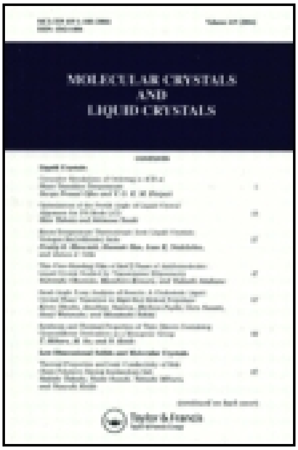

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

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Version of record first published: 24 Sep 2006

Mol. Cryst. Liq. Cryst., 1999, Vol. 328, pp. 383-390
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Electrically Deformed Short Pitch Chiral Smectic Liquid Crystals

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Short pitch chiral smectic C liquid crystals distorted by a transverse electric field are considered. Fully analytic and very simple expressions are given and discussed for the distorted structure and its optical properties, allowing to calculate the field induced rotation of the effective optic axis.

Keywords: optics of chiral media; distorted ferroelectrics; electrooptics

INTRODUCTION

Liquid Crystals (LC) exhibit a great variety of periodic helical phases. In particular, the cholesteric phase and the helical smectic phases (C*, I*, F* and K*), as well as the twist grain boundary (TGB) phases, show a periodic superstructure in only one direction, whereas the blue phases are periodic in three directions. Since usually the spatial periods are very large with respect to the molecular sizes, a description in terms of a continuous periodic medium is generally used and is good enough to any practical purpose[1]. Media having a pitch small with respect to the light wavelength can be treated as optically homogeneous. The true periodic structure and its homogeneous model are referred to as mesoscopic and macroscopic models, respectively. Two different methods have already been developed to find homogeneous models; they are used to describe deformed cholesterics[2] and chiral smectic C's[3] (SmC*). For more complex structures a new approach is more convenient[4], based on a Fourier expansion of the dielectric tensor. In this paper, we consider chiral smectics distorted by an electric field. The interaction between the field and the molecular dipoles leads to a deformation of the helix[5,6,7,8]. If the field is perpendicular to the helix axis, the regions in which the polarization is favorably oriented grow and the system will exhibit a net average polarization in the direction of the applied electric field. Above a critical value (E_C), the helix will unwind, giving a uniform polarization. Below the threshold, the most important optical effect is a rotation of the macroscopic optic axis in the plane defined by the helix direction and the electric field. This electro-optic effect, which is generally called the deformed helical ferroelectric (DHF) effect, provides fast switching speeds at low voltages and grey scale capabilities. Not surprisingly, it has been suggested for use in applications[9] and even been developed for use in active matrix displays[10]. Numerical calculations are generally required to find out the direction of the macroscopic optic axis and the corresponding optical properties of distorted samples. Here we show that, for small enough applied fields, the distortion and the optical properties of short pitch media can be described by fully analytic and very simple expressions.

THE STRUCTURE OF THE DEFORMED HELIX

The undistorted smectic structure is well described by a mesoscopic model, in which the local director is uniformly rotating along the helix axis z:

$$\hat{n} = \sin\theta \cos\varphi \ \hat{x} + \sin\theta \sin\varphi \ \hat{y} + \cos\theta \ \hat{z} \ . \tag{1}$$

Here θ is the tilt angle and $\varphi = 2\pi z / p_o$, where p_o is the pitch of the unperturbed helix. The electric field strongly changes the z-dependence of the azimuthal angle φ, leaving the tilt angle practically unchanged. If the quadratic dielectric coupling and the contributions from the flexoelectric effect are neglected, the free energy density of the smectic can be written as[5]

$$f = \frac{K_3\theta^2}{2}\left(\frac{\partial\varphi}{\partial z} - \frac{2\pi}{p_o}\right)^2 + \vec{E}\cdot\vec{P}, \tag{2}$$

where K_3 is an elastic constant. If the electric field is in the y-direction, $\vec{E} = E\ \hat{y}$, the coupling term with the polarization[11] $\vec{P} = 1/2P_o \sin(2\theta)(\sin\varphi, -\cos\varphi, 0)$ can, to first order in θ, be written as $\vec{E}\cdot\vec{P} = P_o\theta E\cos\varphi$. This yields the Euler-Lagrange equation

$$K_3\theta^2\frac{\partial^2\varphi}{\partial z^2} + P_o\theta E \sin\varphi = 0, \tag{3}$$

whose solution $\varphi(z)$, neglecting possible boundary effects, can be written implicitly as:

$$\frac{z}{p} = \frac{F_1(\varphi/2,k)}{F_1(\pi,k)}, \qquad k = \frac{p_o F_2(\pi,k)}{\pi^2}\left(\frac{P_o E}{K_3\theta}\right)^{1/2} \tag{4}$$

where

$$F_1(t,k)=\int_0^t \frac{dt'}{\sqrt{1-k^2 sin^2 t'}}, \quad F_2(t,k)=\int_0^t \sqrt{1-k^2 sin^2 t'}dt', \qquad (5)$$

are elliptic integrals of the first and second kind respectively. The critical field E_C corresponds to the value $k = 1$ where F_1 diverges.
For small electric fields, the solution of Eq.(3) can be approximated by the simple expression:

$$\varphi = qz + Asin(qz),$$
$$q=\frac{q_o}{1+\frac{1}{2}A^2}, \qquad A=\frac{P_o E}{K_3\theta}\frac{4p_o^2}{\pi^4}. \qquad (6)$$

In Fig. 1 this approximation is compared with the exact solution. The agreement between them is very satisfactory till A=0.62 (the two curves are practically coincident). Even for $A = 1$ the differences are very small.

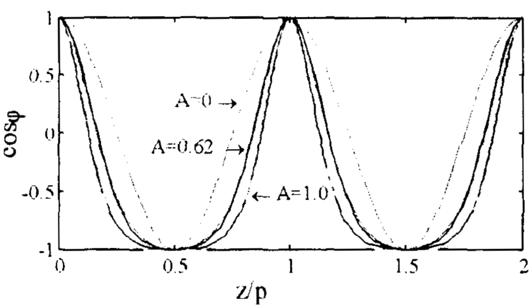

Figure 1 Cosine of the azimuthal angle φ of the helix as a function of the normalized z-coordinate. The solid dark lines are the exact solution while the grey lines are the sinusoidal approximation (6). Increasing values of A give larger induced deformations (for A=0, E =0; for A=0.62, E=0.78 Ec; for A=1.0, E=0.95 Ec).

THE MACROSCOPIC MODEL

The optical properties mesoscopic model for locally uniaxial media are defined by the constitutive equations $\vec{D}=\varepsilon_o \underline{\underline{\varepsilon}}\vec{E}$, $\vec{B}=\mu_o\vec{H}$, where

$$\underline{\underline{\varepsilon}} = \varepsilon_o + \varepsilon_a \hat{n}\cdot\hat{n}. \qquad (7)$$

Here $\varepsilon_a = \varepsilon_e - \varepsilon_o$ is the dielectric anisotropy.

For a plane wave incident on a slab sandwiched between planes orthogonal to z, the internal field is defined by the Berreman vector $\beta(z) = (E_x, H_y, E_y, -H_x)^t$, where t stands for transpose. The propagator $U(z)$, defined by $\beta(z) = U(z)\beta(0^+)$, satisfies the equation:

$$\frac{dU(z)}{dz} = i\frac{2\pi}{\lambda}B(z)U(z), \tag{8}$$

where λ is the vacuum wavelength and the kernel $B(z)$ is a 4x4 matrix, here referred to as the Berreman matrix[12]. The transfer matrix $U(d)$ of a given sample is obtained by integrating Eq.(8) over the sample thickness d. The method used in Ref.[3] to find the macroscopic model requires one to search for the z-independent matrix $\tilde{B}$ and for the corresponding set of constitutive equations, which give the best approximations for $U(d)$, for any given direction of the incident beam. This procedure is rather involved, but it gives also the limits of validity of the macroscopic model and allows us to find possible boundary effects. For periodic media, the optical properties of a sample whose thickness is an integer multiple of the period are fully defined by the transfer matrix over one period. For our samples ($p << \lambda$), this matrix is conveniently written as[4]

$$\begin{aligned} U(\varphi_o) &= \mathbf{1} + \frac{p}{\lambda}\int_{\varphi_o}^{\varphi_o+2\pi} d\varphi' B(\varphi')U(\varphi') = \mathbf{1} + \sum_{n=1}^{\infty}\left(\frac{ip}{\lambda}\right)^n u^{(n)}, \\ u^{(n)} &= \int_{\varphi_o}^{\varphi_o+2\pi} d\varphi_1 B(\varphi_1)\int_{\varphi_o}^{\varphi_1} d\varphi_2 B(\varphi_2) \ldots\ldots \int_{\varphi_o}^{\varphi_{n-1}} d\varphi_n B(\varphi_n), \end{aligned} \tag{9}$$

and explicitly depends on φ_o which is the azimuthal angle at the boundaries ($\mathbf{1}$ is the 4x4 identity matrix).

We expand the Berreman matrices $B[\varphi(z)]$ and $\tilde{B}$ for the mesoscopic and macroscopic models as follows:

$$B(\varphi) = B_o + \sum_{m=1}^{\infty} a_m \cos(m\varphi) + b_m sin(m\varphi), \tag{10}$$

$$\tilde{B} = \tilde{B}_o + (ip/\lambda)\tilde{B}_1 + (ip/\lambda)^2\tilde{B}_2 + \ldots \tag{11}$$

Up to terms in p/λ, the two Berreman matrices give the same transfer matrix if and only if:

$$\tilde{B}_o = B_o,$$
$$\tilde{B}_1 = \frac{1}{2\pi}\sum_{n=1}^{\infty}\frac{1}{n}\;\frac{1}{2}(b_n a_n - a_n b_n) + \cos(n\varphi_o)(B_o b_n - b_n B_o) \qquad (12)$$
$$-\sin(n\varphi_o)(B_o a_n - a_n B_o)$$

The elements of the matrix $\tilde{B}_1$ are very small with respect to the elements of $\tilde{B}_o$ and the higher order matrices $\tilde{B}_2$, ... are negligible to any practical purpose. Despite this fact, the matrix $\tilde{B}_1$ could give important macroscopic effect, since it is related to the optical activity of the medium, as discussed below. The most important feature of this matrix is its dependence on the angle φ_o, which defines the direction of the local optic axis of the periodic sample at its boundary planes. To define the macroscopic model, the φ_o dependent terms must be neglected, as discussed in Ref.[13].

For distorted smectics, the matrix $B(\varphi)$ contains terms of the type $\cos(A\sin qz)$ and $\sin(A\sin qz)$. For small distortions, we have used the following approximations:

$$\cos(A\sin qz) = J_o(A) + 2J_2(A)\cos 2qz$$
$$\sin(A\sin qz) = 2J_1(A)\sin qz \qquad (13)$$

and expanded the Bessel functions J_i in a power series of A up to second order.

RESULTS AND DISCUSSION

The macroscopic model is defined in terms of the constitutive equations first suggested by Tellegen[14]: $\vec{D} = \varepsilon_o \underline{\underline{\tilde{\varepsilon}}}\vec{E} + i\underline{\underline{\tilde{\kappa}}}\vec{H}$; $\vec{B} = \mu_o\vec{H} - i\underline{\underline{\tilde{\kappa}}}'\vec{E}$. The matrix $\tilde{B}_o$ defines the dielectric tensor $\underline{\underline{\tilde{\varepsilon}}}$, whereas $\tilde{B}_1$ defines the chirality tensor $\underline{\underline{\tilde{\kappa}}}$. Within the approximations presented at the end of the preceding section, the only non-zero elements of these tensors are:

$$\tilde{\varepsilon}_{zz} = \varepsilon_o + \varepsilon_a\cos^2\theta,$$
$$\tilde{\varepsilon}_{yy} = \varepsilon_o(1 + \varepsilon_e/\tilde{\varepsilon}_{zz}) + (A/2)^2(\varepsilon_a^2\sin^2\theta)/(4\tilde{\varepsilon}_{zz}),$$
$$\tilde{\varepsilon}_{xx} = \varepsilon_o(1 + \varepsilon_e/\tilde{\varepsilon}_{zz}) + (A/2)^2(5/4\varepsilon_o + \varepsilon_a\sin^2\theta), \qquad (14)$$
$$\tilde{\varepsilon}_{xz} = \tilde{\varepsilon}_{zx} = -A/2\varepsilon_a\sin 2\theta;$$

$$\tilde{\kappa}_{zz} = \frac{p\varepsilon_a^2\sin^2 2\theta}{4\lambda\tilde{\varepsilon}_{zz}}\left(1 - \frac{3}{2}\left(\frac{A}{2}\right)^2\right). \qquad (15)$$

Let us now discuss our results. The macroscopic model has been derived by assuming $p \ll \lambda$ and $A \ll 1$. Concerning the pitch p, the range of validity of the model is the same as for the undistorted SmC*, which has already been exhaustively discussed in Ref.[13]. For any given optical geometry, the model gives generally good results for p values smaller than half internal wavelength.

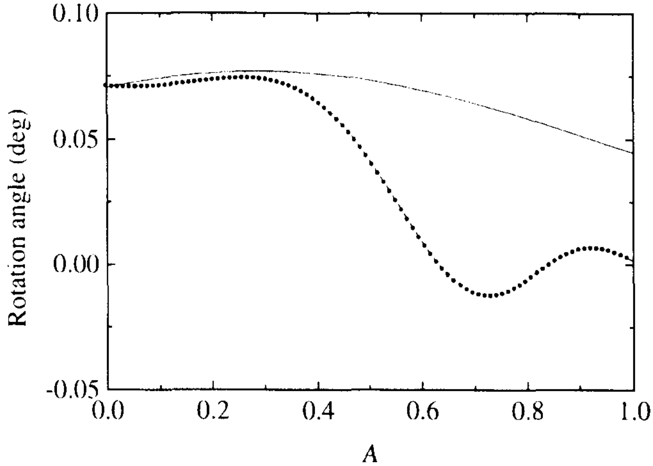

Figure 2 Rotation angle of the output polarization as a function of A for a linearly polarized incident wave with an incident angle in the glass $\theta_g = 10^{\circ}$ on a sample with pitch $p = \lambda/10$, thickness $d = 80p$, tilt angle $\theta = 25^{\circ}$, extraordinary and ordinary refractive indices $n_e = 1.55, n_o = 1.5$, respectively, and glass refractive index $n_g = 1.5$. The solid line stands for the mesoscopic model and the dotted line for the macroscopic one.

For the A- dependence we observe the following:

1. The range of validity of the model is restricted to the interval $0<A<0.3$, as shown in Fig. 2.
2. The macroscopic medium is biaxial. However, in its range of validity the biaxiality is very small and the optic axes are nearly coincident. Practically, the most important optical effect induced by the field is therefore a simple rotation of the effective optic axis, as generally assumed in the literature. For instance, for $A= 0.3$ and with the parameters defined in Fig.2 the rotation angle is equal to 12°.
3. The medium displays optical activity, a fact that in principle could give important macroscopic effects along the optic axis, whose direction depends on A. The structure of the chirality tensor is such that the optical rotation is zero for light propagating along the z-axis and maximum along the orthogonal directions. For $A = 0$ (undeformed chiral smectics), the

macroscopic optic axis is parallel to z and the optical rotation generally gives negligible effects. For the deformed medium, the optical rotation for light propagating along the new optic axis could in principle be considerably higher, but within the considered A-range the effect is too small to have practical consequences (Fig. 3).

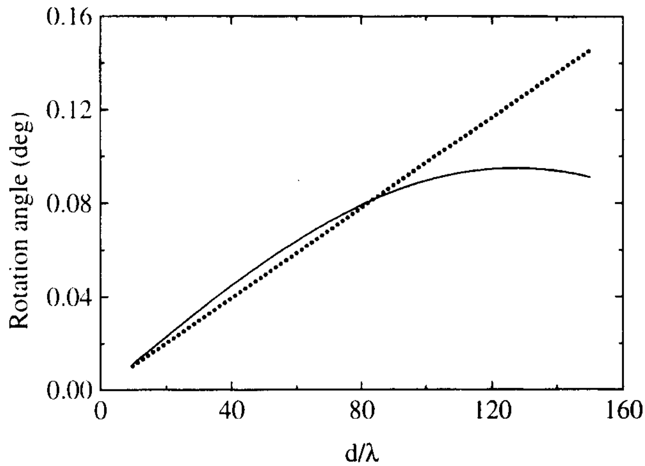

Figure 3 Rotation angle of the output polarization as a function of the normalized sample thickness for incident angle $\vartheta_c = 25°$, equal to the angle of the optic axis for $A = 0.3$, and the same parameters as in Fig. 2.

CONCLUSIONS

The structure of the deformed short pitch SmC* liquid crystal can be well approximated with a simple mathematical expression. Based on this approximation, an homogeneous model has been presented and its limits of validity verified for $A < 0.3$ ($E/E_C < 0.5$) and for p smaller than half the internal wavelength. The medium presents an optical biaxiality and a true optical activity, but this effect seems to be too small to have relevant consequences for applications. Practically, the main effect of the electric field is to rotate the macroscopic optic axis. Explicit expressions for the macroscopic dielectric tensor are given, allowing to calculate the rotation angle as a function of the electric field.

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