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## Liquid Crystals

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# Memory effect in thin samples of chiral smectic C liquid crystals

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# Memory effect in thin samples of chiral smectic C liquid crystals

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On applying an appropriate electric field to a thin sample of ferroelectric liquid crystal a texture of long domains appears perpendicular to the smectic layers. We present observations showing that such treatment of the cell leads to its perfect bistability (i.e. a memory effect). The analysis of the equilibrium equations describing smectic liquid crystal elasticity shows how the director orientation is fixed both by a bulk deformation caused by the electric field treatment and by surface anchoring.

## 1. Introduction

Recently, a memory effect was reported for thin samples of chiral smectic C liquid crystals, $S_C^*$, where a texture of long domains was induced by applying an appropriate electric field [1–5]. When an alternating electric field of a reasonably high amplitude was applied for several seconds, long domains perpendicular to the smectic layers appeared. A similar long domain texture was also created in thicker samples (with sample thickness 25 $\mu$m) in the smectic A phase of a ferroelectric liquid crystal where the sample alignment was improved by an AC electric field 20 Hz, $4 \times 10^3$ kV/m [6]. An analogous stripe texture was reported in [7] where the planar $S_C^*$ sample, about 100 $\mu$m thick, was subjected to dilatation strains. The long domains described in [1–6] can be connected with the creation of a system of parabolic focal conics [8] as shown in [6]. Models based on curvature walls [9] describing this domain texture have also been proposed [6].

Here we will discuss a connection between the electric field induced domain texture and the observed memory effect using the elasticity of $S_C^*$ liquid crystals. The elastic free energy density of a thin $S_C^*$ liquid crystal sample will be approximated by the $S_C$ elastic free energy density because we start from the $S_C^*$ state where the helicoidal structure is unwound by surface anchoring. In the coordinate system $(x, y, z)$ the smectic layers are perpendicular to the $z$ direction (see figure 1). The director orientation in the layers of a $S_C$ phase is described by the $\mathbf{t}$ vector which is the projection of the director on to the layer plane. The rotation of the $\mathbf{t}$ vector around the $z$ axis is characterized by an angle $\Phi$, defined by $\mathbf{t} = (\cos\Phi, \sin\Phi, 0)$. For small gradients of the layer displacement $u$ and for large values of $\Phi$ describing the $\mathbf{t}$ vector orientation Kléman's expression for the $S_C$ free energy density [9] can be

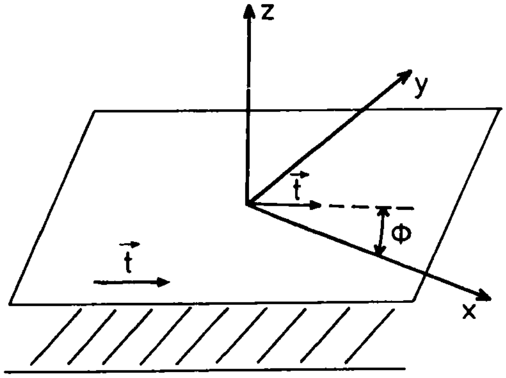

Figure 1. The coordinate system in the layer of a $S_C^*$ liquid crystal. The smectic layer coincides with the $(x, y)$ plane and the $z$ axis is perpendicular to layers. The $\mathbf{t}$ vector is the orientation of director projection on to the smectic layer. The inclination of the $\mathbf{t}$ vector from the $x$ axis is described by the angle $\Phi$.

simplified to

$$
\begin{aligned}
f = {} & \frac{A_{11}}{2}\left(\frac{\partial^2 u}{\partial x\,\partial y}\right)^2 + \frac{A_{12}}{2}\left(\frac{\partial^2 u}{\partial x^2}\right)^2 + \frac{A_{21}}{2}\left(\frac{\partial^2 u}{\partial y^2}\right)^2 \\
& + \frac{B_1}{2}\left(\left(\frac{\partial \Phi}{\partial x}\right)^2 + \left(\frac{\partial \Phi}{\partial y}\right)^2\right) + \frac{B_3}{2}\left(\frac{\partial \Phi}{\partial y}\right)^2 + \frac{\bar{B}}{2}\left(\frac{\partial u}{\partial z}\right)^2 \\
& + B_{13}\frac{\partial \Phi}{\partial z}\left(\frac{\partial \Phi}{\partial x}\cos\Phi + \frac{\partial \Phi}{\partial y}\sin\Phi\right) + C_1\frac{\partial^2 u}{\partial x\,\partial y}\left(\frac{\partial \Phi}{\partial x}\cos\Phi + \frac{\partial \Phi}{\partial y}\sin\Phi\right) \\
& + C_2\frac{\partial^2 u}{\partial y^2}\left(\frac{\partial \Phi}{\partial y}\cos\Phi - \frac{\partial \Phi}{\partial x}\sin\Phi\right). \qquad (1)
\end{aligned}
$$

In this equation the parameters $A_{11}$, $A_{12}$ and $A_{21}$ and the elastic constants describing a layer curvature energy and $\bar{B}$ characterizes the layer compressibility. $B_1$ and $B_3$ are the elastic constants for the $\mathbf{t}$ vector rotation and the constants $C_1$ and $C_2$ describe the interaction between the layer deformation and the $\mathbf{t}$ vector rotation. In the following we take for simplicity $B_{13} \approx 0$, i.e. the variations of $\Phi$ along the $x$, $y$ and $z$ directions are not coupled. The equilibrium equations for the $S_C$ liquid crystal elasticity can be written as

$$
\left.
\begin{aligned}
& A_{12}\frac{\partial^4 u}{\partial x^4} + A_{11}\frac{\partial^4 u}{\partial x^2 \partial y^2} + A_{21}\frac{\partial^4 u}{\partial y^4} - \bar{B}\frac{\partial^2 u}{\partial z^2} \\
& \quad + C_1\frac{\partial^2}{\partial x\,\partial y}\left[\frac{\partial \Phi}{\partial x}\cos\Phi + \frac{\partial \Phi}{\partial y}\sin\Phi\right] \\
& \quad + C_2\frac{\partial^2}{\partial y^2}\left[\frac{\partial \Phi}{\partial y}\cos\Phi - \frac{\partial \Phi}{\partial x}\sin\Phi\right] = 0, \\
& B_1\left[\frac{\partial^2 \Phi}{\partial x^2} + \frac{\partial^2 \Phi}{\partial y^2}\right] + B_3\frac{\partial^2 \Phi}{\partial z^2} \\
& \quad + C_1\left[\frac{\partial^3 u}{\partial x^2 \partial y}\cos\Phi + \frac{\partial^3 u}{\partial x\,\partial y^2}\sin\Phi\right] \\
& \quad + C_2\left[\frac{\partial^3 u}{\partial y^3}\cos\Phi - \frac{\partial^3 u}{\partial y^2\,\partial x}\sin\Phi\right] = 0
\end{aligned}
\right\} \qquad (2)
$$

In the bulk sample there is no difference between the equilibrium equation for smectic C or smectic C* liquid crystals. The terms in the elastic free energy which express the chirality of the $S_C^*$ liquid crystal can be added to the surface anchoring energy [10, 11].

## 2. Experimental

The ferroelectric liquid crystals used in our experiments were the commercial chiral smectic mixtures CS-1011 (CHISSO Corp.) and ZLI 3654 (E. Merck AG). The sample cells of the surface stabilized ferroelectric liquid crystal type were prepared by sandwiching the ferroelectric liquid-crystalline materials between two glass substrates with transparent ITO electrodes on which the PVA aligning layer (MOWIOL 26-88, (Hoechst AG)) was deposited by the conventional spinning technique. The excess solvent was allowed to evaporate by heating the glass phates in an oven at 120°C for 15 min. The dry plates were immediately rubbed in a single direction with a rotating cylinder covered with a plush cloth. Cells with a gap of about 1·5 μm and with antiparallel rubbing directions on the two plates were investigated since in this configuration a significantly smaller number of defects arises than in cells with parallel rubbing. Vacuum deposited spacers on cell borders were used and the thickness of the empty cells was measured interferometrically. A good monodomain alignment with solitary zig-zag defects was obtained by cooling the cells slowly from the isotropic to the $S_C^*$ phase in the absence of an electric field.

Microscopic observations and electrooptical measurements were carried out before and after the application the square-wave electric field with a frequency from 10 to 50 Hz. A new texture resulted with the AC electric field [3]. Careful microscopic

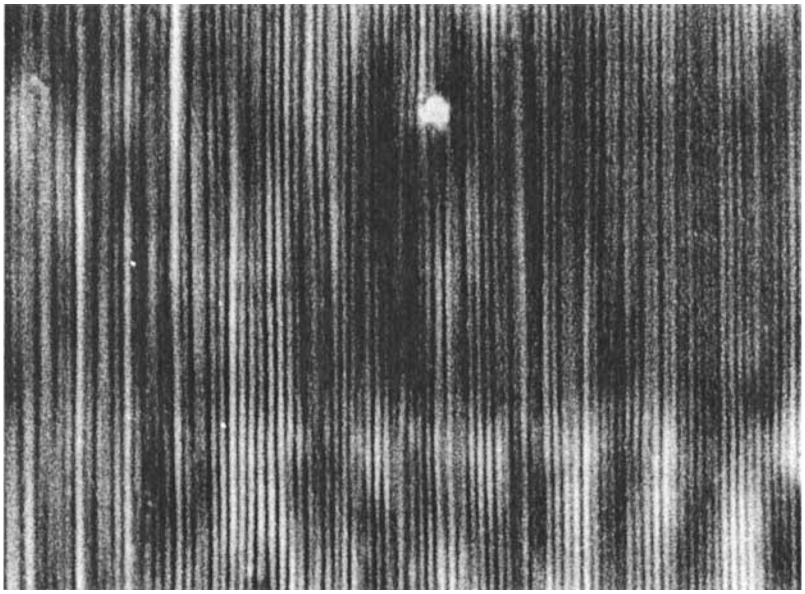

Figure 2. A micrograph showing the striped texture in the surface stabilized ferroelectric liquid crystal cell after low frequency AC electric field treatment (±25 V, 10 Hz); ferroelectric liquid crystal mixture ZLI 3654, cell gap 1·7 μm. The smectic layers are perpendicular to the stripes. Magnification × 550; the contrast was enhanced by the photoprocessing.

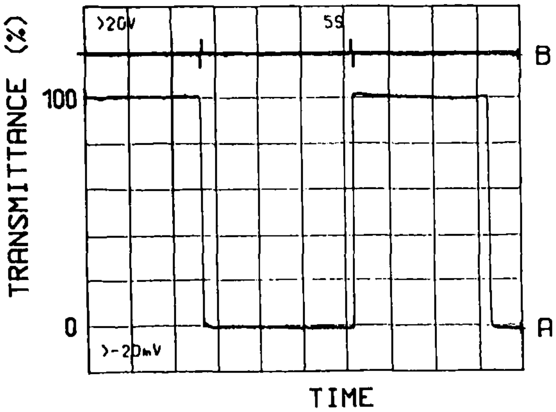

Figure 3. The optical response (trace A) of the surface stabilized ferroelectric liquid crystal cell to the bipolar pulses ±20 V (trace B); ferroelectric liquid crystal mixture ZLI 3654, cell gap 0·7 μm and a time base of 5 s per division.

observations have shown that the new texture, which begins to occur at voltage about ±15 V/μm, is striped with long and slender domains perpendicular to the smectic layers (see figure 2). The contrast between dark and light domains is very weak and the domains are best observed near extinction. The distance between dark or light stripes is about 2·4 μm for both of the mixtures investigated, i.e. approximately half of the pitch value.

The new texture is stable even after the driving voltage is switched off. The initial alignment can be restored only by heating into the isotropic state and on subsequent cooling. The electric field treated cells with the new texture have a perfect bistability (i.e. a memory effect) with high contrast as can be seen in figure 3, even if the switching times are slightly longer as compared with an untreated cell. A perfect optical extinction with almost black colour was obtained and no relaxation phenomenon of the transmittance was observed. Half of the apparent cone angle between the two extinction positions in the memory states was found to be nearly equal to that observed under a DC field, i.e. 23°, for both of the mixtures investigated. This value is also near the tilt angle at room temperature showed by the manufacturers of the mixture.

## 3. Anisotropy of $S_C$ layer and director deformation

Assume a $S_C$ layer deformation described by the displacement $u(x, z)$ which does not depend on the $y$ coordinate i.e. $\partial u/\partial y = 0$ and which satisfies

$$A_{12}\frac{\partial^4 u}{\partial x^4} = \bar{B}\frac{\partial^2 u}{\partial z^2}. \tag{3}$$

By using equations (2) and (3) the conditions for the corresponding $\Phi$ are

$$\left.\begin{aligned} &\frac{\partial}{\partial y}\left\{C_1\frac{\partial}{\partial x}\left[\frac{\partial\Phi}{\partial x}\cos\Phi + \frac{\partial\Phi}{\partial y}\sin\Phi\right]\right. \\ &\qquad\left. + C_2\frac{\partial}{\partial y}\left[\frac{\partial\Phi}{\partial y}\cos\Phi - \frac{\partial\Phi}{\partial x}\sin\Phi\right]\right\} = 0, \\ \text{and}\qquad & B_1\left(\frac{\partial^2\Phi}{\partial x^2} + \frac{\partial^2\Phi}{\partial y^2}\right) + B_3\frac{\partial^2\Phi}{\partial z^2} = 0. \end{aligned}\right\} \tag{4}$$

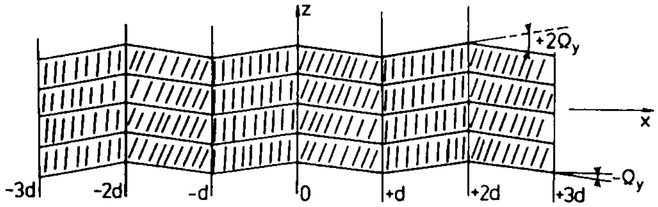

Figure 4. Schematic drawing of the simplest layer deformation of the $S_C^*$ sample after an electric field treatment (see also [6, 13]). The layers are periodically inclined by angles $(+\Omega_y)$ and $(-\Omega_y)$. The periodicity of the layer deformation is $2d$.

By taking $\partial\Phi/\partial y = 0$ the first condition of (4) is satisfied without conflict with the second one which then takes the form

$$B_1 \frac{\partial^2\Phi}{\partial x^2} + B_3 \frac{\partial^2\Phi}{\partial z^2} = 0. \tag{5}$$

The condition $\partial\Phi/\partial y = 0$ is the direct consequence of the anisotropic coupling between the layer distortion and **t** vector deformation.

Equations (1)–(4) describe an infinite $S_C$ liquid crystal. But for the interpretation of possible solutions the orientation of the surfaces with respect to the coordinate system in figure 1 is important. (1) When the sample surfaces are perpendicular to the $y$ axis the conditions $\partial u/\partial y = 0$ and $\partial\Phi/\partial y = 0$ express the fact that the solutions $u$ and $\Phi$ do not depend on the sample thickness. In particular the **t** vector twisted structure (see, for example, [11, 12]) is excluded. (2) When the sample surfaces are perpendicular to the $x$ axis the conditions $\partial u/\partial y = 0$ and $\partial\Phi/\partial y = 0$ only denote that the solutions $u$ and $\Phi$ do not change in the direction along the layers which are parallel to the surfaces. In the last case the **t** vector twisted structure along the sample thickness is permitted, as seen from equation (5). In thicker samples the helicoidal structure $\Phi = qz + \Phi_0$ is also permitted, where $\Phi_0$ is a constant and $q$ is connected with the helicoidal pitch $p$ of a $S_C^*$ liquid crystal via $q = 2\pi/p$.

In the special case of parallel undeformed layers, i.e. when $u = 0$, the orientation of the coordinate axes $x$ and $y$ with respect to the sample surfaces is not important because in our approximation the **t** vector orientation is isotropic in the $(x, y)$ plane. The twisted structure is then always possible for $u = 0$.

The layer deformation leading to the texture of long domains was modelled in [6] as a periodic system of curvature walls [9]. Here we consider the layer deformation similar to that described in [6] and approximated by a zig-zag structure (see figure 4) introduced in [13]. In [13] the textures of a $S_C^*$ liquid crystal in an alternating electric field were studied and a zig-zag layer structure occurred as the consequence of a flow induced by the high frequency electric field. The layer deformation in figure 4 can be described by the periodic displacement $u(x)$ defined by

$$\left.\begin{aligned} u(x) &= \Omega_y(x - 2dn + d/2) \quad \text{for} \quad x \in ((2n-1)d, 2dn), \\ u(x) &= \Omega_y(x - 2dn - d/2) \quad \text{for} \quad x \in (2dn, (2n+1)d), \end{aligned}\right\} \tag{6}$$

with $n = 0, \pm 1, \pm 2, \ldots$. The angle $\Omega_y$ which characterizes the inclination of the smectic layers from the $x$ axis is small in our approximation. The layer deformation defined by the displacement (6) is periodic with the periodicity $2d$ (see figure 4).

## 4. Connection of the layer deformation with the long domain texture and the memory effect

The real sample is finite and therefore the anchoring conditions become important. The surfaces are taken to be perpendicular to the $y$ axis. This choice of the $y$ axis orientation with respect to the surfaces is based on the observation [1] that the layers are nearly perpendicular to the surfaces after treatment by an electric field. Assume further that, during the texture creation in an electric field, the layers sliped on the glass plates as on the free surfaces. Therefore the final configuration of the layers given, for example, by the displacement in equation (6), corresponds to the boundary condition of a free surface, i.e. the projection of stress components to the normal to surfaces $\mathbf{N}$ is zero at the surfaces. The boundary condition, in tensor notation, gives

$$\sigma_{i3}N_i = 0,$$

where the summation over $i = 1, 2, 3$ is adopted. In our sample geometry with $\mathbf{N} = (0, \pm 1, 0)$ the condition of free surfaces requires $\sigma_{23} = 0$. The stress $\sigma_{23}$ is defined as

$$\sigma_{23} = -\frac{\partial}{\partial y}\left[A_{21}\frac{\partial^2 u}{\partial y^2} + \frac{A_{11}}{2}\frac{\partial^2 u}{\partial x^2}\right]$$

[14]. As $\partial u/\partial y = 0$ in the whole sample and even at the surfaces, the boundary condition $\sigma_{23} = 0$ is therefore satisfied.

Now let us discuss the influence of the surface anchoring on the $\mathbf{t}$ vector orientation in $S_C^*$. The anchoring energy $W_S$ (per unit surface) can be written as

$$W_S = W_M \sin^2\Phi \mp W_0 \cos\Phi, \tag{7}$$

where $W_M$ and $W_0$ are positive and the negative sign is valid for the upper plate and the positive sign for the lower plate [11, 15]. Due to the condition $\partial\Phi/\partial y = 0$ the angle $\Phi$ does not depend on the sample thickness. Then the sample surface energy (i.e. on both plates) is

$$2W_S = 2W_M \sin^2\Phi(x, z). \tag{8}$$

For the displacement $u(x, z)$ holding in equation (3) and for the angle $\Phi(x, z)$ which is a general solution of equation (4) the total elastic energy is obtained by a volume integration of relation (1) together with the surface integral over relation (8). That term corresponding to the interaction between fields $u(x, z)$ and $\Phi(x, z)$ which follows the general expression (1) in the form

$$\int_V \left\{C_1\frac{\partial^2 u}{\partial x \partial y}\left(\frac{\partial\Phi}{\partial x}\cos\Phi + \frac{\partial\Phi}{\partial y}\sin\Phi\right) + C_2\frac{\partial^2 u}{\partial y^2}\left(\frac{\partial\Phi}{\partial y}\cos\Phi - \frac{\partial\Phi}{\partial x}\sin\Phi\right)\right\} dV \tag{9}$$

is zero because $\partial u/\partial y = 0$. As we are interested in the $\mathbf{t}$ vector orientation we need study only that part, $E_\Phi$, of the total energy which depends on $\Phi(x, z)$

$$E_\Phi = \int_V \left[\frac{B_1}{2}\left(\frac{\partial\Phi}{\partial x}\right)^2 + \frac{B_3}{2}\left(\frac{\partial\Phi}{\partial z}\right)^2\right] dV + 2W_M \int_S \sin^2\Phi(x, z)\, dS. \tag{10}$$

energy $E_\Phi$ is positive for a general $\Phi$; then for $\Phi = 0, \pm\pi$ the energy $E_\Phi$ is minimal, i.e. $E_\Phi = 0$. Therefore the $\mathbf{t}$ vector distribution is stabilized at the uniform orientation described by $\Phi = 0$ or $\Phi = \pm\pi$. Such a stabilized $S_C^*$ structure can explain the observed memory effect [1–5] also reported in § 2 of this paper. The stabilized states $\Phi = 0$ and $\Phi = \pm\pi$ have antiparallel orientation of the $S_C^*$ spontaneous polarization. Then an applied external electric field can also switch one uniform state to the other uniform and stabilized one.

The texture shown in figure 2 shows the part of the sample between the central parts of the electrodes. In our model (see figure 4) there is a layer undulation between the electrodes. The layer undulation modulates slightly the uniform orientation of the **t** vector giving the modulation of contrast similar to the contrast of long domains shown in figure 2 described in § 2. As seen from the optical contrast in figure 2, the observed texture is probably more complex. Our model gives only an approximation to the observed texture.

## 5. Conclusion

The model of layer deformation can explain the memory effect described in [1–5] and in § 2 of this paper if the following conditions are fulfilled:

(1) The layer deformation of a $S_C^*$ liquid crystal is described by the displacement $u$ which obeys the equilibrium equation (3). The solution $u$ can be modelled, e.g. by the zig-zag layer deformation described in § 3 and shown in figure 4.

(2) In a thin $S_C^*$ sample between two glass plates where the layers are perpendicular to surfaces both $u$ and the **t** vector orientation $\Phi$ do not depend on the sample thickness and $\Phi$ has the same value at the upper and lower glass plates. Therefore the **t** vector twisted structure is excluded which is a consequence of the anisotropic coupling between the layer distortion and the **t** vector deformation. If the layers are not perpendicular to the sample surfaces or if they are curved then the displacement $u$ and the angle $\Phi$ have a general form depending also on the sample thickness and the memory effect discussed here is not present.

(3) The minimum of the total elastic energy $E_\Phi$ (see equation (10)) which determines $\Phi = 0, \pm\pi$ leads to the uniform stabilized orientation of the **t** vector. The fact that the uniform orientation of the **t** vector is stabilized is the essential of the memory effect.

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