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On the thickness dependence of ferroelectric polarization in smectic C* liquid crystal cells

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The thickness dependence of ferroelectric polarization in a SmC* liquid crystal exhibiting a large value of polarization has been investigated. It is observed that the saturation value of the polarization shows no significant thickness dependence. This observation is in accordance with a recent theoretical framework developed by Galerne. It is shown that the wall thickness, where the polarization undergoes a reversal, and hence the self-screening effect, is insignificant in the material studied.

1. Introduction

Since the first observation of ferroelectricity in a smectic C* (SmC*) liquid crystal [1], spontaneous polarization ($\mathbf{P}_s$) in SmC* is one of the most well studied properties; it is essentially a macroscopic bulk property derived from symmetry breaking [1]. The electro-optic properties of liquid crystals are typically studied in a surface-stabilized geometry [2]. Such a bistable state is generally achieved in liquid crystal cells that are about 2 μm or less in thickness.

A priori, the value of $\mathbf{P}_s$, at least in the saturated regime (at temperatures well into the SmC* phase), is not expected to have any dependence on thickness. However, Patel and Goodby [3] showed that for the ferroelectric liquid crystal material, DOBAMBC, the spontaneous polarization does indeed show a thickness dependence in a range of cell thickness, 3–18 μm; beyond this thickness, the polarization remained unchanged. This thickness dependence has been interpreted in terms of the screening of polarization by ions in the liquid crystal cell. In the presence of an electric field, the ions move towards the electrodes. The screening is most effective in thin cells because the ions have sufficient time to move to the electrodes during field switching. As the cell thickness increases, the transit time for ions to move towards the electrodes increases, resulting in less effective screening than in thin cells and hence the lack of a thickness dependence beyond 18 μm.

In a more recent paper, Galerne [4], re-examined this hypothesis and showed that the thickness dependence observed by Patel and Goodby is not due to screening by ions (though this contribution may still be present) but rather due to a self-screening mechanism arising from the polarization itself. Self-screening arises as follows: for planar boundary conditions, the ferroelectric liquid crystal molecules have their dipoles pointing into the bulk of the liquid crystal cell as this configuration minimizes the energy; this is supported by experimental evidence [5]. With an applied field, the polarization vector is predominantly in the direction of the field in the bulk and at one of the electrodes. Since the dipoles at the two electrodes are pointing in opposite directions, the polarization vector reverses direction through a wall d_0 of finite thickness as shown in figure 1. The presence of the wall leads to a dimunition of the polarization. Clearly, therefore, the wall thickness determines the extent of self-screening observed.

The experiments of Patel and Goodby were conducted on DOBAMBC, which exhibits only a small value of polarization ($< 10\,\mathrm{nC\,cm^{-2}}$). It would be important to

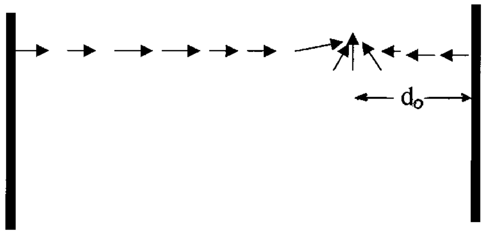

Figure 1. Schematic of the orientation of polarization vectors. The orientation of dipoles at the surfaces leads to the formation of a wall whose thickness dictates the extent of self-screening. The wall moves from one end of the electrode to the other depending on the sign of the field. d_0 signifies the thickness of the wall arising from polarization screening.

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verify if the screening effects discussed above are also valid for materials exhibiting large values of polarization. Such information would also be useful for practical applications in view of the V-shaped switching exhibited in thin cells by ferroelectric liquid crystals (FLCs) with large polarization values [6]. We have now measured the thickness dependence of $\mathbf{P}_s$ for a material exhibiting large polarization (> 200 nC cm^{-2}) values. We show that the saturation value of the measured $\mathbf{P}_s$ is in fact not thickness dependent. We also show that this result is in accord with Galerne's theoretical argument in the limit of very thin screening wall thickness.

2. Experimental

The ferroelectric material used, named 12KN5DSi, has been synthesized in our laboratory, and exhibits the following phase sequence as determined by differential scanning calorimetry:

SmC* 41.5 C SmA 59 C Iso (°C).

The liquid crystal cell was prepared using two glass plates, one with a patterned gold electrode, and the other with a transparent ITO electrode and a SiO alignment layer (EHC. Co. Japan). The patterned electrode area was 0.36 cm^2. This allowed us to measure the thickness of the cell accurately through capacitance measurements. Cells of varying thicknesses were fabricated using glass bead spacers and the plates assembled together and glued on the edges using 5 minute epoxy. The liquid crystal was aligned by cooling slowly (0.01°C min^{-1}) from the isotropic phase to the smectic A phase in the presence of an electric field (5 V μm).

The polarization measurements were made using the triangular wave method [7]. A commercially available instrument, APT III from Display Tech Instruments was used for all the measurements. For each cell, the polarization was first measured as a function of voltage to determine the saturation voltage; this value of the voltage was then used to determine the temperature dependence. The measurements were performed at a frequency of 7 Hz. The software allows measurement of the polarization current from the polarization peak and corrects for any capacitive and resistive currents. The temperature of the sample was controlled using an Instec hotstage and associated PID controller.

3. Results and discussion

Measurements were carried out over a range of temperature in the SmC* phase until the temperature was reached at which the polarization remained unchanged. Experimental data are shown for the various cell thicknesses in figure 2. The most important feature of the data is that the saturated value of the measured polarization does not show the kind of significant dependence on thickness observed for DOBAMBC. We shall now analyse the data using Galerne's approach which enables us to determine the true value of the polarization **P** from the measured data (for the remaining discussion, we use the notation **P** used in [4] for polarization).

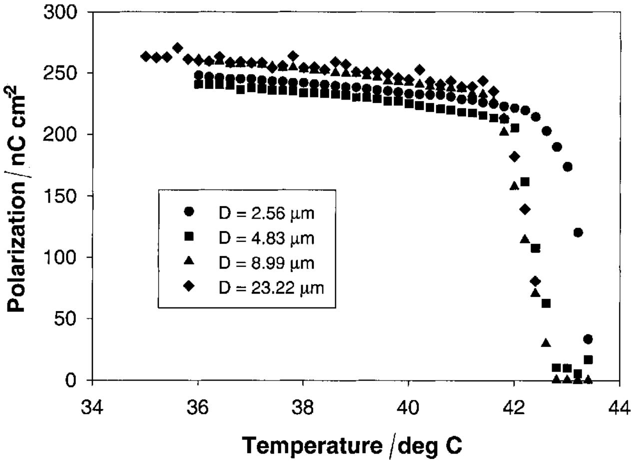

Figure 2. Plot of the spontaneous polarization as a function of temperature for cells of thicknesses, 2.6, 4.9, 8.9 and 23.2 μm. It is observed that there is a slight change in the transition temperature for the cell with the smallest thickness; this result is similar to the effect seen in earlier studies [8].

P may be obtained from the measured value of the polarization **P***, the cell thickness *D* and the wall thickness d_0 wherein the polarization vector undergoes a reversal. The following equation represents the relation between these parameters:

$$\mathbf{P}^* = \frac{\mathbf{P}(D - 2d_0)}{D}. \tag{1}$$

This equation may be rewritten for the sake of convenience as:

$$\mathbf{P}^* = \mathbf{P} - \frac{2\mathbf{P}d_0}{D}. \tag{2}$$

Thus a plot of **P*** vs $1/D$ yields a straight line whose intercept represents the true polarization **P**. The slope can then be used to extract the wall thickness d_0.

To compare polarization values for different thicknesses, we choose **P*** data at a temperature of 40°C. This is a temperature well removed from the transition so that pre-transitional effects do not play a role in determining the value of the polarization. The fit of **P*** vs $1/D$ yields the true value of polarization (243.1 nC cm^{-2}) and the wall thickness (0.08 μm). The experimental data points and the linear fit are shown in figure 3. Table 1 shows values of *D*, **P***, and the % difference between the measured and true value of polarization. It is observed that the maximum deviation between the measured

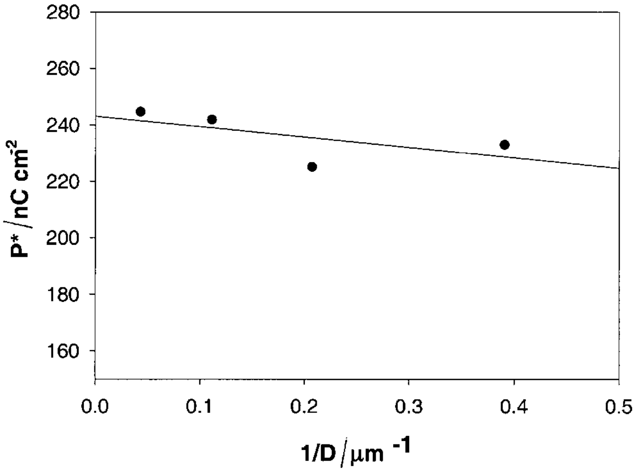

Figure 3. Plot of **P*** vs 1/*D* showing the fit to the Galerne equation. The intercept gives the true value of the polarization and the wall thickness is extracted from the slope of the straight line.

Table 1. Experimentally measured values of the polarization of 12KN5DSi for different cell thicknesses; also shown is the % difference between the true value of the polarization obtained from the fit and the measured values.

Cell thickness *d*/μm	Measured **P***/nC cm^{-2}	% difference (**P*** − **P**)
2.56	233.04	4.3
4.83	225.14	8.0
8.99	241.9	5.0
23.22	244.67	6.4

values and the true value is 8%. To assess the sensitivity of the value of true polarization to the fit, the third data point in the figure corresponding to a thickness of 4.8 μm is omitted and the linear least squares fit performed again using the remaining three data points. The statistical quality of the fit improves, and the intercept value of the polarization is now 245.9 nC cm^{-2} which is a difference of only 1% from the intercept value obtained using all four data points. Considering that a typical measurement uncertainty of the polarization is of the order of 3%, this difference in the intercept value is negligible.

To compare with earlier literature data on DOBAMBC, we have shown in table 2 experimental values obtained by Patel and Goodby and the fit value reported by Galerne. The true value of polarization from the fit is 2.25 nC cm^{-2} at 1°C from the transition. Note however, from the third column in table 2, that the difference between the measured and true values is significantly higher. Also, the value of d_0 for DOBAMBC is 0.8 μm, an order of magnitude larger than for 12KN5DSi (0.08 μm). It is likely that this difference is responsible for the lack of a pronounced cell thickness dependence in our experimental results. The small wall thickness translates into an insignificant self-screening of the polarization. Hence, our polarization data on 12KN5DSi (exhibiting little dependence on thickness) as well as the data of Patel and Goodby on DOBAMBC (which show pronounced dependence on thickness) can be explained quantitatively by the self-screening mechanism proposed by Galerne [4].

Table 2. Data taken from Patel and Goodby [2] showing values of the polarization as a function of cell thickness. Note that the % difference between the true value obtained from Galerne's fit is significantly different for various cell thicknesses.

Cell thickness *d*/μm	Measured **P***/nC cm^{-2}	% difference (**P*** − **P**)
3.0	1.12	101
5	1.38	63
9.0	1.85	22
18	2.06	9.2

As pointed out by Galerne, there may still be a small but finite contribution from ions in screening the polarization. Its existence can be detected by observing the frequency dependence of polarization and is negligible in our cells. The self-screening effect however is as fast as the polarization reversal itself during switching and therefore cannot show a frequency dependence as the ionic current does.

4. Summary

We have shown experimentally, for the first time, that polarization does not have a significant dependence on cell thickness in ferroelectric liquid crystals, if the self-screening effect arising from the reversal of the polarization vector in the bulk through a wall is small. Although this is apparent from theoretical considerations, our experiments establish this on a quantitative basis. The screening of polarization has important implications for applications [8–11] and our results are therefore relevant from this point of view.

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