

## Cell thickness dependence of liquid crystal parameters

Nitin Sood<sup>a,b</sup>, Samriti Khosla<sup>c\*</sup>, Darshan Singh<sup>b</sup> and S.S. Bawa<sup>a</sup>

<sup>a</sup>Engineering Materials Division, National Physical Laboratory, New Delhi 110 012, India; <sup>b</sup>Department of Physics, Punjabi University, Patiala 147 002, India; <sup>c</sup>Liquid Crystals Research Lab, Department of Physics, JCDAV College, Dasuya 144 205, India

(Received 15 September 2011; Revised 10 December 2011; Accepted for publication 12 December 2011)

Ferroelectric liquid crystal parameters, spontaneous polarization, and transition temperature were studied as a function of cell thickness. These parameters were found to increase with increasing cell thickness, but an exception was observed for the transition temperature in the case of a thin cell. A simple Landau model is presented to interpret the theoretical and experimental observations. The anomalous behavior is attributed to the electroclinic effect and is explained using the Landau model.

**Keywords:** electrical properties; spontaneous polarization; ferroelectric liquid crystals

### 1. Introduction

In 1975, Meyer *et al.* established the existence of ferroelectricity in liquid crystals (LCs). It was first discovered in the chiral smectic C (SmC\*) phase of LCs [1]. In this phase, a helical superstructure is generated, whose net polarization is zero (Figure 1). These ferroelectric LCs (FLCs) are highly suitable for display devices and offer high speed, high contrast, and switching characteristics when their intrinsic helical structure is suppressed [2–8]. FLCs have states with opposite polarization, which can be switched from one to another on a time scale of a few microseconds with the application of an external AC field sufficient to unwind the helix.

Spontaneous polarization ( $P_S$ ) and rotational viscosity ( $\gamma_\varphi$ ) are the major parameters that determine the switching time of FLC cells. In the past two decades, various methods were developed for the measurement of these parameters [9–16]. The literature review conducted in this study revealed that despite the extensive work performed by various research groups, very little attention has been paid to their cells' thickness dependence. The helical superstructure of the SmC\* phase is disturbed by the effect of the surface interactions. Moreover, the switching mechanism in electro-optic devices depends on the cell thickness, as the polarization and transition temperature are expected to be influenced by the thickness of the cell. The small-thickness sample cells also show electroclinic (EC) behavior.

The systematic investigation of the cell thickness dependence of the electrical parameters is expected to clarify the effect of the intersurface interaction between cell surfaces

and molecules on the electrical properties. To investigate the dynamics of molecules under the influence of such inter-surface interactions, an attempt was made to study the FLC parameters with varying cell thicknesses, using a universal procedure. The experiment that was conducted in this study involved two phenomena: the cell thickness dependence of the FLC parameters and the existence of the EC effect.

### 2. Experiment

Investigations were performed on the FLC mixture FLC-6304 (Rolic, Switzerland). FLC-6304 exhibits ferroelectric SmC\* (chiral SmC\*) and SmA\* phases, with the corresponding transition temperatures given by SmC\*–SmA-isotropic: 59°C, 64°C (Hoffmann La Roche, Rolic). It has high spontaneous polarization (110 nC/cm<sup>2</sup>), a short pitch (0.35  $\mu$ m), and a large tilt angle (27°) (Hoffmann La Roche, Rolic). The thermotropic mesomorphism and the transition temperatures of the material were also confirmed using an optical–thermal polarizing microscope and a differential scanning calorimeter (DSC-7 Perkin-Elmer, Mumbai, India). Planar cells with 100, 62.5, 25, 7.5, 6, and 3  $\mu$ m thicknesses were filled with the FLC mixture via capillary action at the isotropic temperature. A well-aligned texture was obtained by cooling the sample to room temperature at 0.1°C/min. The microtextures of the samples were visualized through a polarizing microscope (Model Select P1, Dewinter, New Delhi, India) interfaced with a computer.

In the setup that was used, a square wave was applied to the sample, and the output current was integrated from an

\*Corresponding author. Email: samritis@yahoo.com

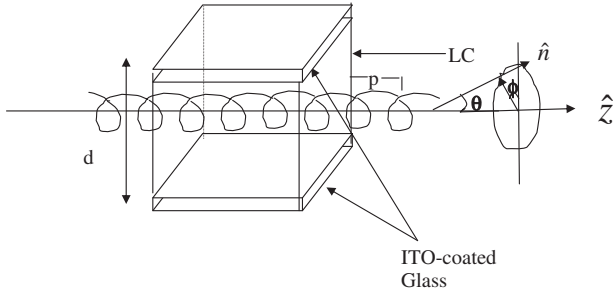


Figure 1. Helical structure of the SmC\* phase.

external capacitor. The output waveform was then obtained on a digitizing storage oscilloscope for further analysis. The circuit [14] is a modified form of current reversal method [13–16] (where resistance is used in the external circuit) and measures the response time ( $\tau$ ), azimuthal angle ( $\varphi_o$ ), spontaneous polarization ( $P_S$ ), rotational viscosity ( $\gamma_\varphi$ ), and dielectric permittivity ( $\epsilon_\perp$ ) in a single experiment. When a symmetric square wave is applied to the LC sample, the instantaneous value of the output voltage ( $V_o$ ) over capacitor  $C$  can be written as [13,14]

$$V_o = \frac{1}{C} \int_0^t i dt = \frac{V_{IN}}{RC} t + \frac{2V_{IN}C_{LCC}}{C} + \frac{P_S A}{C} \cos(\varphi(t)) \Big|_0^t$$

$$\equiv V_R + V_C + V_P,$$

where  $t = 0$  represents the instant of reversing the sign of the voltage,  $R$  is the resistance of the cell,  $C_{LCC} = (\epsilon_0 \epsilon_\perp A)/d$  is the capacitance of the cell,  $\epsilon_0$  is the permittivity of air,  $\epsilon_\perp$  is the permittivity of the LC, and  $A$  and  $d$  are the area and thickness of the cell, respectively. The shape of the waveform that was obtained is shown in Figure 2. The output voltage is the sum of three components, consisting of the resistive part ( $V_R$ ) (Region III), recharging of the cell capacitance ( $V_C$ ) (Region I), and the voltage across  $C$  due to the repolarization current in the cell ( $V_P$ ) (Region II).

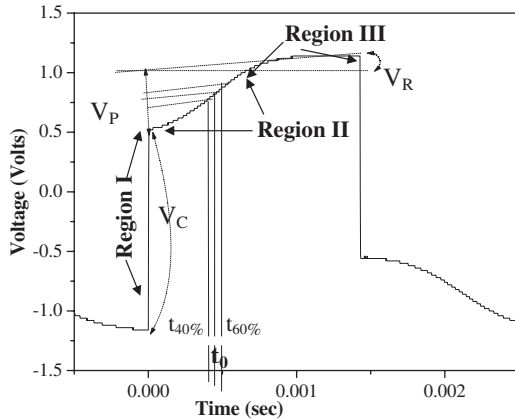


Figure 2. Shape of the waveform obtained in the case of the improved current reversal method.

The polarization was measured using the following equation [13,14]:

$$V_P = \frac{2AP_S}{C}.$$

### 3. Results and discussion

The temperature dependence of the electrical parameters of FLC as a function of cell thickness has been investigated by very few authors. As revealed by the literature review that was conducted in this study, only two groups have studied it experimentally [17,18]. In the present work, the temperature and cell thickness dependence of spontaneous polarization were studied. The results obtained were found to be reproducible.

The polarization behavior of sample cells with different thicknesses is shown in Figures 3 and 4. As

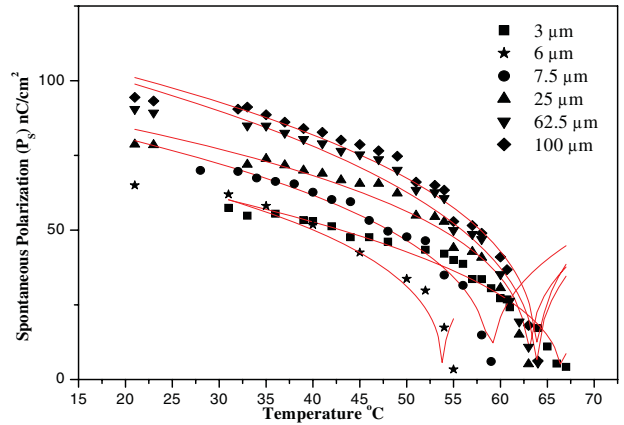


Figure 3. Variation of the spontaneous polarization as a function of temperature for the FLC-6304 sample: 3- $\mu\text{m}$  cell,  $\blacksquare$ ; 6- $\mu\text{m}$  cell,  $\star$ ; 7.5- $\mu\text{m}$  cell,  $\bullet$ ; 25- $\mu\text{m}$  cell,  $\blacktriangle$ ; 62.5- $\mu\text{m}$  cell,  $\blacktriangledown$ ; and 100- $\mu\text{m}$  cell,  $\blacklozenge$ .

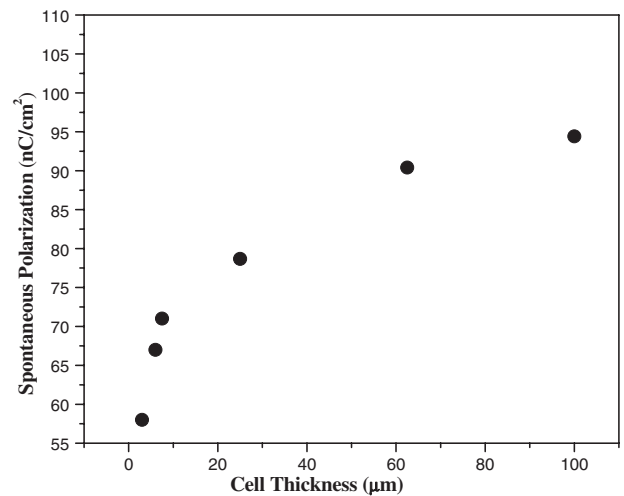


Figure 4. Cell thickness dependence of the spontaneous polarization ( $P_S$ ).

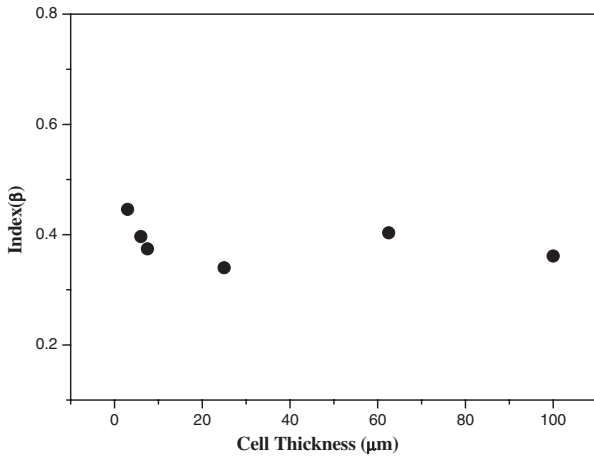


Figure 5. Cell thickness dependence of the critical index ( $\beta$ ).

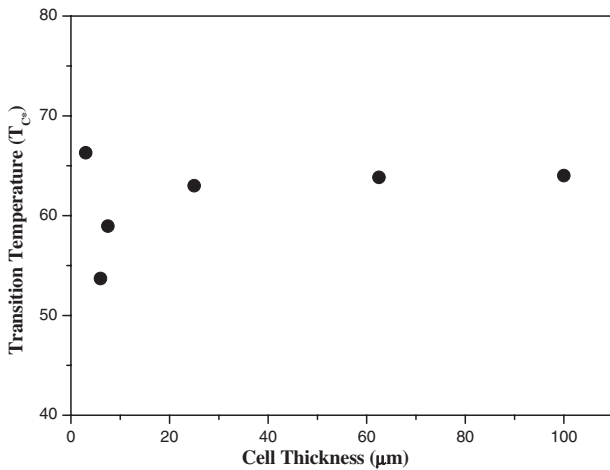


Figure 6. Cell thickness dependence of the transition temperature ( $T_{C^*}$ ).

expected theoretically, the temperature dependence shown in Figure 3 is observed to follow the power law given by

$$P_S = A(T - T_{C^*})^\beta. \quad (1)$$

Although  $P_S$  was observed to follow the power law in each case, an increase in its magnitude was observed with increasing cell thickness (Figure 4). The critical exponent ( $\beta$ ), as obtained from theoretical fit, is shown in Figure 5 for different cell thicknesses. Index  $\beta$  is almost constant, except for the 3- $\mu\text{m}$  cell. The small deviation from the theoretical value of 0.5 in each case is due to the field-induced terms [2,18–21]. The cell thickness dependence of  $T_{C^*}$  (SmC\*–SmA\* transition temperature) for FLC, as obtained from the curve fitting in Figure 3, is shown in Figure 6.  $T_{C^*}$  tends to decrease with diminishing cell thickness. An exception was observed, however, in the case of the 3- $\mu\text{m}$  cell, where an unusual increase in  $T_{C^*}$  was observed. Here, a shift of  $-6^\circ\text{C}$  was observed. A trace of the switching current after passing through the transition temperature, as seen in the specimen

with 3  $\mu\text{m}$  cell thickness, may be due to the smearing of  $T_C$  because of the EC effect that comes into play above a critical value of the biasing field. The increasing value of  $P_S$  and  $T_{C^*}$  with the cell thickness and anomalous behavior in the 3- $\mu\text{m}$ -thick sample can be explained in terms of the mathematical models cited below.

### 3.1. Cell thickness dependence

The decrease in  $P_S$  and  $T_{C^*}$  with diminishing cell thickness can be explained in terms of the generalized Landau expansion of free-energy density [22–27]. For a non-chiral system, at any temperature  $T$ , the free-energy density is given by

$$F = F_0 + \frac{1}{2}a\theta^2 + \frac{1}{4}b\theta^4 + \frac{1}{6}c\theta^6, \quad (2)$$

where  $\theta$  is the tilt angle, the primary-order parameter for the SmC–SmA transition;  $a = \alpha(T - T_C)$  is the only temperature-dependent term; and  $\alpha$ ,  $b$ , and  $c$  are the positive constants for the case of the second-order transition. The values of these constants are very small near the transition temperature.  $T_C$  is the transition temperature from the SmC–SmA phase and  $F_0$  is the singular part of  $F$ .

The expression of free energy in the case of an unwound system (for which the wave vector is  $q = 0$ ) for the SmC\*–SmA\* transition is given by [22–27]

$$F^*(\theta, E) = F_0 + \frac{1}{2}a\theta^2 + \frac{1}{4}b\theta^4 + \frac{1}{6}c\theta^6 + \frac{1}{2\chi_0}P^2 - CP\theta - \frac{1}{2}\Omega P^2\theta^2 + \frac{1}{4}\eta P^4 - EP. \quad (3)$$

Chiral interactions are introduced into the equation by the term  $P^2/2\chi_0$ , where  $\chi_0$  is the high-frequency dielectric constant, which is due to the effect of dipolar ordering.  $C$  is a piezoelectric bilinear coupling term,  $\Omega$  is a biquadratic coupling term, and the term  $\eta$  (a constant) was added to stabilize the system. The last term was included to represent the decrease in energy on the field application.

Minimizing Equation (3) with respect to  $P$ ,

$$\frac{\partial F^*}{\partial P} = 0,$$

gives

$$\left(\frac{1}{\chi_0} - \Omega\theta^2\right)P - C\theta + \eta P^3 - E = 0.$$

Recently, Gouda et al. [28] pointed out that the term  $\eta$  is related to the low-temperature saturation of polarization, and is therefore irrelevant near the transition temperature.

Thus, at higher temperatures, the following is obtained:

$$\left(\frac{1}{\chi_0} - \Omega\theta^2\right)P - C\theta = E. \quad (4)$$

From Equation (4), the following is obtained:

$$\begin{aligned} P &= \frac{(E + C\theta)}{((1/\chi_0) - \Omega\theta^2)} = \frac{\chi_0(E + C\theta)}{(1 - \chi_0\Omega\theta^2)} \\ &= \chi_0(E + C\theta)(1 - \chi_0\Omega\theta^2)^{-1}. \end{aligned} \quad (5)$$

In the case of the second-order transition, the tilt angle continuously approaches zero at the transition temperature, and Equation (5) is reduced to the form  $P = E\chi_0$ .

Applying condition  $\partial F^*/\partial\theta = 0$ , we get

$$a\theta + b\theta^3 + c\theta^5 - CP - \Omega P^2\theta = 0.$$

Substituting P from Equation (5), neglecting the contribution from the higher powers of  $\theta$ , the following is obtained [29]:

$$(a - C^2\chi_0 - E^2\Omega\chi_0^2)\theta - CE\chi_0 = 0. \quad (6)$$

Considering the first term in Equation (6) [29],

$$\begin{aligned} (a - C^2\chi_0 - E^2\Omega\chi_0^2)\theta &= a^*\theta = (\alpha(T - T_c) - C^2\chi_0 - E^2\Omega\chi_0^2)\theta \\ &= \alpha\left(T - T_c - \frac{C^2\chi_0}{\alpha} - \frac{E^2\Omega\chi_0^2}{\alpha}\right)\theta \\ a^*\theta &= \alpha(T - T_{C^*})\theta. \end{aligned} \quad (7)$$

Now,  $a^* = \alpha(T - T_{C^*})$  is a temperature-dependent term where  $T_{C^*}$  is the SmC\*-SmA\* phase transition temperature and is given by

$$T_{C^*} = T_c + \frac{C^2\chi_0}{\alpha} + \frac{E^2\Omega\chi_0^2}{\alpha}.$$

Using Equation (5) at the transition,

$$T_{C^*} = T_c + \frac{C^2\chi_0}{\alpha} + \frac{P^2\Omega}{\alpha} \quad (8)$$

The mean field coefficients  $C$ ,  $\alpha$ ,  $\chi_0$ , and  $\Omega$  have been found to be positive [30]. As the last two terms are always positive in Equation (8), the temperature  $T_{C^*}$  always increases. Also, except for polarization, all the other terms in Equation (8) are nearly constant for a particular FLC compound and, hence, the transition temperature ( $T_{C^*}$ ) reaches a maximum value when  $P$  becomes  $P_{\max}$ . Equation (8) can thus be written as

$$T_{C^* \max} = T_c + \frac{C^2\chi_0}{\alpha} + (P_{\max})^2 \frac{\Omega}{\alpha}, \quad (9)$$

where  $P_{\max}$  and  $T_{C^* \max}$  are the maximum values of the polarization and transition temperature, respectively. The critical

temperature for FLC can also be written as [31]

$$T_{C^*} = T_{C^* \max} - \left(\frac{\Pi^2 g}{2\alpha'}\right) \frac{1}{(d + (2g/|W|))^2}, \quad (10)$$

where  $T_{C^* \max}$  is the maximum critical temperature when thickness  $d \rightarrow \infty$ ,  $g$  is the elastic modulus, and  $W$  is the effective anchoring energy. From Equations (8) and (9), the following is obtained:

$$T_{C^*} = T_{C^* \max} + \frac{\Omega}{\alpha}(P^2 - P_{\max}^2). \quad (11)$$

Comparing Equations (10) and (11) yields the following:

$$P = \left[ P_{\max}^2 - \left(\frac{\Pi^2 g}{2\Omega}\right) \frac{1}{(d + 2g/|W|)^2} \right]^{1/2}. \quad (12)$$

Hence, from Equation (12), it is clear that as  $d \rightarrow \infty$  and  $P \rightarrow P_{\max}$ , the spontaneous polarization increases with the cell thickness. Also, when  $d|W| \gg g$ , the following is obtained:

$$P = \left[ P_{\max}^2 - \left(\frac{\Pi^2 g}{2\Omega}\right) \frac{1}{(d)^2} \right]^{1/2}. \quad (13)$$

Equation (13) also shows that polarization increases with cell thickness. At  $d|W| \ll g$ , the dependence of  $P(d)$  on the cell thickness is very small. Equation (8), (10), or (11) conforms to an increase in transition temperature with cell thickness, as observed in Figures 3 and 4.

### 3.2. Anomalous behavior

The anomaly in the case of the 3- $\mu\text{m}$  sample, which shows an abnormal jump in its transition temperature, can be explained in terms of the EC effect. In 1975, Meyer *et al.* showed that in the SmC\* phase, each layer exhibits PS due to the chiral nature of the molecules. The other consequence of chirality is the helical superstructure due to the precession of the director around the layer normal. The application of an electric field with sufficient strength unwinds the helix of the SmC\* phase, producing a homogenous tilt and, hence, a net polarization current. This coupling between the tilt and polarization can induce itself in the SmA\* phase, for a field above a critical value, and the effect is called the *EC effect*. The director in the SmA\* phase, usually parallel to the layer normal, becomes tilted at some angles. Thus, the SmC\* and SmA\* phases show the same homogeneous tilt (i.e. unwound) and have identical symmetries [32]. The phase transition from SmC\* to SmA\*, which is generally second-order, becomes less abrupt. At this point, the transition temperature is difficult to analyze and appears to increase in value.

In this study, the transition from SmC\* to SmA\* in the 3- $\mu\text{m}$  cell was found to be less abrupt and conforms to the induction of the tilt angle in the SmA\* phase. The EC effect

was observed only in this sample because in thin samples, a smaller field is required to attain the EC effect.

The EC behavior, particularly the required critical field and the shift in the transition temperature, can be interpreted in terms of a simple Landau model. The Landau free-energy density is given by Equation (3).

$$F^*(\theta, E) = F_0 + \frac{1}{2}a\theta^2 + \frac{1}{4}b\theta^4 + \frac{1}{6}c\theta^6 + \frac{1}{2\chi_0}P^2 - CP\theta - EP.$$

Here, the contribution from the biquadratic term was neglected as the EC effect strongly depends on bilinear coupling. To calculate the value of the critical field required to observe the EC effect, the following condition was applied:

$$\frac{\partial F^*}{\partial \theta} = 0.$$

The equation for the electric field in terms of the tilt angle that was obtained is

$$E = \frac{1}{C\chi_0} \{ [\alpha(T - T_C) - C^2\chi_0]\theta + b\theta^3 + c\theta^5 \}. \quad (14)$$

At the critical field, the following conditions must be satisfied [33]:

$$\frac{\partial E}{\partial \theta} = 0, \quad \frac{\partial^2 E}{\partial \theta^2} = 0.$$

Using these conditions, the following are obtained:

$$T_{Cr^*} = T_C + \frac{9b^2}{20\alpha c} + \frac{C^2\chi_0}{\alpha} \quad (15)$$

and

$$E_{Cr^*} = \frac{1}{C\chi_0} \left[ \frac{6\sqrt{3}}{25\sqrt{10}} \frac{(-b)^{5/2}}{c^{3/2}} \right], \quad (16)$$

where  $T_{Cr^*}$  is the transition temperature when the field is above the critical value. The transition temperature at the zero field, after neglecting the biquadratic term, is [33]

$$T_{C^*} = T_C + \frac{3b^2}{16\alpha c} + \frac{C^2\chi_0}{\alpha}. \quad (17)$$

The difference in the transition temperature for zero-field Equation (17) and critical-temperature Equation (15), or the change in transition temperature  $T_{C^*}$  due to the EC effect, is given by

$$T_{Cr^*} - T_{C^*} = \frac{21b^2}{80\alpha c}. \quad (18)$$

As  $\alpha$ ,  $b$ , and  $c$  are different for different FLCs, the increase in  $T_{C^*}$  due to the EC effect is also different. Based on the order of each constant, however, the transition temperature can vary up to a few K, as can be observed in Figures 3 and 6.

Equation (15) can be modified using Equation (8) (neglecting the biquadratic term).

$$T_{Cr^*} = T_{C^*} + \frac{9b^2}{20\alpha c}. \quad (19)$$

Thus, the critical-field equation becomes

$$E_{Cr^*} = \frac{1}{C\chi_0} \left\{ \left[ \alpha \left( T_{Cr^*} - T_C + \frac{9b^2}{20\alpha c} \right) - C^2\chi_0 \right] \theta + b\theta^3 + c\theta^5 \right\}. \quad (20)$$

As discussed in Section 3.1 and as proven by Pikin and Yoshina [31],  $T_{C^*}$  decreases with decrease in cell thickness. As a result, the value of the critical field required to attain the EC effect is comparatively low in thin samples for a given material.

#### 4. Conclusions

The spontaneous polarization is measured from the relaxation voltage curve corresponding to the repolarization current of the FLC material with high polarization. The results of this study coincide with the results predicted by the theory. The temperature dependence of polarization was observed to follow the power law, and the critical exponent of the obtained polarization curve was found to be close to 0.5 eV.

The cell thickness dependence of polarization was observed to show an increase in its magnitude with cell thickness. The critical exponent and transition temperature obtained from the theoretical fit of polarization vs. temperature curve were also studied for various cell thicknesses. Although  $\beta$  remains constant and  $T_{C^*}$  increases with increasing cell thickness, an anomaly is observed for the 3- $\mu\text{m}$  cell. A simple Landau model is presented to support the experimental results. The anomalous behavior is attributed to the EC effect and is explained using the Landau model.

#### Acknowledgement

The authors thank the referees for their valuable suggestions that helped them in improving the quality of this paper.

#### References

- [1] R.B. Meyer, L. Liebert, L. Strzelecki and P. Keller, *J. Phys. Lett. (Paris)* **36**, L69 (1975).
- [2] L.A. Bersnev, L.M. Blinov, M.A. Osipov and S.A. Pikin, *Mol. Cyst. Liq. Cryst.* **158A**, 1 (1988).
- [3] Y. Kimura and R. Hayakawa, *Jpn. J. Appl. Phys.* **32**, 427 (1993).
- [4] K.K. Raina and H.J. Coles, 9th IEEE International Symposium on Application of Ferroelectrics, Vol. CH3416-50-7803, p. 788, USA, (IEEE, Varsity Park, PA, 1995).

- [5] K.K. Raina and J.K. Ahuja, *Mol. Cryst. Liq. Cryst.* **338**, 125 (1999).
- [6] N.A. Clark and S.T. Lagerwall, *Appl. Phys. Lett.* **36**, 11 (1980).
- [7] L.A. Bersnev, V.G. Chigrinov, D.I. Drgachev, E.P. Poshtadaev, J. Funfschilling and M. Schadt, *Liq. Cryst.* **5**, 1171 (1989).
- [8] J. Funfschilling and M. Schadt, *J. Appl. Phys.* **66**, 3877 (1989).
- [9] J.K. Ahuja and K.K. Raina, *Jpn. J. Appl. Phys.* **39**, 4076 (2000).
- [10] L.M. Blinov, M.I. Barnik, M. Ozaki, N.M. Shtykov and K. Yoshino, *Phys. Rev. E*, **62** (6), 8091 (2000).
- [11] M.L.M. Madhumohan, B.V.S. Gaud, P.A. Kumar and V.G.K.M. Pissipati, *Mater. Res. Bull.* **34** (14), 2167 (1999).
- [12] S.S. Bawa, A.M. Biradar and S. Chandra, *Jpn. J. Appl. Phys.* **26**, 189 (1987).
- [13] S. Khosla and K.K. Raina, *Indian J. Pure Appl. Phys.* **42**, 49 (2004).
- [14] S. Khosla, N. Sood, S.S. Bawa and D. Singh, *J. Inform. Disp.* **12** (3), 129 (2011).
- [15] Yu. P. Panarin and V.M. Vaksman, *Mol. Mater.* **1**, 147 (1992).
- [16] V. Panov, J.K. Vij and N.M. Shtykov, *Liq. Cryst.* **28** (4), 615 (2001).
- [17] J.S. Patel and G.W. Goodby, *Chem. Phys. Lett.* **137**, 91 (1987).
- [18] H. Miyata, M. Maeda, I. Suzuki, *Liq. Cryst.* **20** (3), 303 (1996).
- [19] S.L. Srivastava, *Proc. Natl. Acad. Sci. India*, **LXIII** (63), 311 (1993).
- [20] S.L. Srivastava, V.K. Agarwal, *Indian J. Pure Appl. Phys.* **31**, 59 (1993).
- [21] S. Wrobel, S. Hiller, M. Pfeiffer, M. Marzec, W. Haase, *Liq. Cryst.* **18** (1), 21 (1995).
- [22] I.C. Khoo, *Optics and Nonlinear Optics of Liquid Crystals* (World Scientific, Singapore, 1993).
- [23] T. Carlsson, B. Zeks, C. Fillipic, A. Levstik and R. Blinc, *Mol. Cryst. Liq. Cryst.* **163**, 11 (1988).
- [24] A. Levstik, T. Carlsson, C. Fillipic, I. Levstik and B. Zeks, *Phys. Rev. A* **35** (8), 352 (1987).
- [25] C. Fillipic, T. Carlsson, A. Levstik, B. Zeks and R. Blinc, *Phys. Rev. A* **38** (11), 5833 (1988).
- [26] T. Carlsson, B. Zeks, C. Fillipic and A. Levstik, *Phys. Rev. A* **42** (1), 877 (1990).
- [27] T. Carlsson, B. Zeks, C. Fillipic, A. Levstik and R. Blinc, *Mol. Cryst. Liq. Cryst.* **163**, 11 (1988).
- [28] F. Gouda, T. Carlsson, G. Andersson and S.T. Lagerwall, *Liq. Cryst.* **16**, 315 (1994).
- [29] S.T. Lagerwall, *Ferroelectric and Antiferroelectric Liquid Crystals* (Wiley-VCH, New York, 1999).
- [30] F. Giesselmann and P. Zugenmaier, *Phys. Rev. E* **52**, 1762 (1995).
- [31] S.A. Pikin and K. Yoshina, *Jpn. J. Appl. Phys.* **30**, L557 (1981).
- [32] S. Garoff and R.B. Mayer, *Phys. Rev. A* **19** (1), 338 (1979).
- [33] Ch. Bahr and G. Heppke, *Phys. Rev. A* **41** (8), 4335 (1990).