesses. The experiment appears to be poorly fit by the so-called ‘rigid’ hypothesis leading to the conclusion that the switching of polarization is not uniform but rather starts locally from orientational defects and propagates quickly throughout the sample. This reinforces the view that the driving torque equation is nothing but the Sine–Gordon equation reminiscent of soliton propagation.

1. Introduction

Since the realization of the surface stabilized ferroelectric liquid crystal (SSFLC) geometry [1], with its potential for bistable switching and hence in device technologies, there has been much interest in the SmC* phase of liquid crystals. Various studies have been performed from the standpoints of not only fundamental interest but also of potential applications to electro-optical devices [2]. Much effort has been made to optimize the physical constants of ferroelectric liquid crystals, such as spontaneous polarization \mathbf{P}_S , helical pitch p , tilt angle θ , switching time τ and rotational viscosity η , in order to produce high quality devices. Indeed the switching times under an electric field obey a scaling law with respect to the fundamental time $\tau = \eta / \mathbf{P}_S \mathbf{E}$. Therefore, materials with high \mathbf{P}_S and low η are ideal. These materials may be considered as stacks of two-dimensional liquid layers in which the rod-like molecules tend to align along a common direction, denoted by the director \mathbf{n} , which is inclined by a tilt angle θ (temperature-dependent) with respect to the layer normal \mathbf{z} (see figure 1). The ferroelectric polarization \mathbf{P}_S is in the layer plane (xy). Its direction is perpendicular to both \mathbf{n} and \mathbf{z} . When an electric field \mathbf{E} is applied parallel to the xy plane, it causes azimuthal motions (changes of Ψ) but not polar motions (θ remains fixed). There, Ψ defines the angle

between the orthogonal projection of \mathbf{n} into the smectic layers and the direction x or equivalently, the angle between \mathbf{P}_S and y (see figure 1).

2. Experimental

2.1. The experiment

The best known electro-optical phenomenon in FLCs is the Clark–Lagerwall effect (in SSFLC geometry), which

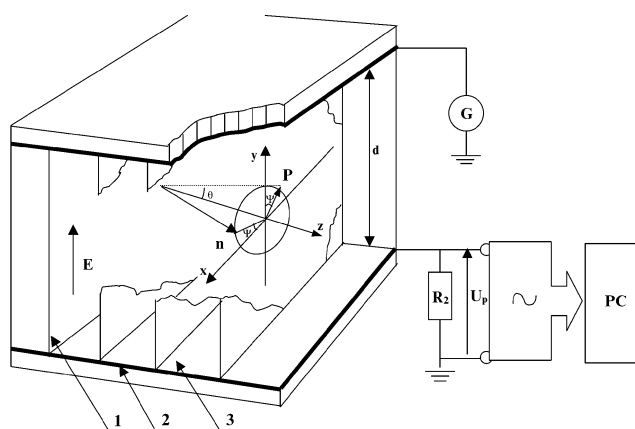


Figure 1. Geometry of the FLC cell, coordinate system and schematic diagram of the set-up. \mathbf{E} is the electric field, \mathbf{P} is the polarization, both inside the x, y plane; d is the cell thickness. (1) smectic layers, (2) conducting layer, (3) alignment layer.

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results in director reorientation from one bistable state to the other when an external electric field changes its signs. The set-up scheme and the cell geometry are shown in figure 1. SmC* liquid crystal phases are layered structures in which the director \mathbf{n} is tilted at an angle θ with respect to the layer normal \mathbf{z} . Let Ψ be the azimuthal angle of \mathbf{P} ($\mathbf{P} \propto \mathbf{n} \times \mathbf{z}$). It changes gradually from layer to layer, forming the well known helical structure [2]. We suppress the helix intrinsic to director orientation in FLCs by applying an a.c. electric field with a square wave shape [3, 4]. This holds when the zero field phase is helielectric or is any of the tilted subphases, for example, SmC_A*, SmC_α* or the so-called ferroelectric phases [5], as soon as the field is sufficiently high enough to have driven the phase transition to the uniform SmC*. Within the SmC* phase, the effect of the field change is to switch gradually the azimuthal angle Ψ from 0 for $\mathbf{E} > 0$ to π for $\mathbf{E} < 0$.

A liquid crystal cell can be regarded as a resistor R_1 and a capacitor C connected in parallel. When an electric field with a square wave alternative shape is applied to a ferroelectric liquid crystalline sample, the resulting current flowing in the circuit is the sum of three contributions [6]:

$$I = I_i + I_c + I_p \quad (1)$$

where I_i is due to the ionic current, I_c to the capacitor charge accumulation and I_p to the polarization realignment. We consider the cell resistance to be constant [6].

$$I_i = \frac{V(t)}{R_1} \quad (2)$$

where $V(t)$ is the voltage drop across the cell.

$$I_c = C \frac{dV}{dt} \quad (3)$$

$$I_p = -S \mathbf{P}_S \frac{d\Psi}{dt} \sin \Psi(t) \quad (4)$$

where S is the transverse section of the sample in the cell.

When connecting a resistor R_2 in series with the cell, the voltage drop across R_2 is:

$$U = R_2 I. \quad (5)$$

This signal is digitized by an oscilloscope (Hewlett Packard 54502A) and averaged over 64 periods. The data are stored in a computer for further treatment by a simple program to obtain the various parameters under investigation. A typical sketch of the switching current is given in figure 2.

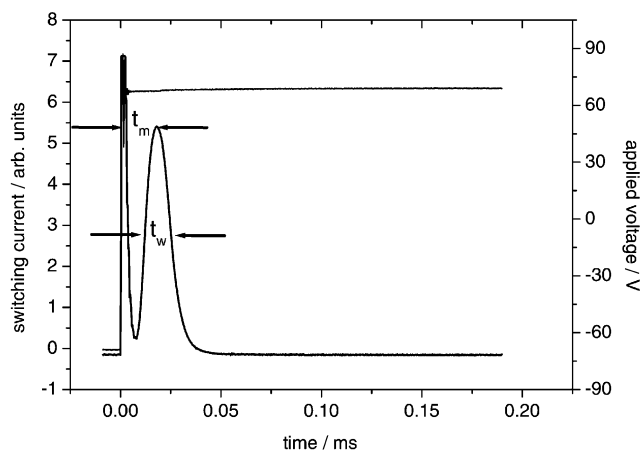
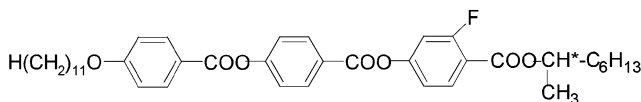


Figure 2. The measured signal and the parameters t_m and t_w under investigation at $T=106^\circ\text{C}$ ($\mathbf{E}=5.3 \text{ V } \mu\text{m}^{-1}$, $\nu=41 \text{ Hz}$).

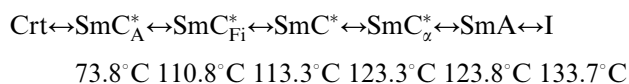
2.2. Material and mesomorphic properties

The molecular structure of the compound under investigation, called C11-F2, is given:

it belongs to a group of six homologous compounds showing a variety of different SmC* subphases.



The first assessment of the mesomorphic properties of C11-F2 was made by DSC measurements (Perkin-Elmer DSC 7), texture observations (Mettler FP5) and electro-optical measurements. The phase sequence between the crystalline (Cr) and isotropic (I) phases and the corresponding transition temperatures on cooling suggested for this compound were:



There exists a voltage threshold above which the compound experiences a phase transition from any zero-field subphase to the unwound ferroelectric phase. The experiment has been performed with voltage values well above the thresholds so we are dealing only with the SmC* phase under field.

3. Model and method of analysis

At time equal to zero, the external voltage $V(t)$ applied across the FLC cell changes its sign from $-V$ to $+V$. The polarization \mathbf{P} will begin to rotate within the layer-plane

under the influence of the external field. The variation of the azimuthal director angle Ψ is described by the equation of the torque equilibrium, which comes from the condition of minimization of the FLC free energy density with respect to Ψ . It follows the Sine–Gordon equation during the half period when Ψ decreases from π to 0 (see appendix):

$$\eta \frac{\partial \Psi}{\partial t} = -\mathbf{P}_S \mathbf{E} \sin \Psi + \varepsilon_0 \varepsilon_a \mathbf{E}^2 \sin^2 \theta \sin \Psi \cos \Psi + K \Delta \Psi \quad (6)$$

where \mathbf{E} is the applied electric field and $\Delta \Psi$ the laplacian of Ψ taking into account the elastic torque when the angle Ψ is non-uniform in space. This dynamic equation governs the director orientation within an FLC sample, where an electric field is applied parallel to the smectic planes. A few hypotheses are implied, such as the uniformity of the viscosity η and the elastic constant K as well as the assumption that the alignment is perfectly planar with the layers normal to the boundary plates. Equation (6) has been studied by different groups [7–12], where some neglect the director spatial deformation torques and the contribution of the surface interactions. Indeed, since only the uniform collective mode is considered and surface anchoring effects are neglected, the transient states during polarization switching are determined from the balance between the ferroelectric and the dielectric anisotropy terms. Also some make the assumption, the validity of which is questionable, that the polarization will begin to rotate within the layer plane under the influence of field reversal only if Ψ is not exactly 0 or π at the beginning, otherwise one would be in a metastable state where \mathbf{P} and \mathbf{E} are antiparallel but the driving torque is null. In this case the azimuthal angle would rotate from $(\pi - \Psi_0)$ to Ψ_0 (with $\Psi_0 \in]0, \pi/2[$). A direct consequence of this hypothesis is that at $t=0$ or $t \rightarrow \infty$, $d\Psi/dt \neq 0$, which is absurd as it would mean that Ψ never reaches an equilibrium value. Hence the initial and final orientations must be determined by surface interactions and these cannot be readily estimated. In addition the concept of a uniform Ψ_0 has no physical meaning. Nevertheless one can assume that in the middle of the switching, i.e. during the peak of the current in figure 2, the dynamical equation (6) could hold without the elastic torque. One can then solve the problem analytically and try to fit the theory to experimental data. In order to simplify the problem, equation (6) has been solved by Xue *et al.* [7], without the elastic torque $K \Delta \Psi$. The solution of this equation is given by (see appendix):

$$\frac{t-t_0}{\tau} = \frac{1}{1-\alpha^2} \left[-\ln \left(\tan \frac{\Psi}{2} \right) + \alpha \ln \left(\frac{1-\alpha \cos \Psi}{\sin \Psi} \right) \right] \quad (8)$$

where $\alpha = \varepsilon_0 \varepsilon_a \sin^2 \theta \mathbf{E} / \mathbf{P}_S$ measures the effect of the

dielectric anisotropy; the important times are t_0 which is equal to the time when the angle Ψ is equal to $\pi/2$, and $\tau = \eta / \mathbf{P}_S \mathbf{E}$ which is the basic switching time to which all measured times are proportional. None of these is directly measurable in the experiment, so one has to define several times, which are accessible and which may be compared with the theoretical predictions.

We note that α is the central point of this study because the case when it is neglected, i.e. when the dielectric torque is negligible, has been extensively studied [8, 9]. The results can be summarized thus: when $\alpha=0$, the current peak is symmetric and the width at half-height is equal to 1.76 times the fundamental time τ . When ε_a is taken into account, it should be noted that most of molecules yielding SmC* phases are designed to maximize the transverse permanent dipole with respect to the longitudinal one. So most of the time, ε_a and thus α are expected to be negative (for the compound under study there is a fluorine atom that should increase the transverse dipole). The dielectric anisotropy modifies the switching process regardless of its sign. If $\varepsilon_a < 0$, the director has a tendency to stay normal to the field. Thus the liquid crystal initially responds ‘slowly’, but takes a shorter time to complete the switching from the midpoint to the final state, so one expects as sketched in figure 3, a disymmetry with $t_m - t_- > t_+ - t_m$. For $\varepsilon_a > 0$, the disymmetry is reversed. At worse, if $\alpha > 1$, the equilibrium state under field would be at $\cos \Psi = \pm 1/\alpha \propto 1/\mathbf{E}$, that would mean an apparent tilt angle dependent on the field.

Before proceeding, we should note that an estimate of the order of magnitude of the parameter α , with the following values of $\varepsilon_a = -5$, $\varepsilon_0 = 8.842 \times 10^{-12}$ SI, $\theta = 15^\circ$, $\mathbf{E} = 5 \text{ V } \mu\text{m}^{-1}$, $\mathbf{P}_S = 80 \text{ nC cm}^{-2}$, is $\alpha = -1.85 \times 10^{-2}$. Thus

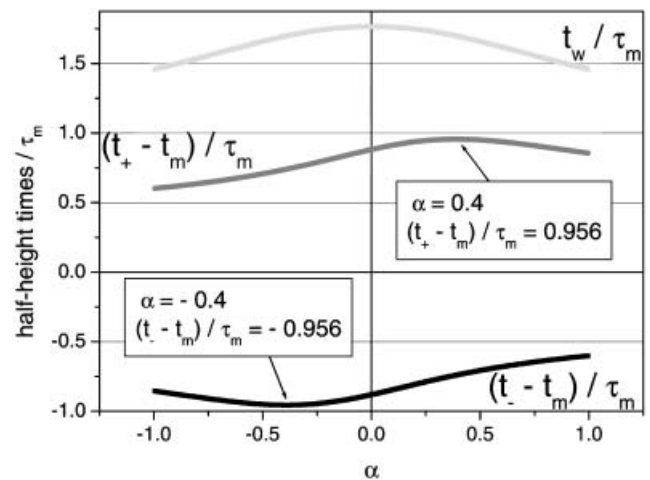


Figure 3. From bottom to top, ratios of the widths at half-height (left half, right half and full width) to the basic time τ_m .

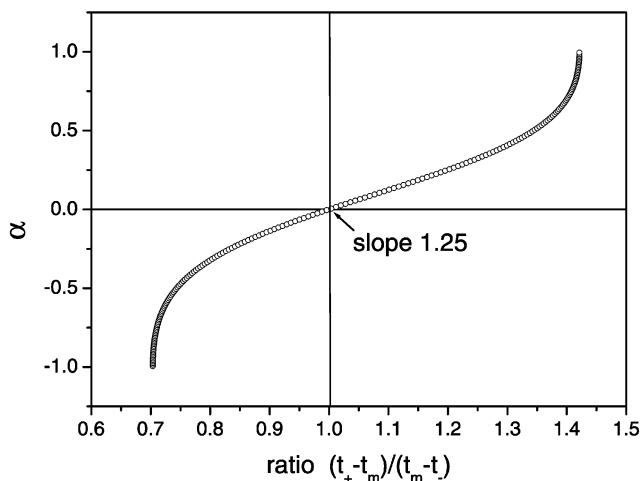


Figure 4. Graphical determination of α from the ratio of the right to left half width at half-height.

we are dealing with small values of this parameter even if it is proportional to the applied field.

From the experimental point of view, one can measure the area under the peak A , its height i_{\max} , the corresponding time t_m , t_- to the left and t_+ to the right of the maximum when the current is at half-height, then the width at half-height is $t_w = t_+ - t_-$. One can then compute the empirical switching time $\tau_m = A/2i_{\max}$ which should be equal to τ when $\alpha = 0$, the dimensionless ratios $(t_+ - t_m)/\tau_m$, $(t_m - t_-)/\tau_m$, t_w/τ_m and $(t_+ - t_m)/(t_m - t_-)$ can be computed as functions of α as shown in the appendix; see figure 4.

4. Results

Electro-optical studies were performed using the planar configuration. Commercial cells (EHC from Japan, 15 and $5.4 \mu\text{m}$ thick) coated with ITO (indium tin oxide) and rubbed polyimide were used, ensuring planar geometry. The sample alignment was improved by using an electric field of 10 to 20 Hz, $4 \text{ V} \mu\text{m}^{-1}$ applied for 10 to 30 min. During measurements, the alignment was checked using a polarizing microscope. The active area was 25 mm^2 . Uncertainties in cell spacings was $\pm 1 \mu\text{m}$. Slow cooling through the isotropic to SmA phase transition ($0.05^\circ\text{C min}^{-1}$) to avoid the appearance of focal-conic textures [13] leads to planar alignment. A classical electro-optical set-up was used for the measurement of the switching current, response time and apparent tilt angle [14]. All the measurements on heating and cooling runs were made between the isotropic and the SmC_A^* phases ranges, without going into the crystalline phase; the voltage across the resistor R_2 is digitized by an oscilloscope (Hewlett Packard 54502A) and the data stored in a computer for further

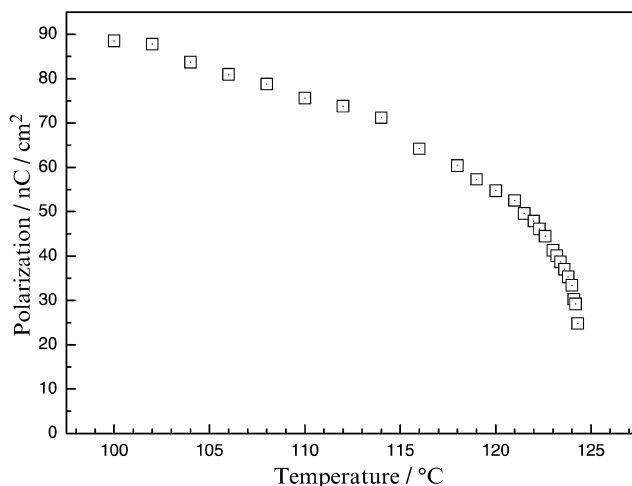


Figure 5. Polarization of the smectic C^* phase under field versus the temperature.

treatment. The ‘permanent’ polarization was determined by integrating the area under the polarization current peak, and the switching time τ_m was computed with the help of the area and the maximum of the polarization current peak upon field reversal.

The polarization (figure 5) under saturation conditions and the apparent tilt angle (figure 6) were measured at various temperatures ($E = 4.7 \text{ V} \mu\text{m}^{-1}$, $\nu = 41 \text{ Hz}$) for the polarization measurements, and $\nu = 0.1 \text{ Hz}$ for the apparent tilt angle. Such a field is sufficient to unwind the helical structure and accomplish the phase transition from SmC_A^* , SmC_{FI}^* , helielectric SmC^* and SmC_α^* phases to the unwound SmC^* . Both curves show a regular decrease with increasing temperature. The upper values are 88.5 nC cm^{-2} for the polarization and 31 degrees for the tilt angle.

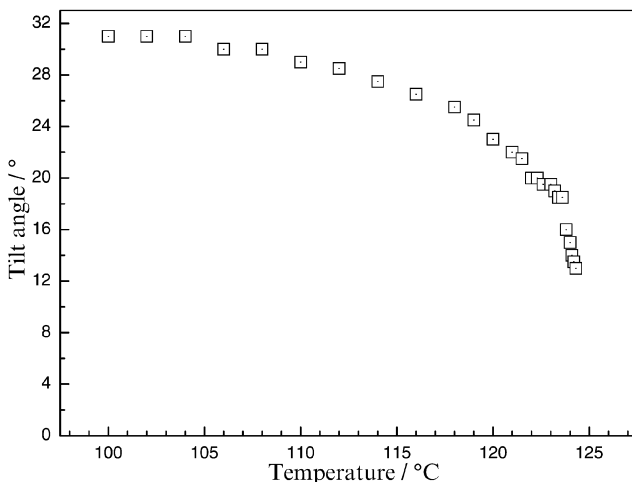


Figure 6. Tilt angle of the smectic C^* phase under field versus the temperature.

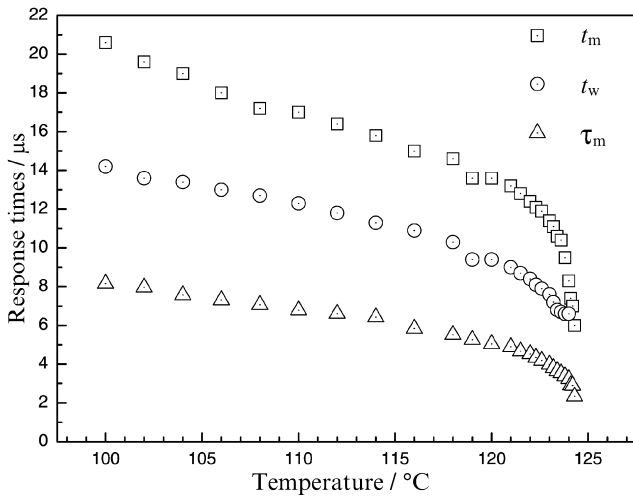


Figure 7. Response times t_m , t_w and τ_m of the smectic C* phase under field versus the temperature.

The other parameters t_m , τ_m and t_w are shown in figure 7. These experimental data revealed that the characteristic times are approximately proportional to each other as expected from the scaling law with respect to τ . They decrease monotonically with increasing temperature. As shown in figure 7, the behaviour of these times versus temperature showed no anomalies at the phase transition temperatures. The rotational viscosity η , which is related to rotations about the smectic C cone, is one of the most important parameters of the SmC* phase and strongly influences the switching time between the field-induced states of the FLC. Indeed some progress has been made in developing reasonably simple methods for viscosity determinations.

In the current response method, the rotational viscosity is usually calculated by means of the relationship [8, 9, 15]:

$$\eta = \frac{1}{1.76} t_w \mathbf{P}_S \mathbf{E}. \quad (8)$$

This has been demonstrated under the assumption $\alpha=0$ [9] in which case, one can also use:

$$\eta = \mathbf{P}_S \mathbf{E} \frac{A}{2i_{\max}} = \tau_m \mathbf{P}_S \mathbf{E}. \quad (9)$$

The result of our investigations is that both determinations are equivalent to within a few percent and the second one is shown in figure 8. The rotational viscosity of the liquid crystal shows practically a decrease with temperature far from the phase transition and falls in the vicinity of the phase transition (SmC $_{\alpha}$ * \leftrightarrow SmA).

The validity of the model has still not been assessed, so in order to compare with the theoretical expectations, we have

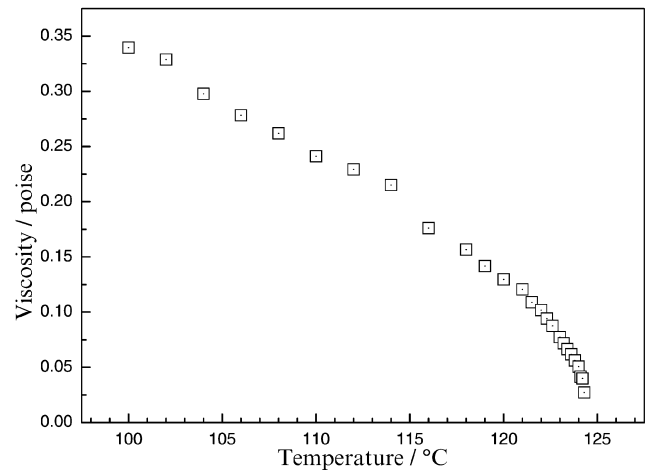


Figure 8. Estimated rotational viscosity of the smectic C* phase under field versus the temperature.

determined the four terms $(t_+ - t_m)/\tau_m$, $(t_m - t_-)/\tau_m$, t_w/τ_m and $(t_+ - t_m)/(t_m - t_-)$ as functions of temperature and plotted three of them in figure 9; the fourth should be compared with figure 3 if eventually the theory is validated, thus giving a graphical determination of α and thus ϵ_a .

In short we summarize the results thus: the asymmetry of the current peak appears to be what is expected for $\alpha > 0$ with values larger than allowed in the theory. Thus one can safely assert that the ‘rigid’ assumption of the angle Ψ being the same everywhere in the sample does not hold.

5. Discussion and conclusion

In this paper a tentative procedure for determining some physical parameters of a ferroelectric liquid

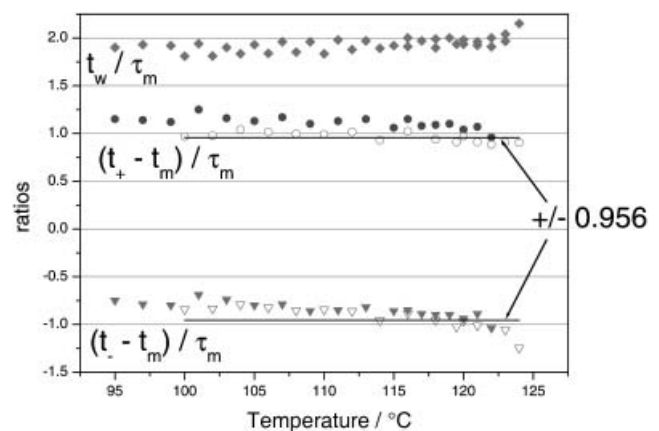


Figure 9. Experimental determination of the response times ratios as defined in figure 3 versus temperature for different sample thicknesses (15 μm for open symbols and 5.4 μm for full symbols). Note that these values are higher than the 0.956 upper limit of the theory thus ruling out the ‘rigid’ model.

crystal is described. The calculation described assumes the simplified uniform-director configuration that would be obtained in the case where the liquid crystal were free from interactions with its bounding surfaces. The main prediction is that one would obtain an asymmetric current peak, for example wider to the left than to the right for a negative dielectric anisotropy ϵ_a , which is to be expected for such molecules. This does not hold experimentally, as the asymmetry is of the other sense and much too high as an absolute value. A more realistic model would take into account that the angle Ψ cannot be uniform in space, which leads us to consider the case when many domains coexist that have different azimuthal angles at the time when the field switches up, due to the boundary conditions, or due to thermal agitation. The fact that the peak asymmetry is better seen for thinner samples, see e.g. figure 9, supports this view because the defects which are the starting points of the switching [9] are in this case closer together. An improved theory already exists [11, 12] in the case where the Ψ gradient is unidimensional, in which case the solution of the Sine–Gordon equation (6) is a progressive wave. The real solution is probably a mix of progressive waves and uniform rotation as discussed here.

Acknowledgements

We wish to acknowledge the support of Dr Nguyen Huu Tinh for the supply of the chemical compound and the Tuniso-French C.M.C.U. committee whose help was invaluable.

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APPENDIX

A: Calculation of the response times for the polarization reorientation in ferroelectric liquid crystals

We start at $t=0_-$ in the situation where the applied field and the permanent polarization lie in the layer plane and are oriented downwards, with $\cos \psi \simeq -1$ and $\psi = \pi$. When the applied field \mathbf{E} changes sign, the angle ψ goes from π to 0, so the derivative $\partial\psi/\partial t$ is negative.

The equilibrium of the viscous, elastic and electrical torques is given by:

$$\eta \frac{\partial\psi}{\partial t} = -\mathbf{P}_S \mathbf{E} \sin \psi + \epsilon_0 \epsilon_a \mathbf{E}^2 \sin^2 \theta \sin \psi \cos \psi + K \Delta\psi \quad (\text{A1})$$

At the end of \mathbf{P} 's reversal one gets $\partial\psi/\partial t=0$ everywhere and one assumes that above a given field value, one obtains $\psi=0$ or π almost everywhere except locally close to defects and thermal fluctuations where the elastic torque $K\Delta\psi$ has to compensate for the electric torques.

For lower voltages, the reorientation starts in disoriented domains and propagates as a progressive wave (soliton=solution of the Sine–Gordon equation). However, for higher voltages, we know that all response times obey a scaling law as they are proportional to $\tau = \eta/\mathbf{P}_S \mathbf{E}$.

In order to solve the equilibrium equation, we assume that the elastic torque is needed just at the beginning of the reorientation, otherwise $\psi(t)$ is the same everywhere. One gets:

$$\frac{\partial\psi}{\partial t} = \frac{-1}{\tau} \sin \psi (1 - \alpha \cos \psi) \quad (\text{A2})$$

with

$$\alpha = \epsilon_0 \epsilon_a \sin^2 \theta \frac{\mathbf{E}}{\mathbf{P}_S}. \quad (\text{A3})$$

The equation can be solved by multiplying on both sides by $\sin \psi$ and defining $u = -\cos \psi$ giving:

$$\frac{dt}{\tau} = \int du \left[\frac{1}{2(\alpha-1)(u+1)} - \frac{1}{2(\alpha+1)(1-u)} - \frac{\alpha^2}{(\alpha^2-1)(\alpha u+1)} \right]. \quad (\text{A4})$$

Defining t_0 as the time when $\psi = \pi/2$:

$$\frac{t-t_0}{\tau} = \frac{\ln(u+1)}{2(\alpha-1)} + \frac{\ln(1-u)}{2(\alpha+1)} - \frac{\alpha \ln(\alpha u+1)}{\alpha^2-1} \quad (\text{A5})$$

with the angle $\psi(t)$ it becomes:

$$\frac{t-t_0}{\tau} = \frac{1}{1-\alpha^2} \left[-\ln\left(\tan\frac{\psi}{2}\right) + \alpha \ln\left(\frac{1-\alpha\cos\psi}{\sin\psi}\right) \right]. \quad (\text{A6})$$

We see that the signs are coherent; indeed for t between 0 and t_0 we have $\psi > \pi/2$; for $t=t_0$ the second member vanishes and for $t > t_0$, $\psi < \pi/2$.

B: Relations between the experimental data and the theory

As stated in the paper, one can measure the peak area A , its height i_{\max} at time t_{\max} and the half-height times t_- , t_+ and $t_w = t_+ - t_-$. One must note that t_{\max} is equal to the time t_0 of the theory only if $\alpha=0$, otherwise t_0 is located 'somewhere' on the peak.

The first time that can be computed is $\tau_m = A/2i_{\max}$; it coincides with the fundamental response time $\tau = \eta/\mathbf{P}_S \mathbf{E}$ only when $\alpha=0$. We can also compute the normalized ratios $(t_+ - t_{\max})/\tau_m$, $(t_{\max} - t_-)/\tau_m$, t_w/τ_m and $(t_+ - t_{\max})/(t_{\max} - t_-)$ which indicate the symmetry of the current peak and can be calculated as a function of α in the theory.

From the theory, the summit of the current peak corresponds to the vanishing of the derivative di_p/dt . First the current reads:

$$i_p = -\mathbf{P}_S \frac{d\cos\psi}{dt} = \frac{A}{2\tau} \sin^2\psi [1 - \alpha\cos\psi]. \quad (\text{B1})$$

As $d\psi/dt$ is not zero, the peak summit corresponds to $di_p/d\psi=0$ for $\psi=\psi_m$:

$$\cos\psi_m = \frac{1 - (1 + 3\alpha^2)^{\frac{1}{2}}}{3\alpha}. \quad (\text{B2})$$

One readily gets:

$$i_{\max} = \frac{A}{2\tau_m} = \frac{A}{2\tau} \sin^2\psi_m (1 - \alpha\cos\psi_m) \quad (\text{B3})$$

$$\tau = \tau_m \sin^2\psi_m (1 - \alpha\cos\psi_m)$$

together with a first dimensionless number:

$$\frac{t_{\max} - t_0}{\tau_m} = \frac{1}{1-\alpha^2} \left[-\ln\left(\tan\frac{\psi_m}{2}\right) + \alpha \ln\left(\frac{1-\alpha\cos\psi_m}{\sin\psi_m}\right) \right] \quad (\text{B4})$$

$$\sin^2\psi_m (1 - \alpha\cos\psi_m).$$

In order to compute t_- and t_+ we need the angles ψ_- and ψ_+ , such that $i_p = i_{\max}/2$:

$$\sin^2\psi_{\pm} (1 - \alpha\cos\psi_{\pm}) = \sin^2\psi_m (1 - \alpha\cos\psi_m)/2 \quad (\text{B5})$$

so by defining $x = \cos\psi_{\pm}$:

$$(1-x^2)(1-\alpha x) = \frac{[2 + (1 + 3\alpha^2)^{\frac{1}{2}}][3\alpha^2 - 1 + (1 + 3\alpha^2)^{\frac{1}{2}}]}{27\alpha^2} = A(\alpha). \quad (\text{B6})$$

The terms $\cos\psi_-$ (resp. $\cos\psi_+$) are the negative (resp. positive) roots with a modulus lower than 1 of the third degree equation:

$$\alpha x^3 - x^2 - \alpha x + 1 - A(\alpha) = 0 \quad (\text{B7})$$

The system can be solved numerically and the plot of the dimensionless parameters versus α is to be found in figures 3 and 4.