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Field-induced reorientation dynamics in liquid crystalline hexatic phases

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A smectic hexatic phase with tilted molecules is assumed to be confined between two plane boundaries. At a finite boundary distance two dielectric modes are excited by an a.c. external electric field. Using the elastic continuum theory, equations for the relaxation times and the corresponding dielectric susceptibilities are derived and discussed in the limit of high boundary distances and for different material parameters.

1. Introduction

Smectic liquid crystals consist of a stack of fluid layers formed by rod-like molecules. In the smectic A phase the molecular long axes are parallel to the smectic layer normal. There are also several smectic phases with long molecules tilted towards the smectic layers. In the smectic C phase the tilt angle θ is fixed in thermodynamic equilibrium, while a variation of the azimuthal angle φ is possible without changing the free energy of a homogeneous sample (figure 1). Weak torques induced by an external electric field can produce large deformations accompanied by non-zero gradients of φ . Thus, the Fréedericksz transition can be observed in thin smectic C films. Compared with the nematic phase, the director rotation in the smectic C phase is confined to a cone with apex 2θ . In addition to the orientational order of the molecular long axes hexatic smectic phases also exhibit a long range bond-orientational order [1]. The net of the bonds is formed by the lines connecting the centres of gravity of nearest neighbour molecules within a smectic layer. In the hexatic smectic B phases the molecules are aligned with their long axes perpendicular to the smectic planes. The bond-net of this phase produces a six-fold rotational axis parallel to the normal of the smectic layer. A cooperative rotation of the bonds around the smectic layer normal produces a viscous torque, but does not change the free energy. However, if gradients of the bond angle η (figure 1) are different from zero,

an elastic free energy contribution arises due to the bond net deformations. The hexatic smectic I and F phases are characterized by a bond-orientational order and a tilt of the molecular long axes. In the smectic I phase the molecular long axis is tilted towards a vertex of the bond hexagon, i.e. $\varphi - \eta = 0$, whereas in the smectic F phase a tilted molecule is directed towards the edge of the hexagon ($\varphi - \eta = \pi/6$). If the tilt direction and the bond hexagons are rotated by the same angle $\delta\varphi = \delta\eta$, the free energy remains constant. In the case of different rotation angles $\delta\varphi \neq \delta\eta$, however, a free energy contribution proportional to $(\delta\varphi - \delta\eta)^2$ results from the coupling between the director and the bonds. This coupling is moderately strong and causes a remarkable enhancement of the Fréedericksz transition threshold of smectic I and F films in comparison with smectic C films [2, 3]. If the molecules are chiral, the smectic phases with tilted molecules become electrically polarized. The vector of the electric polarization in smectic C*, I* and F* phases is perpendicular to the molecular tilt plane. Furthermore, chirality produces a helical alignment of both the bond-net and the molecular long axes. This helical configuration can be suppressed by boundary interactions in thin films, if the helix period is larger than the sample thickness.

Some insight into the structure and the dynamics of smectic I* and F* phases can be given by dielectric spectroscopy. The dielectric behaviour should be influenced by the spontaneous electric polarization, the helical alignment of the molecules and the couplings between the polarization, the tilt order and the bond-orientational order. A comprehensive theoretical approach [4] based on the Landau theory predicts six

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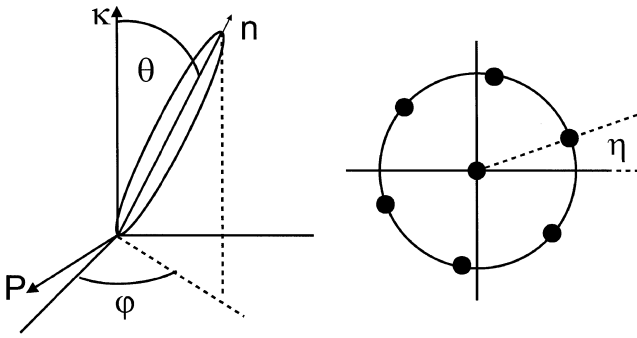


Figure 1. Director and bond-net reorientations, characterized by the angles φ and η . The director n (direction of the molecular long axes) and the smectic layer normal κ enclose a constant tilt angle θ . Chiral hexatic phases have a spontaneous electric polarization \mathbf{P} perpendicular to the tilt plane spanned by the director n and the smectic layer normal κ .

modes, which are related to amplitude and phase fluctuations of the order parameters defined for chiral hexatic I^* and F^* phases. However, recent experimental observations indicated only one dielectric mode related to combined fluctuations of the tilt direction and the bond net [5, 6].

In this paper we investigate the dielectric response of thin chiral smectic I^* and F^* films confined between two parallel plates. It is assumed that the bounding plates of the thin film produce a homogeneous alignment of the molecular long axes and the bond net. We assume that the temperature is chosen to be sufficiently far away from the hexatic–smectic A or hexatic–SmC* phase transition point. In this case amplitude fluctuations of order parameters can be neglected. An alternating electric field applied across the film couples to the electric polarization and gives rise to a viscoelastic response of the director and the bond net. This response influences the dielectric constant and can be recorded by dielectric spectroscopy. As there are two degrees of freedom ($\delta\varphi$ and $\delta\eta$), which are accompanied by relatively low free energy variations, two dielectric modes are suggested to occur. We investigate how the frequency and the amplitude of these modes depend on the film thickness.

2. Mesoscopic dynamics

We consider a ferroelectric liquid crystalline film with hexatic order confined between two plane boundaries at distance d . The smectic layers may enclose a non-zero angle μ with the film normal (figure 2). A planar reorientation of the molecular long axes leads to the relation $\sin \varphi_* = \tan \mu / \tan \theta$ for the azimuthal angle φ_* . If the structure is disturbed by an external electric field \mathbf{E} , the disturbance of the initial director and the hexagon

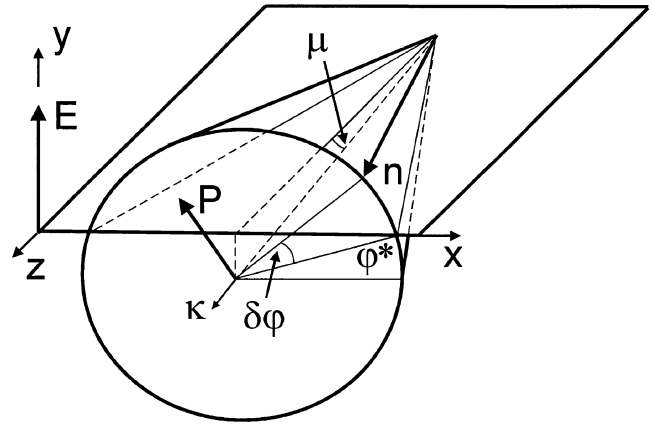


Figure 2. Alignment of the smectic phase in thin films (director n , normal of the smectic layers κ , electric field \mathbf{E}). The xz -plane is parallel to the bounding plates. The smectic layers are not exactly perpendicular to the film plane. At the boundary plates, i.e. for $y = \pm d/2$, the azimuthal director angle is always equal to the angle φ_* , corresponding to a strong surface anchoring of the director n . Inside the liquid crystal film an electric field \mathbf{E} can produce director rotations $\delta\varphi \neq 0$ accompanied by rotations of the electric polarization \mathbf{P} .

orientations are characterized by angles $\delta\varphi(y)$ and $\delta\eta(y)$, respectively. Both angles are assumed to be fixed at the boundaries so that the conditions $\delta\varphi(y = \pm d/2) = 0$ and $\delta\eta(y = \pm d/2) = 0$ are satisfied (strong anchoring). Additionally, it is supposed that the helix is completely unwound. Thus, the liquid crystalline structure in the cell is determined by surface constraints. To satisfy the boundary conditions we use the expression $\delta\varphi = \delta\varphi_0 \cos(ky)$ and $\delta\eta = \delta\eta_0 \cos(ky)$ with $k = \pi/d$. The field-induced distortions of the director and the bond net configuration can be described in the framework of a continuum theory. The starting point is a free energy expression, which includes the elastic deformation of the director field and the bond net, the direct coupling between them, and an external electric field contribution. The elastic free energy density can be written as [7]

$$f_{\text{elast}} = \frac{1}{2} \left[K \left(\frac{\partial \delta\varphi}{\partial y} \right)^2 + L \left(\frac{\partial \delta\eta}{\partial y} \right)^2 + 2G \left(\frac{\partial \delta\varphi}{\partial y} \right) \left(\frac{\partial \delta\eta}{\partial y} \right) \right] \quad (1)$$

Since $f_{\text{elast}} \geq 0$, the elastic constants obey the conditions $K > 0$, $L > 0$, and $KL - G^2 > 0$. The coupling term

$$f_{\text{coupl}} = \frac{1}{2} h (\delta\varphi - \delta\eta)^2 \geq 0 \quad (2)$$

allows us to consider small deviations of the director from the equilibrium orientation imposed by the bond net. We assume that the applied electric field is perpendicular to the film surface and spatially

homogeneous inside the sample. Then we obtain $\mathbf{P} = P(-\sin(\varphi_* + \delta\varphi), \cos(\varphi_* + \delta\varphi), 0)$ for the polarization and $\mathbf{E} = E(0, \cos\mu, \sin\mu)$ for the electric field. The electric field free energy density $f_{\text{ext}} = -P_y E$ is written as

$$f_{\text{ext}} = -PE \cos(\varphi_* + \delta\varphi) \cos\mu. \quad (3)$$

Finally, the free energy of the smectic film results from the integral

$$F = \frac{1}{d} \int_{-d/2}^{+d/2} dy (f_{\text{elast}} + f_{\text{coupl}} + f_{\text{ext}}) \quad (4)$$

where d is the film thickness. For small fluctuations this integration leads to

$$F = \frac{k^2}{4} (K\delta\varphi_0^2 + L\delta\eta_0^2 + 2G\delta\varphi_0\delta\eta_0) + \frac{h}{4} (\delta\varphi_0 - \delta\eta_0)^2 - \frac{2}{\pi} \left(\frac{\partial P_y}{\partial \delta\varphi} \right)_0 E \delta\varphi_0.$$

In equilibrium, a minimum of F requires that the conditions $\partial F / \partial \delta\varphi_0 = 0$ and $\partial F / \partial \delta\eta_0 = 0$ holds. For small deviations from equilibrium the dynamics is described by the equations of motion

$$-\frac{1}{2} \Gamma_\varphi \frac{\partial \delta\varphi_0}{\partial t} = \frac{\partial F}{\partial \delta\varphi_0}$$

and

$$-\frac{1}{2} \Gamma_\eta \frac{\partial \delta\eta_0}{\partial t} = \frac{\partial F}{\partial \delta\eta_0} \quad (5)$$

where Γ_η and Γ_φ are rotational viscosities. This leads to the following linear differential equations

$$-\Gamma_\varphi \frac{\partial \delta\varphi_0}{\partial t} = (k^2 K + h) \delta\varphi_0 + (Gk^2 - h) \delta\eta_0 - \frac{4}{\pi} \left(\frac{\partial P_y}{\partial \delta\varphi} \right)_0 E$$

and

$$-\Gamma_\eta \frac{\partial \delta\eta_0}{\partial t} = (Gk^2 - h) \delta\varphi_0 + (k^2 L + h) \delta\eta_0. \quad (6)$$

3. The susceptibility

The eigenmodes of the coupled director-bond dynamics can be obtained from equation (6), for $E=0$. Using the expressions $\delta\varphi_0 = \delta\tilde{\varphi}_0 \exp(-t/\tau)$ and $\delta\eta_0 = \delta\tilde{\eta}_0 \exp(-t/\tau)$, we arrive at the equations

$$(\tau^{-1} - \lambda_\varphi) \delta\tilde{\varphi}_0 + \lambda_1 \delta\tilde{\eta}_0 = 0 \quad (7)$$

and

$$\lambda_2 \delta\tilde{\varphi}_0 + (\tau^{-1} - \lambda_\eta) \delta\tilde{\eta}_0 = 0 \quad (8)$$

where the coefficients are defined as

$$\lambda_\varphi = \frac{h}{\Gamma_\varphi} \left[1 + \left(\frac{k}{k_0} \right)^2 \right], \quad \lambda_\eta = \frac{h}{\Gamma_\eta} \left[1 + R \left(\frac{k}{k_0} \right)^2 \right]$$

and

$$\lambda_1 = \frac{h}{\Gamma_\varphi} \left[1 - S \left(\frac{k}{k_0} \right)^2 \right], \quad \lambda_2 = \frac{h}{\Gamma_\eta} \left[1 - S \left(\frac{k}{k_0} \right)^2 \right] \quad (9)$$

with $k_0 = (h/K)^{1/2}$, $R = L/K$ and $S = G/K$. The elastic constant G essentially does not influence the general behaviour of the system. Therefore, we assume in further evaluations that S is equal to zero. The solution condition for equations (7) and (8) leads to the inverse decay times

$$\tau_{\pm}^{-1} = \frac{\lambda_\varphi + \lambda_\eta}{2} \mp \left[\left(\frac{\lambda_\varphi - \lambda_\eta}{2} \right)^2 + \lambda_1 \lambda_2 \right]^{1/2} \quad (10)$$

which are attributed to two relaxation modes and satisfy the inequality $\tau_+^{-1} < (\lambda_\varphi, \lambda_\eta) < \tau_-^{-1}$. The relaxation of the system towards the equilibrium state $\delta\varphi_0(\infty) = \delta\eta_0(\infty) = 0$ can be described by

$$\begin{pmatrix} \delta\varphi_0(t) \\ \delta\eta_0(t) \end{pmatrix} = C_+ \begin{pmatrix} 1 \\ a \end{pmatrix} \exp\left(-\frac{t}{\tau_+}\right) + C_- \begin{pmatrix} -1 \\ b \end{pmatrix} \exp\left(-\frac{t}{\tau_-}\right) \quad (11)$$

where $a = \lambda_2 / (\lambda_\eta - \tau_+^{-1}) > 0$, $b = \lambda_2 / (\tau_-^{-1} - \lambda_\eta) > 0$ and the constants C_+ , C_- are chosen to satisfy the initial conditions. The first mode with the decay time τ_+ corresponds to in-phase fluctuations of the director and the bond-net, while the second mode is related to out-of-phase fluctuations, characterized by τ_- . Both processes are shown schematically in figure 3. Because of $\tau_- < \tau_+$, out-of-phase fluctuations relax faster than in-phase fluctuations. It can be shown that τ_+ increases with decreasing values of k . If $k \rightarrow 0$, we obtain $\tau_+ \rightarrow \infty$ and $a \rightarrow 1$. In contrast, τ_- remains finite in this limit. The result is $\tau_- = (h/\Gamma_\varphi + h/\Gamma_\eta)^{-1}$ and $b \rightarrow \Gamma_\varphi/\Gamma_\eta$ for $k \rightarrow 0$. Figure 4 depicts the ratio τ_+/τ_- for different values of the elastic constants ratio $R = L/K$. For small wave vectors, $k/k_0 \ll 1$, the inverse decay times can be written as ($\Gamma = \Gamma_\varphi/\Gamma_\eta$)

$$\left(\frac{h}{\Gamma_\varphi} \tau_+ \right)^{-1} = \frac{\Gamma(1+R)}{1+\Gamma} \left(\frac{k}{k_0} \right)^2 + \mathcal{O}\left((k/k_0)^4 \right) \quad (12)$$

and

$$\left(\frac{h}{\Gamma_\varphi} \tau_- \right)^{-1} = 1 + \Gamma + \frac{1 + \Gamma^2 R}{1 + \Gamma} \left(\frac{k}{k_0} \right)^2 + \mathcal{O}\left((k/k_0)^4 \right). \quad (13)$$

The response of the hexatic phase to a weak alternating

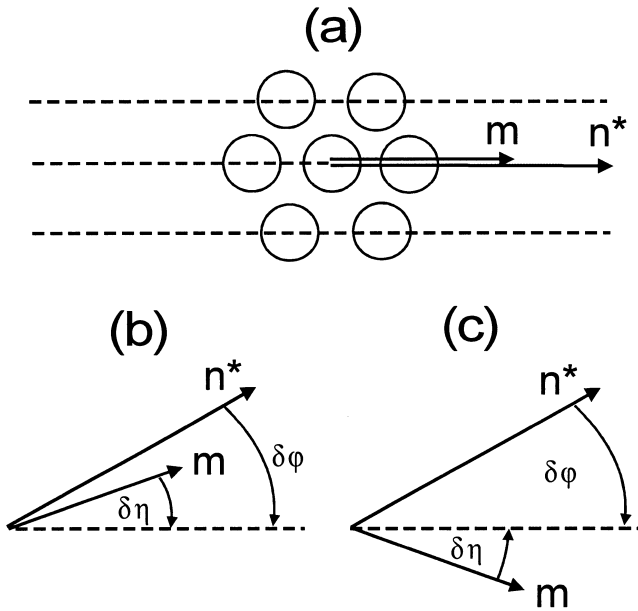


Figure 3. Modes for director and bond relaxations towards the equilibrium configuration $\delta\varphi_0=0$ and $\delta\eta_0=0$ for smectic I* phases. The vector m defines the direction of the bonds and n^* is the projection of the director onto the smectic layer plane. (a) In equilibrium the vectors m and n are parallel to the dashed line, which defines their stable alignments imposed by the boundary conditions (director and bond anchoring at the plates). (b) Slow relaxation mode with relaxation time τ_+ . (c) Fast mode with relaxation time τ_- .

electric field is different from the relaxation process described by equation (11), but the relaxation times τ_+ and τ_- enter into the final results. A periodic external field $E(t) = \tilde{E}(\omega) \exp(i\omega t)$ leads to harmonic oscillations $\delta\eta_0(t) = \delta\tilde{\eta}_0(\omega) \exp(i\omega t)$ and $\delta\varphi_0(t) = \delta\tilde{\varphi}_0(\omega) \exp(i\omega t)$.

Then the equations of motion (5) are expressed as

$$(i\omega + \lambda_\varphi)\delta\tilde{\varphi}_0 - \lambda_1\delta\tilde{\eta}_0 = \frac{4}{\pi} \left(\frac{\partial P_y}{\partial \delta\varphi} \right)_0 \tilde{E} \quad (14)$$

and

$$-\lambda_2\delta\tilde{\varphi}_0 + (i\omega + \lambda_\eta)\delta\tilde{\eta}_0 = 0. \quad (15)$$

Solving equations (14) and (15) leads to

$$\begin{pmatrix} \delta\tilde{\varphi}_0 \\ \delta\tilde{\eta}_0 \end{pmatrix} = \frac{\tau_- \tau_+}{(1+i\omega\tau_-)(1+i\omega\tau_+)} \begin{pmatrix} 1+i\omega\tau_\eta \\ \lambda_2\tau_\eta \end{pmatrix} \frac{4}{\pi\tau_\eta\Gamma_\varphi} \left(\frac{\partial P_y}{\partial \delta\varphi} \right)_0 \tilde{E} \quad (16)$$

($\tau_\eta = 1/\lambda_\eta$). The dielectric susceptibility is defined by the

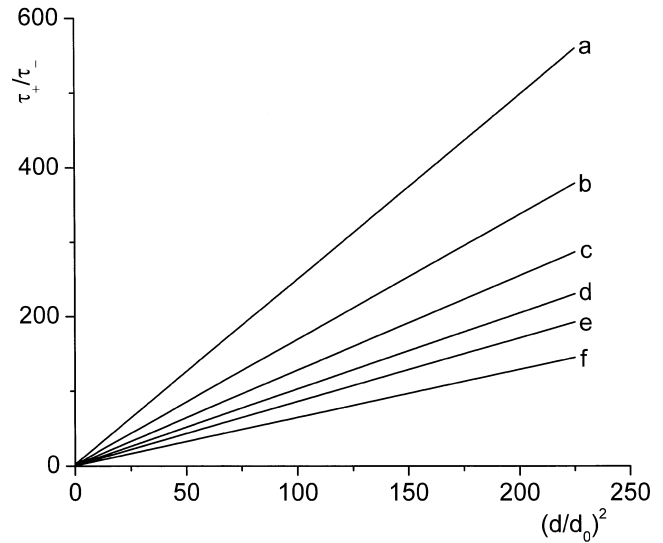


Figure 4. Plot of the relaxation time τ_+/τ_- versus reduced film thickness $(d/d_0)^2$ for the viscosity ratio $\Gamma = 1/50$ and several ratios of the elastic constants $R=L/K$. (a) $R=20$; (b) $R=30$; (c) $R=40$; (d) $R=50$; (e) $R=60$; (f) $R=80$.

relation

$$\chi = \lim_{E \rightarrow 0} \frac{\langle \delta P_y \rangle}{E} \quad (17)$$

where

$$\langle \delta P_y \rangle = \frac{1}{d} \int_{-d/2}^{+d/2} \delta P_y(y) dy. \quad (18)$$

By using the relation $\delta P_y = (\partial P_y / \partial \delta\varphi)_0 \delta\varphi$ and the equations (16)–(18) we arrive at $\langle \delta \tilde{P}_y(\omega) \rangle = \chi(\omega) \tilde{E}(\omega)$, with the frequency-dependent dielectric susceptibility

$$\chi(\omega) = \frac{\chi(0)}{\tau_+ - \tau_-} \left[\frac{\tau_+ - \tau_\eta}{1+i\omega\tau_+} + \frac{\tau_\eta - \tau_-}{1+i\omega\tau_-} \right] \quad (19)$$

and its static value

$$\chi(0) = (\tau_- \tau_+ / \tau_\eta \Gamma_\varphi) (4\pi^{-1} P \sin \varphi_* \cos \mu)^2.$$

Equation (19) can be rewritten as a sum $\chi(\omega) = \chi_+(\omega) + \chi_-(\omega)$, where each term is attributed to a Debye relaxation process. For $\omega=0$ the ratio of susceptibilities $\chi_+(0)/\chi_-(0) = (\tau_+ - \tau_\eta)/(\tau_\eta - \tau_-)$ is plotted in figure 5.

4. Discussion

In the absence of an electric field the coupled dynamics of molecular tilt and bond-net reorientations can be described by superimposing in-phase and out-of-phase fluctuations with decay times τ_+ and τ_- , respectively. These relaxation times satisfy the condition $\tau_+ > \tau_-$. For small wave vectors, $k \rightarrow 0$, the inverse

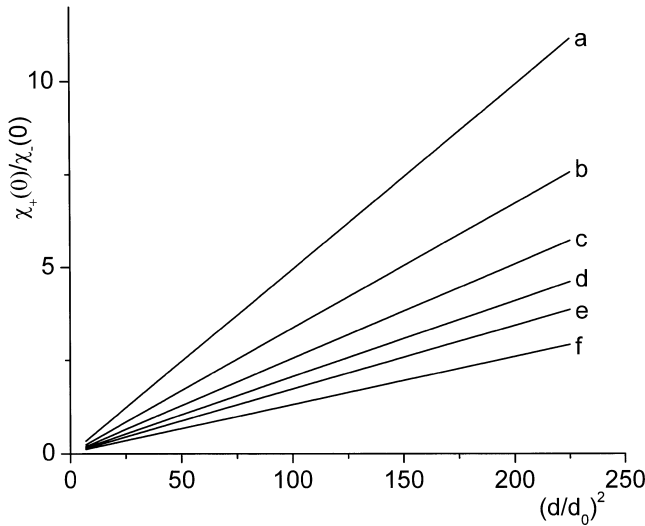


Figure 5. Plot of the susceptibility ratio $\chi_+(0)/\chi_-(0)$ versus $(d/d_0)^2$ for the viscosity ratio $\Gamma=1/50$ and several ratios of the elastic constants $R=L/K$. (a) $R=20$; (b) $R=30$; (c) $R=40$; (d) $R=50$; (e) $R=60$; (f) $R=80$.

relaxation time τ_+^{-1} tends to zero, whereas τ_-^{-1} remains finite. Thus, the relaxation rates behave similarly to an acoustic and an optical branch known from solid crystals [8]. But, in contrast to the zero-frequency mode of solids, where the dispersion is linear, the inverse decay time behaves as $\tau_+^{-1} \sim k^2$. If an alternating electric field is applied, the dielectric susceptibility $\chi(\omega)$ can be expressed as a sum of two Debye spectra $\chi_+(\omega)$ and $\chi_-(\omega)$, which are associated with the characteristic relaxation times τ_+ and τ_- . In experimental investigations both relaxation processes should be distinguishable if $\tau_+/\tau_- \gg 1$ and if the amplitude of the weaker Debye process is not negligibly small. The latter requirement may be written as $0.1 < \chi_+(0)/\chi_-(0) < 10$, although this condition is too stringent for practical cases. For evaluating the ratios τ_+/τ_- and $\chi_+(0)/\chi_-(0)$ experimental data are required for the ratios R , Γ and the characteristic distance d_0 . Numerical values for R are available for example from investigations of the Fréedericksz transition [3] and from light scattering experiments [9]. The ratio R ranges over two orders of magnitude, $5 \lesssim R \lesssim 200$. Values for the viscosity ratio Γ are not yet available. Since the viscosity Γ_ϕ for rotations of the molecular tilt plane is much lower than the viscosity Γ_η of bond-net reorientations, the condition $\Gamma \ll 1$ should be satisfied. Experimental results [9] suggest that the length $d_0 = \pi(K/h)^{1/2}$ is of the order of a micrometre. Accordingly, the condition $k/k_0 = d_0/d \ll 1$ is satisfied, since in typical experiments the cell thickness d is considerably larger than one micrometre. Then, for $R \gg 1$ and $\Gamma \ll 1$ equations (12), (13) and the

relation $h = K(\pi/d_0)^2$ lead to the relaxation times

$$\tau_+ \simeq \frac{1}{\Gamma R} \left(\frac{\Gamma_\phi}{\pi^2 K} \right) d^2 \quad (20)$$

and

$$\tau_- \simeq \left(\frac{\Gamma_\phi}{\pi^2 K} \right) d_0^2. \quad (21)$$

It is expected that the elastic constant K and the viscosity Γ_ϕ are comparable to the twist elastic constant and the rotational viscosity of a nematic liquid crystal, respectively. By using $K \simeq 10^{-6}$ dyn, $\Gamma_\phi \simeq 0.1$ P [10], $d_0 = 1 \mu\text{m}$ and $d = 10 \mu\text{m}$, the estimations $\tau_+ \simeq 10^{-2}$ [s]/ (ΓR) and $\tau_- \simeq 10^{-4}$ [s] result. If $\Gamma R < 10$, both relaxation modes are well separated. However, for $\Gamma R < 10^{-2}$ the relaxation frequency $1/\tau_+$ is rather low, namely smaller than 1 Hz. On the other hand, if $d_0 \simeq 0.1 \mu\text{m}$, the relaxation frequency $1/\tau_-$ could be greater than 10^6 Hz.

Figures 4 and 5 depict the ratios of the characteristic times τ_+/τ_- and susceptibility amplitudes $\chi_+(0)/\chi_-(0)$ in dependence of the reduced film thickness $d/d_0 = (k/k_0)^{-1}$ for different values of the elastic ratio R . Both ratios are almost linear functions of the square $(d/d_0)^2$. Using relations (10) and equations (9), this behaviour is described by the leading terms of the asymptotic expansions ($d/d_0 \gg 1$, $\Gamma \ll 1$)

$$\frac{\tau_+}{\tau_-} = \frac{1}{\Gamma R} \left(\frac{d}{d_0} \right)^2 + \frac{1}{\Gamma R} + O\left((d/d_0)^{-2} \right) \quad (22)$$

and

$$\frac{\chi_+(0)}{\chi_-(0)} = \frac{1}{R} \left(\frac{d}{d_0} \right)^2 + 2\Gamma - \frac{1}{R} + O\left((d/d_0)^{-2} \right). \quad (23)$$

Obviously, the relaxation times are well separated for a wide range of the product ΓR , because $(d/d_0)^2$ can exceed even the value 10^3 . Additionally, at least for large R , the ratio of the susceptibility amplitudes can satisfy the required condition $0.1 < \chi_+(0)/\chi_-(0) < 10$. In this case two modes should be clearly observable. Because the helix is assumed to be completely unwound, the distance between the boundaries d should not exceed the helix pitch p .

Finally, let us consider a very thick cell satisfying $d \gg p$, where p is the helix period. Then a helical alignment of the director and the bond-net is stable. For sufficiently thick cells the disturbance of the helical alignment due to the boundaries can be neglected. As shown in the appendix, equations (16), (19), (22) and (23) remain essentially valid if d is replaced by $p/2$. In this case the replacement $(d/d_0)^2 \rightarrow (dp)^2/4$ in the relations (22) and (23) leads to analogous conclusions as previously. Two separated relaxation modes could be detected experimentally if the square $(p/d_0)^2$ is appropriately adjusted.

In recent experiments only one dielectric relaxation mode has been observed [5, 6]. These measurements are carried out in a frequency range from 1 Hz to 10^7 Hz and for a thick sample. Systematic investigations with varying cell thickness d or helical pitch p are required to check the theoretical predictions.

Appendix

Very thick cells

For a sufficiently thick cell the disturbance of the helix due to the boundary alignment can be neglected. In this case the helix period p satisfies the condition $d \gg p$. For an disturbed sample the helix configuration is described by $\varphi = (2\pi/p)z + \delta\varphi(z, t)$ and $\eta = (2\pi/p)z + \eta_1 + \delta\eta(z, t)$, where $\eta_1 = 0$ (smectic I* phase) or $\eta_1 = \pm\pi/6$ (smectic F* phase). Assuming that the helix is parallel to the z -axis and the electric field parallel to the y -axis, the free energy density is written as [11]

$$f_h = \frac{1}{2} \left[K \left(\frac{\partial\varphi}{\partial z} \right)^2 + L \left(\frac{\partial\eta}{\partial z} \right)^2 \right] + \frac{h}{2} (\delta\varphi - \delta\eta)^2 - P_y E \quad (\text{A1})$$

where $P_y = P \cos\varphi$. Linear gradient terms, which produce the helical alignment, are not considered here, since these terms do not enter into the equations of motion

$$-\Gamma_\varphi \frac{\partial\varphi}{\partial t} = \frac{\delta F_h}{\delta\varphi}$$

and

$$-\Gamma_\eta \frac{\partial\eta}{\partial t} = \frac{\delta F_h}{\delta\eta}.$$

Using the free energy $F_h = p^{-1} \int_0^p dz f_h(z)$ these equations are written as

$$-\Gamma_\varphi \frac{\partial\varphi}{\partial t} = -K \frac{\partial^2\varphi}{\partial z^2} + h(\delta\varphi - \delta\eta) + PE \sin\varphi \quad (\text{A2})$$

and

$$-\Gamma_\eta \frac{\partial\eta}{\partial t} = -L \frac{\partial^2\eta}{\partial z^2} - h(\delta\varphi - \delta\eta). \quad (\text{A3})$$

Taking into account that a small alternating external field $E = \tilde{E} \exp(i\omega t)$ produces only very weak disturbances $\delta\tilde{\varphi}(z) \exp(i\omega t)$ and $\delta\tilde{\eta}(z) \exp(i\omega t)$ of the orientation fields, the equations of motions are rewritten as

$$-i\omega\Gamma_\varphi \delta\tilde{\varphi} = -K \frac{\partial^2\delta\tilde{\varphi}}{\partial z^2} + h(\delta\tilde{\varphi} - \delta\tilde{\eta}) + P \sin(qz) \quad (\text{A4})$$

and

$$-i\omega\Gamma_\eta \delta\tilde{\eta} = -L \frac{\partial^2\delta\tilde{\eta}}{\partial z^2} - h(\delta\tilde{\varphi} - \delta\tilde{\eta}). \quad (\text{A5})$$

These equations can be solved by using the expressions $\delta\tilde{\varphi} = \delta\tilde{\varphi}_0 \sin(qz)$ and $\delta\tilde{\eta} = \delta\tilde{\eta}_0 \sin(qz)$, where $q = 2\pi/p$. Thus we obtain

$$-i\omega\Gamma_\varphi \delta\tilde{\varphi}_0 = q^2 K \delta\tilde{\varphi}_0 + h(\delta\tilde{\varphi}_0 - \delta\tilde{\eta}_0) + P\tilde{E} \quad (\text{A6})$$

and

$$-i\omega\Gamma_\eta \delta\tilde{\eta}_0 = q^2 K \delta\tilde{\eta}_0 - h(\delta\tilde{\varphi}_0 - \delta\tilde{\eta}_0). \quad (\text{A7})$$

These equations formally coincide with the equations (14) and (15) if the length d is replaced by $p/2$, where p is the helix period p (pitch). The polarization P corresponds to $(-4/\pi)(\partial P_y / \partial \delta\varphi)_0$.

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