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Reorientation of the Director and the Bonds in a Hexatic Smectic Phase

P. Schiller (*), K. Camara, G. Pelzl, S. Diele and F. Zeidler

Martin-Luther-Universität Halle-Wittenberg, Institut für Physikalische Chemie, Mühlpforte 1,
06108 Halle/S., Germany

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Abstract. — Some smectic liquid crystal phases exhibit a long range bond-orientational order. In hexatic smectic phases with molecules tilted towards the layers director rotations are combined with distortions of the bond net. The coupling of the director to the bond field influences strongly the threshold behaviour of the Freedericksz transition of planar oriented smectic I films sandwiched between parallel plates. In contrast to nematic and smectic C films, the threshold voltage depends on the film thickness. Investigating this thickness dependence several material parameters can be obtained by adjusting the experimental data to the threshold formula.

1. Introduction

The broken translational symmetry in a crystal necessarily implies long-range orientational order. A nematic liquid crystal does not exhibit a translational order, but there is an anisotropy due to the orientation of non-spherical molecules. Another possibility to break the rotational symmetry results from the bond-orientational order. This ordering of hexatic liquids is associated with the orientation of the lines joining neighbouring atoms or molecules [1]. Hexatic order has proven to exist in both two-dimensional and three-dimensional systems, for example in free standing liquid crystal films, colloidal systems, in magnetic bubble arrays and Langmuir-Blodgett films.

In some three-dimensional smectic liquid crystals the parallel alignment of the molecular long axes is combined with a long range hexatic ordering of the centres of gravity [1–3]. The molecules of a smectic I phase form a stack of layers in which the molecular long axes are tilted with respect to the layer normal. The preferred direction of the molecules is characterized by a coordinate axis w , which is parallel to a unit vector called the director. More precisely, the axis w is chosen to be parallel to one of the major axes of the dielectric tensor of the biaxial structure. The second major axis is perpendicular to the tilt plane of the molecules, *i.e.* perpendicular to the axes w and κ defined in Figure 1. This direction coincides with the twofold rotation axis of the tilted smectic structure. Finally, the third major axis v is defined to be perpendicular to the former ones. The corresponding principal values of the dielectric tensor are denoted by ϵ_{ww} , ϵ_{uu} and ϵ_{vv} , respectively.

(*) Author for correspondence (e-mail: schiller@indigo2.chemie.uni-halle.de)

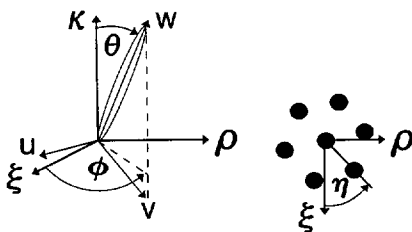


Fig. 1. — The angles ϕ and η define rotations of the long molecular axes and the bonds, respectively. A Cartesian system with the axes ξ , ρ and κ is attached to the smectic layers, while u , v and w are the major axes of the dielectric tensor.

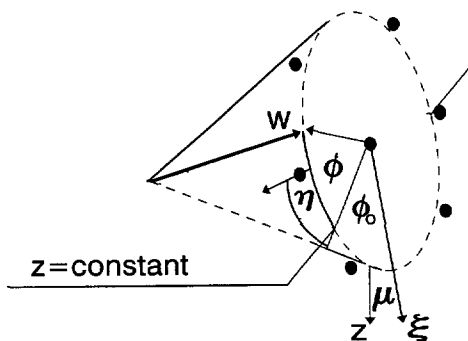


Fig. 2. — Reorientation in a planar film of the smectic I phase. The axis z is perpendicular to the film interface, whereas the axis ξ lies on the smectic plane. Deviations from the bookshelf geometry (with layers perpendicular to substrates) are characterized by a non-zero layer tilt angle μ . (In comparison to Fig. 1 the reference axis for the angles η and ϕ is chosen differently.)

In the hexatic smectic I phase the local hexagon formed by the six nearest neighbours of a molecule is oriented macroscopically in an undisturbed sample and the director is tilted towards an apex of the hexagon. Elastic distortions are usually accompanied with both director and bond reorientations. The major axis w and the normal of the smectic layers enclose a fixed angle θ . Therefore only azimuthal rotations defined by the angle ϕ must be taken into account. Rotations of the local hexagon are defined by the angle η .

In this paper we investigate the reorientation of the director and the bonds in a smectic I film driven by an electric field. The elastic continuum theory predicts that the threshold voltage of the Fredericksz transition of the smectic I phase should depend on the film thickness [4]. This behaviour of the threshold provides some information on the bond-orientational order. Especially, it is possible to determine the elastic constant for bond distortions and a material parameter which characterizes the coupling between the director and the bond field.

2. Free Energy and Boundary Conditions

We consider a smectic I film with director orientation parallel to the boundaries. Figure 2 depicts the orientation of the hexagon and the director with respect to the plane $z = \text{constant}$ which is parallel to the bounding plates. The layers could be perpendicular to the bounding plates (bookshelf geometry) or tilted by an angle μ . Below the Fredericksz-threshold the

director is homogeneously aligned parallel to the plates. Assuming a strong anchoring the alignment of the molecules remains unchanged at the bounding plates, even if the director field is remarkably distorted in the inner part of the sample. It is also reasonable to suppose that the bond directions are fixed to the boundaries. Otherwise, if the direction of the long molecular axes (director) is strongly anchored, a rotating bond-net would change the number of particles in contact with the surface. Probably, weak elastic torques are not sufficient to alter the surface density and thus we can assume that a strong anchoring of the director implies fixed bonds at the surfaces. Then the azimuthal director angle ϕ and the bond angle η , which characterize deviations from the perfect planar alignment, are always zero at the boundaries $z = -d/2$ and $z = d/2$. For describing small director and bond rotations in hexatic smectic phases with tilted molecules Pleiner and Brand [5] proposed a general elastic continuum theory, while a simplified model introduced by Nelson, Halperin and Selinger [2,3] is also applicable if the distortions are rather large. In this paper we perform a linear stability analysis using a simple mathematical model for describing the Freedericksz effect. Let the small parameter $\epsilon \propto \phi, \eta$ characterize the magnitude of weak distortions ($\epsilon < \pi/6$). For small deviations from the planar alignment the elastic energy can be expanded in the series

$$f_1 = \frac{1}{2}K_1[1 + c_1\phi + c_6\eta + \dots](\partial_z\phi)^2 + \frac{1}{2}K_6[1 + d_1\phi + d_6\eta + \dots](\partial_z\eta)^2 + K_{16}[1 + g_1\phi + g_6\eta + \dots]\partial_z\phi\partial_z\eta$$

where the coefficients K_i , c_i , d_i and g_i depend on the layer tilt angle μ (Fig. 2). In the framework of a linear stability analysis, it is sufficient to consider only lowest order terms proportional to ϵ^2 :

$$f_1 = \frac{1}{2}K_1(\partial_z\phi)^2 + \frac{1}{2}K_6(\partial_z\eta)^2 + K_{16}\partial_z\phi\partial_z\eta + O(\epsilon^3).$$

Stability of the homogeneous film state requires $K_1 > 0$, $K_6 > 0$ and $K_1K_6 - K_{16}^2 > 0$. The coupling between the director and the bond orientation could be taken into account by a series

$$f_H = - \sum_{n=1}^{\infty} h_{2n} \cos 2n(\phi - \eta),$$

which is a generalization of the expression $f_H = h \cos 6(\phi - \eta)$ used in reference [2]. In the case of small rotation angles we can write

$$f_H = \text{const.} + \frac{1}{2}H(\phi - \eta)^2 + O(\epsilon^4)$$

where the coupling constant H is positive for a smectic I phase. Finally, the free energy density of the electric field is defined by

$$f_2 = -\frac{1}{2}\Sigma\epsilon_{ij}E_iE_j.$$

Since the electric field \mathbf{E} is parallel to the z -axis, the corresponding free electric energy density is expressed as

$$f_2 = -\frac{1}{2}(\epsilon_{uu}u_z^2 + \epsilon_{vv}v_z^2 + \epsilon_{ww}w_z^2)E^2,$$

where u_z , v_z and w_z are the z -components of the unit vectors \mathbf{u} , \mathbf{v} and \mathbf{w} which are parallel to the coordinate axes u , v and w , respectively. If a uniaxial behaviour with $\epsilon_{uu} = \epsilon_{vv} < \epsilon_{ww}$ is assumed, the free energy density of the electric field can be written as [4]

$$f_2 = -\frac{1}{2}(\epsilon_{ww} - \epsilon_{uu})[\cos\theta\sin\mu - \sin\theta\cos\mu\cos(\phi + \phi_0)]^2E^2 + \text{const.}$$

where ϕ_0 is determined by the relation $\cos\phi_0 = \tan\mu/\tan\theta$. As the mathematical expression is rather complicated in the general biaxial case, we only consider two borderline cases, namely the bookshelf geometry $\mu = 0$ and the geometry with maximum layer tilt $|\mu| = \theta$. Then the effective dielectric anisotropy

$$\Delta\epsilon_{\text{eff}} = \begin{cases} (\epsilon_{ww} - \epsilon_{vv})\sin^2\theta \cos^2\mu \sin^2\phi_0 & |\mu| \leq \theta \quad (\text{uniaxial}) \\ \epsilon_{vv} - \epsilon_{uu} + (\epsilon_{ww} - \epsilon_{vv})\sin^2\theta & \mu = 0 \quad (\text{bookshelf}) \\ (\epsilon_{uu} - \epsilon_{vv})\cos^2\theta & |\mu| = \theta \end{cases} \quad (1)$$

enters in the expansion of the free electric energy density $f_2 = \text{const.} - \frac{1}{2}\Delta\epsilon_{\text{eff}}\phi^2 + O(\epsilon^3)$. Considering only terms with lowest order of magnitude, the free energy of the planar smectic I film is expressed as

$$F = \frac{1}{2} \int_{-d/2}^{+d/2} dz [K_1(\partial_z\phi)^2 + K_6(\partial_z\eta)^2 + 2K_{16}\partial_z\phi\partial_z\eta + H(\phi - \eta)^2 - \Delta\epsilon_{\text{eff}}E^2\phi^2] + O(\epsilon^3). \quad (2)$$

If deviations from the equilibrium state occur, the dynamics of the director and the bond angle is described by the phenomenological equations

$$-\Gamma_1 \frac{\partial\phi}{\partial t} = \frac{\delta F}{\delta\phi} \quad \text{and} \quad -\Gamma_2 \frac{\partial\eta}{\partial t} = \frac{\delta F}{\delta\eta}, \quad (3)$$

where Γ_1 and Γ_2 are rotational viscosities and t is the time. In these equations any material flow accompanied with the reorientation process of the director and the bonds is neglected. It should be noted that the Freedericksz threshold voltage U_c could be also obtained from the (static) Eulerian equations which are related to the free energy (2). For $U < U_c$ the free energy (2) is a minimum if $\phi = 0$ and $\eta = 0$, whereas this minimum disappears when U exceeds the critical value U_c . Since elastic distortions appear above the threshold, the electric field is not longer homogeneous in the sample. But this small deviation $\Delta E \propto A^2$ ($A = \phi(z=0)$, distortion amplitude) from the applied field $E = U/d$ causes only corrections of higher order of magnitude which do not influence the threshold value E_c of the Freedericksz transition [6].

Strong anchoring at the substrates leads to the conditions

$$\phi(z = -d/2) = \phi(z = +d/2) = 0 \quad \text{and} \quad \eta(z = -d/2) = \eta(z = +d/2) = 0. \quad (4)$$

The Freedericksz threshold voltage of a planar nematic film does not depend on the film thickness [7]. Smectic I liquid crystals, however, which exhibit a bond-orientational order, should behave differently. Due to the coupling of the director to the bonds a characteristic dependence of the threshold voltage on the layer thickness is expected. Theoretical and experimental investigations suggest that a weak hexatic order should exist even in the smectic C phase [1]. In this case the results obtained below also provide an extension of the theory for the Freedericksz transition in planar films of the smectic C phase [8].

3. Threshold of the Freedericksz Transition

Equations (3) are written as

$$\begin{aligned} \Gamma_1 \partial_t \phi &= K_1 \partial_z^2 \phi + K_{16} \partial_z^2 \eta + \Delta\epsilon_{\text{eff}} E^2 \phi - H(\phi - \eta) \quad \text{and} \\ \Gamma_2 \partial_t \eta &= K_{16} \partial_z^2 \phi + K_6 \partial_z^2 \eta + H(\phi - \eta). \end{aligned} \quad (5)$$

These equations are solved by using the ansatz

$$\begin{pmatrix} \phi \\ \eta \end{pmatrix} = \begin{pmatrix} \phi_0 \\ \eta_0 \end{pmatrix} \exp(ikz + \lambda t) \quad (6)$$

where $i = \sqrt{-1}$. Applying the condition of neutral stability $\max[\lambda(k)] = 0$ and taking into account the boundary conditions (4) leads to $k = \pi/d$ and the critical value [4]

$$\Delta\epsilon_{\text{eff}} E_c^2 = H + K_1 k^2 - \frac{(H - K_{16} k^2)^2}{H + K_6 k^2} \quad (7)$$

above that the initial configuration $\phi = \eta = 0$ becomes unstable. Taking into account the relation $U = Ed$ between the voltage and the electric field strength, the square of the threshold voltage is expressed as

$$U_c^2 = U_0^2 \left\{ 1 + (d/d_0)^2 - \frac{[(K_{16}/K_1)^2 - (d/d_0)^2]^2}{K_6/K_1 + (d/d_0)^2} \right\} \quad (8)$$

where

$$U_0 = \sqrt{\frac{\pi^2 K_1}{\Delta\epsilon_{\text{eff}}}} \quad \text{and} \quad d_0 = \sqrt{\frac{\pi^2 K_1}{H}}. \quad (9)$$

The unstable mode, which is growing at a voltage slightly above the Freedericksz-threshold, is found to be

$$\begin{pmatrix} \phi(x) \\ \eta(x) \end{pmatrix} = A_1 \begin{pmatrix} 1 \\ R(k) \end{pmatrix} \cos kz,$$

where $R(k) = \frac{H - K_{16} k^2}{H + K_6 k^2}$ and the amplitude A_1 could be obtained by taking into account additional nonlinear terms [4] which enter in the equations (5).

Some insight in the temperature dependent behaviour of the elastic constants K_i and the coupling parameter H provides the Landau-theory of the SmC-SmI phase transition. The complex order parameter $\xi = \theta \exp(i\phi)$ is useful to characterize the tilt order in smectic liquid crystals [8], while the order parameter $\beta = |\beta| \exp(6i\eta)$ describes the strength of the bond-orientational order and the preferred direction of the bonds [1]. Then the free energy density is expressed by an expansion [10]

$$\begin{aligned} \hat{F} = & a_1 |\xi|^2 + b_1 |\xi|^4 + \dots + a_6 |\beta|^2 + b_6 |\beta|^4 + \dots \\ & + \frac{1}{2} K_1^0 |\nabla \xi|^2 + \frac{1}{2} K_6^0 |\nabla \beta|^2 + \frac{1}{2} h [\xi^6 \beta^* + (\xi^*)^6 \beta] + \dots \end{aligned} \quad (10)$$

If the tilt angle is not equal to zero ($|\xi| = \theta \neq 0$), the coupling term proportional to h produces always a bond-orientational order accompanied with $|\beta| \neq 0$. In the smectic C phase ($a_6 > 0$) this order is expected to be very weak, whereas the ordering of the bonds is much more pronounced in the smectic I phase ($a_6 < 0$). If elastic distortions occur the tilt angle θ remains constant ($\nabla \theta = 0$) and the relations $\frac{1}{2} h [\xi^6 \beta^* + (\xi^*)^6 \beta] = h |\beta| \theta^6 \cos 6(\phi - \eta)$, $\frac{1}{2} K_1^0 |\nabla \xi|^2 = \frac{1}{2} K_1^0 \theta^2 (\nabla \phi)^2$ and $\frac{1}{2} K_6^0 |\nabla \beta|^2 = \frac{1}{2} K_6^0 |\beta|^2 (\nabla \eta)^2$ demonstrate how the material constants depend on the order parameters. Comparing the free energy expressions (2) and (10), we obtain $H = 18h |\beta| \theta^6$, $K_1 = K_1^0 \theta^2$ and $K_6 = K_6^0 |\beta|^2$.

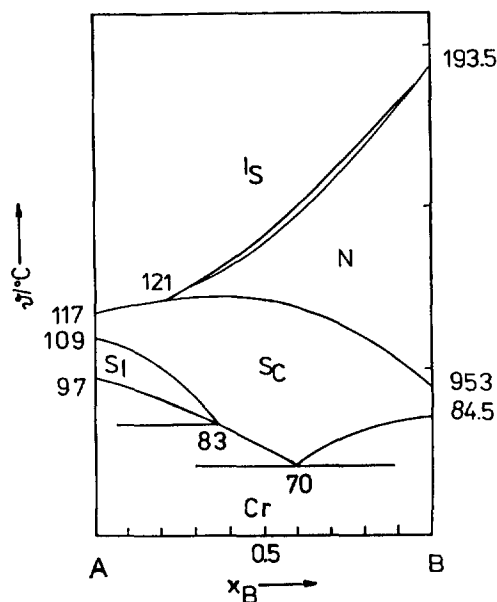
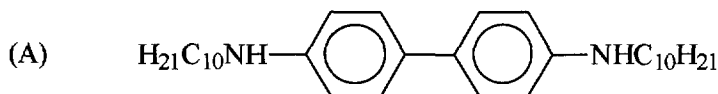


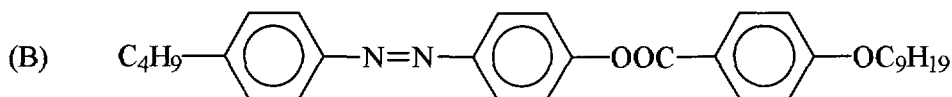
Fig. 3. — The phase diagram of the binary mixture.

4. Substances and Experimental Setup

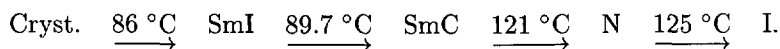
Experimental investigations were performed on planar oriented smectic C and smectic I films using a mixture which is in the nematic state at higher temperatures. The planar alignment, which is characterized by a director orientation parallel to the substrate planes, was achieved by slowly cooling the planar oriented nematic phase. Homologous 4,4'-bis (n-alkylamino)-biphenyls with intermediate length of the alkylchains exhibit both the SmC and the SmI phase in a convenient temperature range [11]. The requirement that the phase sequence should also include the nematic state satisfies the binary mixture of the compounds 4,4'-bis (n-decylamino)-biphenyl



and 4-n-butylphenylazophenyl-4-n-nonyloxybenzoate [12]



in a certain concentration range. The phase diagram of the binary system is presented in the Figure 3. For the electrooptical investigations we used a mixture with mole fraction $x_A \approx 0.7$, which has the phase sequence:



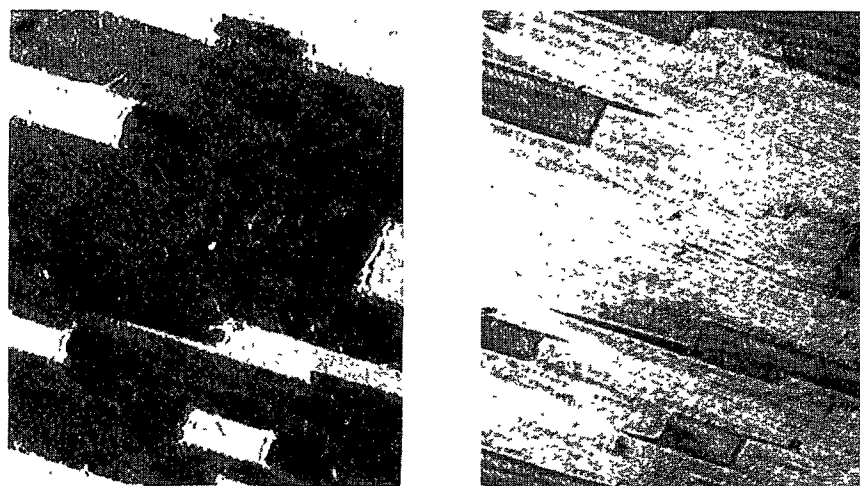


Fig. 4. — Fredericksz transition of the planar oriented smectic I film (temperature $\vartheta = 88^\circ\text{C}$, film thickness $6\ \mu\text{m}$, crossed polarizer). Voltage: $U = 0\ \text{V}$ (left) and $U = 35\ \text{V}$ (right).

The nematic mixture was filled into a commercial electrooptical sandwich cell (E.H.C. Co. Tokyo). The inner transparent conductive surfaces are coated with a prerubbed polyimide layer so that a planar alignment results. On decreasing the temperature two types of homogeneous domains appeared in the SmC phase. The director orientation in the domains deviated slightly from the alignment in the nematic phase. This orientation remained unchanged in the smectic I phase (Fig. 4) at lower temperatures. Detailed optical investigations suggest that the smectic layers are arranged in the chevron geometry [13, 14], which is also indicated by some zig-zag defects.

The electrooptical cell was placed on the hot stage (Linkam) of a polarizing microscope (Leitz Orthoplan) equipped with a photo tube and a photomultiplier (1P 28 PR 1402 RF, Enatechnik) to measure the light intensity. The Fredericksz effect was observed by applying an a.c. electric field and the threshold voltages were obtained from the light intensity-voltage curves. The frequency of the electric field was chosen to be high enough (1 kHz) to avoid flexoelectric distortions and convective instabilities. Electrically driven convective instabilities, which occur in the smectic C phase of less purified substances, are visible by striped textures (similar to the textures of the well-known convective instabilities of nematic layers). The instability described in this paper is not accompanied with stripes and material flow, which would be detectable by the motion of dust particles. Since the material is non-chiral, also the ferroelectric switching mechanism can be excluded.

5. Experimental Results and Discussion

In the phases SmC and SmI the director rotates around the layer normal if the voltage exceeds the Fredericksz-threshold. This transition is indicated by a uniform change of the interference colour (Fig. 4). We found that neighbouring domains are distinguished by the sense of the director rotation around the layer normal. The threshold voltage amounts approximately 6 V for the nematic phase (122°C), whereas for the smectic I phase its value depends on the sample thickness. Even for the smectic C phase (92°C) we found a weak alteration of the

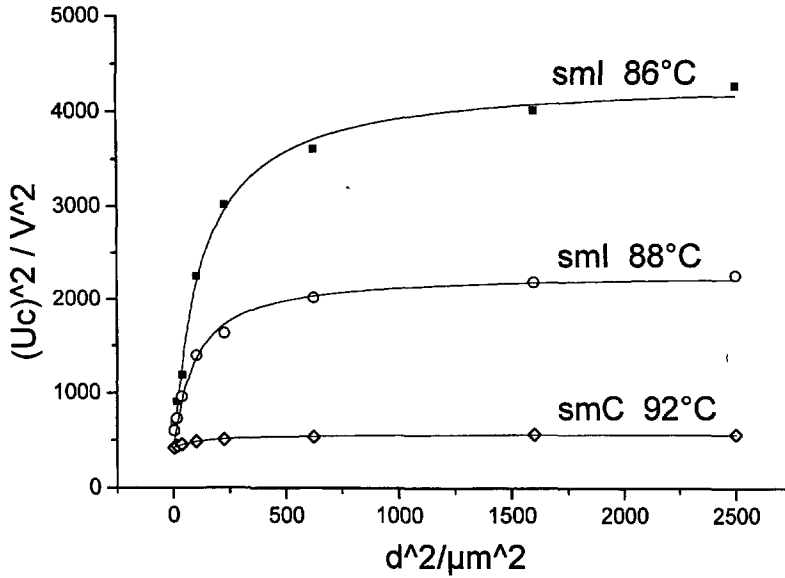


Fig. 5. — The square of the threshold voltage for the Fredericksz transition is plotted *versus* the square of the sample thickness. The experimental data are fitted by using the formula (11).

threshold voltage between the values 20.6 V ($d = 2 \mu m$) and 23.8 V ($d = 50 \mu m$). In Figure 5 the square of the threshold voltage is plotted *versus* the square of the sample thickness for 8 different cells ($d = 2, 4, 6, 10, 15, 25, 40$ and $50 \mu m$). Assuming that the elastic coupling is small ($K_{16}^2 \ll K_1 K_6$), the experimental data for different layer thicknesses are fitted by using the simplified expression

$$U_c^2 = U_0^2 \left\{ 1 + (d/d_0)^2 - \frac{(d/d_0)^4}{K_6/K_1 + (d/d_0)^2} \right\}. \quad (11)$$

Actually, the application of the complete expression (8) has only a minor effect on the parameters U_0 , d_0 and K_6/K_1 obtained from the fitting procedure. Furthermore, as the Landau theory predicts the relation $K_1 = K_1^0 \theta^2$, the value of $U_0 = \sqrt{\pi^2 K_1 / \Delta \epsilon_{\text{eff}}}$ should be nearly the same in the phases SmC and SmI as long as the tilt angle θ remains constant. Performing X-ray diffraction experiments, we found that the thickness of the smectic layers do not change significantly in the region between 86 °C and 92 °C and thus the values for θ and U_0 should be approximately constant. The experimental data for the threshold voltages obtained for the phases SmC and SmI are consistent with this conclusion. It is reasonable to determine U_0 by using the threshold data of the smectic C phase ($\vartheta = 92^\circ$), since in this phase the bond-orientational order is rather weak and therefore U_0 is not much different from the Fredericksz threshold voltage U_c . Fitting with formula (11) we obtain $U_0 = 20.6$ V, while the other parameters d_0^2 and K_6/K_1 strongly depend upon the temperature:

		d_0^2	K_6/K_1
SmC	($\vartheta = 92^\circ$ C)	$430 \mu m^2$	0.35
SmI	($\vartheta = 88^\circ$ C)	$21.8 \mu m^2$	4.4
SmI	($\vartheta = 86^\circ$ C)	$13.3 \mu m^2$	9.3

Considering the relation $K_6 = K_6^0 |\beta|^2$, the growth of the ratio K_6/K_1 with decreasing temperature is explained by a remarkable increase of the order parameter $|\beta|$ for the bond-orientational order. Furthermore, the relations $d_0^2 = \pi^2 K_1/H$ and $H = 18h|\beta|\theta^6$ indicate that d_0^2 should decrease with growing $|\beta|$ in accordance with the experimental results. Since there is only a weak hexatic ordering in the smectic C phase [15], the parameters K_6 and H are rather small. Thus the influence of the bond-orientational order on electrooptic effects in the smectic C phase can be neglected. At higher temperature ($\vartheta = 100^\circ\text{C}$) the threshold voltage was found to be almost constant justifying the assumption that the values of H and K_6 are very small beyond the SmI-SmC phase transition temperature.

Unfortunately, the experimental data are not sufficient to extract the values for the material constants K_1 , K_6 and H , as the dielectric constants of the considered biaxial smectic material are unknown. Assuming that $K_1 \approx 10^{-11}$ N, which is a reasonable value for nematic and smectic C liquid crystals, the coupling constant is estimated to be $H \approx 1$ N/m² for $\vartheta = 86^\circ\text{C}$. It should be noted that a weak hysteresis was observed. This hysteresis and the observation of moving domain walls during the switching process indicated that the Freedericksz transition was discontinuous [16] (first order transition). Raising slowly the voltage, the threshold U_c was found to be a little higher than the voltage U_d at which the elastic distortions vanish after decreasing the voltage. Unfortunately, the linear stability analysis used in this paper is not sufficient to predict whether a transition should be continuous (second order transition) or discontinuous. However, a non-linear analysis was recently carried out [4] for materials with uniaxial dielectric properties ($\epsilon_{uu} = \epsilon_{vv}$). It was shown that a discontinuous transition accompanied with hysteresis should always occur if the layer orientation declines from the bookshelf geometry (if $\mu \neq 0$). Then the switching from the homogeneous to the distorted state is usually performed by domain wall motion which starts at a voltage somewhat below the stability limit U_c . This deviation from U_c could impair the accuracy of the fitted material constants obtained from the threshold data depicted in Figure 5. Nevertheless, the general concept proposed for hexatic smectic phases with tilted molecules [2, 3], which involves a coupling of the director to the bond-net, agrees very well with the predicted threshold behaviour of the Freedericksz-effect.

Acknowledgments

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