Book Chapter

Dynamics of the Director Reorientation and Light Modulation in Helix-Free Ferroelectric Liquid Crystals

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Abstract

The dynamics of the director reorientation in new helix-free ferroelectric liquid crystals (FLC) is considered. These materials are specially designed helix-free FLCs with a rather low value of the spontaneous polarization (less than $50 \text{ nC} / \text{cm}^2$) and high viscosity (from 0.3 to 1.0 Poise), which are characterized by a spatial periodic deformation of smectic layers in the absence of an electric field. FLC director reorientation is due to the motion of solitons – spatially localized waves of a stationary profile that arise in an alternating electric field upon transition to the Maxwellian mechanism of energy dissipation. A theoretical model is proposed for describing the spatial-periodic deformation of FLC and reorientation of its director. The frequency and field experimental dependences of FLC electro-optical response time are presented for the modulation of the light transmission with fastest response among all LC materials. The novel helix-free FLC are able to efficiently modulate the visible and near IR radiation at frequencies up to 7 kHz at the electric field strength of the order of $1\text{-}2 \text{ V}/\mu\text{m}$. The conditions for the continuous hysteresis-free electro-optical response were determined, and such a response was realized for the first time in the frequency range up to 6 kHz.

Key Words

Liquid Crystal; Ferroelectric; Smectic Layer; Helix; Soliton; Director Orientation; Transparent Mode; Bistable Light Scattering

Introduction

The character of the process of FLC main optical axis (director) reorientation under the action of an alternating electric field depends on which of the two dissipative coefficients – rotational or shear viscosity predominates [1, 2].

If the FLC is subjected to an electric field varying periodically with the frequency f , and the period of variation is large ($\tau_m f \ll$

1) in comparison with the Maxwellian relaxation time τ_m [3], then the FLC behaves as a fluid with a rotational viscosity γ_φ . In this case, a time of the director reorientation $\tau_R \sim \gamma_\varphi / (P_S \cdot E)$, where E is the strength of an external electric field, and P_S is the FLC spontaneous polarization [4]. Correspondingly, a time of an electro-optical response $\tau_{0,1-0,9}$, proportional to the time τ_R , is also determined by the ratio of the viscosity γ_φ to the polarization P_S and does not depend on the frequency of an electric field change.

However, to increase the speed of the response, it is not possible to significantly reduce the ratio γ_φ / P_S , because P_S increasing usually leads to an increase in γ_φ . This circumstance limits the frequency of the light modulation if the voltage applied to an electro-optical cell does not exceed several volts. Besides, at P_S that is sufficiently more than 50 nC/cm^2 the saturation voltage increases, ferroelectric domains begin to arise, and the residual light scattering appears after electric field switching off.

On the other hand, at rather high frequencies ($\tau_m \cdot f \gg 1$) FLC can behave as an amorphous solid body, and in this case the dissipative coefficient is the shear viscosity. Its predominance leads to a change in the character of the motion of the helix-free FLC director in weak electric fields: the director reorientation occurs due to the motion of domain boundaries – 180-degree domain walls [1,2]. Such a reorientation process makes it possible to obtain the light modulation frequency of the order of 3 kHz at the electric field strength about $1 \text{ V} / \mu\text{m}$. In such a case the electro-optical response time is $50 \div 70 \mu\text{s}$.

The 180-degree domain wall separates two equilibrium domains, in which the spontaneous polarization vector lies either along the direction of an external electric field or against the field direction [5]. 180-degree walls (or topological solitons) separating two different basic states of a system characterize the general nonlinear properties of a medium. The motion of such solitons in the direction orthogonal to smectic layers changes the azimuth angle φ of the director's orientation from 0 to π (or from π to 0). The smectic layers are treated here as the periodic ordering of the FLC molecules mass centers in the direction of FLC director with a period of the order of the molecule length. Thus, the

topological soliton dynamics describes the motion of the orientational inflection (kink or antikink) in an external electric field.

The combined influence of the medium nonlinearity and the presence of the dispersion of velocities of deformation (or displacement) waves sharply increases the steepness of the wave front and the growth of gradients of field variables. This results in a spatial redistribution of the excitation energy and its localization – the wave period tends to infinity. Such solitons called dynamic solitons characterize local nonlinear properties of a medium. For the first time a director reorientation through solitons was proposed in [6]. In contrast to topological solitons, the dynamic soliton is continuously transformed into the homogeneous major state. The shape of dynamic solitons is definitely relates to independent parameters, in particular, the motion velocity [7,8].

Below, we consider the process of the director reorientation in the helix-free FLC due to the motion of dynamic solitons that arise during the transition to the Maxwellian mechanism of energy dissipation in an alternating electric field.

A new type of low-voltage FLC materials with a small value of spontaneous polarization is considered. In the volume of this FLC at the absence of an electric field the spatially inhomogeneous structures with periodic deformations of smectic layers are observed with a deformation pitch from 1.5 to 5 μm , and the birefringence depends on the electric field frequency.

In such FLCs the change in the energy dissipation channel, that is, the transition to the shear viscosity, is accompanied by a strong frequency dependence of the electro-optical response time. If the shear viscosity prevails, then the reorientation of the FLC director is due to the motion of soliton waves. This provides the light modulation frequency in the visible and near infrared range of a few kilohertz at the electric field strength of $1\text{V}/\mu\text{m}$. The electro-optical response time in the soliton mode depends on the electric field change frequency rather weak [9].

Deformation of Smectic Layers

For the studied helix-free FLC the physical cause of the frequency dependence of the birefringence is the periodic deformation of smectic layers. It results in the formation of spatially inhomogeneous structures in FLC volume, the change of the director position and therefore the position of the main optical axis along smectic layers [10,11]. This periodic deformation at the absence of an electric field is observed behind the analyzer in the form of alternating light and dark stripes, when the smectic layers are parallel to substrates of an electro-optical cell (homeotropic director orientation) [9].

Let us consider the energy nature of origing the periodic deformation. Its presence means that in smectic layers the FLC molecules initially inclined at an angle Θ_0 with respect to the normal to the layer at a given point are additionally deflected by some angle Ψ relatively to the substrate plane (Figure 1).

Figure 1: (Reprinted with permission from SID 2012 DIGEST, pp. 452-455). The electro-optical cell with helix-free FLC (a) and a fragment of deformed smectic layer (b). 1 – glass substrates with conductive covers, 2 – smectic layers, Θ_0 – the angle of molecule's tilt in smectic layers, ψ – the local tilt angle of a deformed smectic layer with respect to z-axis, P_s – vector of the spontaneous polarization, d – FLC cell thickness, l – smectic layer thickness.

As the result, the projection of the director on the xy plane changes, and the thickness of a curved smectic layer in the projection onto the z axis can be written as

$$l = l_0 \cos \Theta_0 / \cos \psi. \quad (1)$$

The bulk free energy density associated with such a deformation of layers can be written in the following form [10, 12]:

$$F = \frac{1}{2} K \left(\frac{d\psi}{dy} \right)^2 + \frac{1}{2} M \left(\frac{l - l_0}{l_0} \right)^2, \quad (2)$$

where K is the elasticity coefficient describing the director deformation with respect to the angle ψ due to the smectic layers periodic distortion, M is the bending energy of smectic layers. From relation (1), taking into account the smallness of the angles ψ and Θ_0 ($\psi, \Theta_0 \ll 1$), we obtain a relative change in the thickness of a smectic layer [13]:

$$(l - l_0) / l_0 \cong (\psi^2 - \Theta_0^2) / 2. \quad (3)$$

Using Euler's equation, we minimize the functional (2). Taking into account relation (3) and denoting $\alpha = M d^2 / 2K$ we arrive now to an equation that describes the distribution of the angle ψ along the coordinate y ($0 \leq y \leq d$, and d is the thickness of the electro-optical cell):

$$d^2 \psi / dy'^2 + \alpha \Theta_0^2 \psi - \alpha \psi^3 = 0, \quad (4)$$

where $y' = y/d$, $0 \leq y' \leq 1$.

Until we will not consider the energy of interaction of FLC molecules with confining surfaces and assume that

$$\psi(y=0) = 0 \text{ and } d\psi/dy=0 \text{ at } \psi_{max} = \pm \Theta_0. \quad (5)$$

The last condition means that the maximum value of the angle ψ is equal to the molecular tilt angle Θ_0 in smectic layers. If the angle $\psi = \Theta_0$, the main optical axis (the FLC director) deviates by an angle Θ_0 from the plane of the incident light polarization.

There are three trivial solutions of equation (4): $\psi = 0$ and $\psi = \pm \Theta_0$. The solution $\psi = 0$ corresponds to a structure with smectic layers that are orthogonal to the substrate plane. The solutions ψ

$= \pm \Theta_0$ give the absolute minimum of the free energy $F = 0$ and correspond to a structure with tilt layers.

The exact solution of equation (4) is written in terms of the Jacobi elliptic sine [7]:

$$\psi = \sqrt{2} \Theta_0 \frac{k}{\sqrt{1+k^2}} \operatorname{sn} \left(\Theta_0 d \sqrt{\frac{M}{2K}} \frac{(y' + C_1)}{\sqrt{1+k^2}}, k \right), \quad (6)$$

$$\text{where } k = \sqrt{\frac{\alpha \Theta_0^4}{4C_2}} - \sqrt{\frac{\alpha \Theta_0^4}{4C_2} - 1}, 0 < C_2 < \frac{M \Theta_0^4 d^2}{8K},$$

$0 < k < 1$ is the Jacobi elliptic sine module, C_1 and C_2 being integration constants.

To find the solution of equation (4) in elementary functions, we used the van der Pol method (the averaging method) [7]. The general solution (the basic approximation) for the equation (4) has the following form: $\psi_0(y') = C \sin(\Theta_0 \sqrt{\alpha} y' + \eta) \equiv C \sin \delta$, where C and η are arbitrary constants.

We seek the solution of equation (4) in the same functional form as the solution of the generating equation, but taking into account the fact that the amplitude C and phase η depend on the coordinate:

$$\psi(y') = C(y') \sin(\Theta_0 \sqrt{\alpha} y' + \eta(y')) \equiv C(y') \sin \delta(y'). \quad (7)$$

Expression (7) contains two independent coordinate functions, and to find them we need two equations. We also require the derivative $d\psi/dy'$ has the same functional form as for the generating equation, that is,

$$\frac{d\psi}{dy'} = C(y') \Theta_0 \sqrt{\alpha} \cos \delta(y'). \quad (8)$$

Differentiating (7) and equating the derivative (8), we obtain the first equation:

$$\frac{dC}{dy'} \sin \delta + C \frac{d\eta}{dy'} \cos \delta = 0. \quad (9)$$

Differentiating (8) and substituting into equation (4), we obtain the second equation:

$$\frac{dC}{dy'} \cos \delta - C \frac{d\eta}{dy'} \sin \delta = \frac{C^3}{\Theta_0} \sqrt{\alpha} \sin^3 \delta. \quad (10)$$

Equations (9) and (10) represent a system of first-order differential equations for $C(y')$ and $\eta(y')$, which is solved with respect to the derivatives:

$$\frac{d\eta}{dy'} = -\frac{\sqrt{\alpha}}{\Theta_0} C^2 \sin^4 \delta, \quad (11)$$

$$\frac{dC}{dy'} = \frac{\sqrt{\alpha}}{\Theta_0} C^3 \sin^3 \delta \cos \delta. \quad (12)$$

We replace the right-hand sides of equations (11) and (12) by their average values over a period in which the phase δ changes by 2π , where $\delta = \Theta_0 \sqrt{\alpha} y' + \eta$. From the averaged equations, we obtain:

$$\frac{d\delta}{dy'} = \Theta_0 \sqrt{\alpha} - \frac{\sqrt{\alpha}}{\Theta_0} \frac{3C^2}{8}, \quad (13)$$

$$\frac{dC}{dy'} = 0. \quad (14)$$

From equations (13) and (14) it follows that

$$\delta = \left(\Theta_0 \sqrt{\alpha} - \frac{\sqrt{\alpha}}{\Theta_0} \frac{3C^2}{8} \right) y' + B \quad \text{and} \quad C = \text{const.}$$

Thus, the approximate solution of equation (4) has the following form: (15)

$$\psi = C \sin \delta = C \sin \left(\Theta_0 \sqrt{\frac{M}{2K}} \left(1 - \frac{3}{8} \frac{C^2}{\Theta_0^2} \right) y + B \right)$$

From the boundary conditions (5) we find the constants: $C = \Theta_0$, $B = 0$. Substituting them into the solution (15) we obtain:

$$\psi = \Theta_0 \sin \left(\frac{5}{8} \Theta_0 \sqrt{\frac{M}{2K}} y \right). \quad (16)$$

To find the solution (16) the interaction of FLC molecules with the surface that is the anchoring energy (the anisotropic part of the surface tension) was not taken into account. However, for electro-optical cells, whose thickness is $d \leq 2 \mu\text{m}$, surface interactions affect rather strongly on the over molecular structure of FLC. The anchoring energy will be taken into account if we will determine the energy M of smectic layer deformation corresponding to these thicknesses.

The energy of layer deformation can be found from the following relation for the static value of the dielectric susceptibility [11]:

$$\chi_{st} = \frac{1}{2} \frac{P_s^2}{\Theta_0^2 M}, \quad (17)$$

that corresponds to the linear response of a structure to the electric field action and does not depend on the electric field strength [14,15].

When the electric field change is slow (the frequency of the bipolar voltage of triangular shape is less than 1 Hz), the moment of viscous friction forces practically does not affect on the susceptibility χ_{st} . In this case, the susceptibility χ_{st} corresponds to an elastic (reversible) process of changing the distribution of the angle ψ , which describes the deformation of smectic layers under

conditions, when there are weak structure distortions and relaxation to the unperturbed state with decreasing the electric field strength.

If the reorientation of the dipole moments of molecules occurs in a phase with a change of an electric field, that is, there is no phase shift between the exciting action and the dielectric response of the structure, then the susceptibility χ_{st} is a constant. In particular, for FLC with helicoidally twisted directors, the section of the dependence $\chi_{st}(f)$, where the static susceptibility does not depend on the frequency of the electric field change, is in the range from 1 to 10 Hz [15].

For the electro-optical cell with a thickness of 1.7 μm , where the specific helix-free FLC was used as the modulating medium (the pitch of smectic layers deformation $d \approx 2 \mu\text{m}$, spontaneous polarization $P_S = 40 \text{ nC} / \text{cm}^2$, rotational viscosity coefficient $\gamma_\varphi = 0.7 \text{ P}$, tilt angle of molecules in smectic layers $\Theta_0 \approx 22^\circ$), the value $\chi_{st} \approx 5.5$ corresponds to the reversible process of changing the angle ψ distribution (Figure 2). In this case, the energy M of smectic layers deformation, found from the relation (17), is about $12 \cdot 10^3 \text{ erg} / \text{cm}^3$. For an electro-optical cell with a thickness of 5 μm the deformation energy decreases to $4 \cdot 10^3 \text{ erg} / \text{cm}^3$ [11].

Figure 2: Frequency dependence of the dielectric susceptibility of the helix-free FLC with periodic deformations of smectic layers. Bipolar voltage of triangular shape, amplitude $\pm 0.1 \text{ V}$. The electro-optical cell thickness is 1.7 μm .

The analytical solution (16) describes the FLC structure with smectic layers local deformation ψ as a function of the y -coordinate, normal to the cell substrates – Figure 3).

The pitch of smectic layers deformation increases with increasing a thickness of the electro-optical cell approximately up to $7\text{ }\mu\text{m}$ and then practically does not change that is confirmed experimentally (Figure 4).

Figure 3: The calculated values of the angle ψ for electro-optical cells $1.7\text{ }\mu\text{m}$ (1) and $5\text{ }\mu\text{m}$ (2) thick. $\Theta_0 = 23^\circ$, $K = 5 \cdot 10^{-12}\text{ N}$.

Figure 4: Dependence of the deformation pitch of smectic layers on the thickness of the electro-optic cell with the composition HF-32F. The orientation of molecules is homeotropic.

The Mechanism of FLC Director Reorientation

The reorientation of the FLC director due to the interaction of the alternating electric field E (applied along the coordinate y in Figure 1) with spontaneous polarization P_s can occur both at the change of the azimuthal orientation angle φ of the director by 180° (when the director is reoriented along the cone generatrix with its turn $2\Theta_0$) and at the change of the angle ψ distribution characterizing the deformation of smectic layers (Figure 1). In the first case, the dissipative coefficient is the rotational viscosity γ_φ , and in the second case – the viscosity γ_ψ for the shear deformation.

We add to the expression (2) the bulk free energy density of the interaction between the electric field and the FLC spontaneous polarization [10]:

$$\frac{1}{2} P_s E \cos \varphi_0 \left(\frac{l - l_0}{l_0} \right)^2, \quad (18)$$

where φ_0 is the initial azimuth angle of the director's orientation. Minimizing the new expression for the free energy, we obtain the dynamic equation describing the change of the angle ψ in the external electric field, which is determined by the balance of electric, elastic and viscous moments of forces acting on the FLC director:

$$-\gamma_\psi \frac{\partial \psi}{\partial t} = K \frac{\partial^2 \psi}{\partial y^2} + \frac{(P_s E \cos \phi_0 + M)}{2} \psi (\Theta_0^2 - \psi^2), \quad (19)$$

where $K \cdot \partial^2 \psi / \partial y^2$ and $\gamma_\psi \cdot \partial \psi / \partial t$ are the elastic and viscous moments, K is the elasticity coefficient describing the deformation of the director with respect to the angle ψ , γ_ψ is the viscosity for shear deformation, t is the time, and the electric field is turned at $t = 0$.

Equation (19) does not take into account the change of the angle Θ_0 in the external electric field that is correct far from the phase transition to the paraelectric (smectic A^*). In addition, the reorientation of FLC director over the azimuth angle φ is not

considered. Here we consider the director reorientation due to the dynamic soliton motion along smectic layers that occurs during the transition to the Maxwellian mechanism of energy dissipation, when the shear viscosity prevails.

The boundary conditions for equation (19) are analogous to the boundary conditions (5). In addition, it is assumed that at $t = 0$ the distribution (16) is preserved.

We introduce new variables: (20)

$$a' = d^2(P_s E \cos \varphi_0 + M)/(2K), y' = y/d, t' = t/t_0, t_0 = \gamma_\psi d^2/K.$$

Then equation (19) is transformed to the following form:

$$\frac{\partial \psi}{\partial t'} + \frac{\partial^2 \psi}{\partial y'^2} + \alpha' \Theta_0^2 \psi - \alpha' \psi^3 = 0. \quad (21)$$

In general case ψ depends both on the coordinate and time: $\psi = \psi(y', t') = \psi_0 \exp(-i\zeta t')$, where $\zeta > 0$ is a constant, and ψ_0 is an amplitude [7]. Then the equation (21) can be written in the following form:

$$\frac{\partial \psi_0}{\partial t'} + \frac{\partial^2 \psi_0}{\partial y'^2} + \psi_0 (\alpha' \Theta_0^2 + i\zeta) - \alpha' \psi_0^3 = 0 \quad (22)$$

We substitute $\psi_0 = \Phi \exp(-i\eta)$ into the equation (22) and obtain a system of two equations:

$$\begin{cases} \partial^2 \Phi / \partial y'^2 + \Phi \alpha' \Theta_0^2 - \alpha' \Phi^3 = 0, \\ \Phi (\partial \eta / \partial t') - \Phi \zeta = 0. \end{cases} \quad (23)$$

From the system (23) we obtain:
 $\Phi = (\sqrt{2} \Theta_0 k / \sqrt{1+k^2}) \operatorname{sn}[\Theta_0 \sqrt{\alpha'} (y' + C_1) / \sqrt{1+k^2}, k]$ and $\eta = \zeta t' + C_2$. Then the expression $\psi = \psi_0 \exp(-i\zeta t')$ is transformed to the following form: (24)

$$\psi = (\sqrt{2} \Theta_0 k / \sqrt{1+k^2}) \operatorname{sn}[\Theta_0 \sqrt{\alpha'} (y' + C_1) / \sqrt{1+k^2}, k] \exp(-2i\zeta t' - iC_2) \quad .$$

For $k \rightarrow 1$, when the wave period tends to infinity, from (24) we obtain a spatially localized solution describing waves of the stationary profile:

$$\psi = \Theta_0 \operatorname{th} \left(\frac{\Theta_0 \sqrt{\alpha'} (y' + C_1)}{2} \right) \exp(-2i\xi t' - iC_2). \quad (25)$$

From the initial and boundary conditions we have $\eta = \text{constant}$ and the constants $C_1 = C_2 = 0$. Substitute them to (25) we obtain:

$$\psi = \Theta_0 \operatorname{th} \left(\frac{\Theta_0 \sqrt{\alpha'} y'}{2} \right) \exp(-2i\xi t'). \quad (26)$$

The relation (26) gives the width of the soliton localization region and its amplitude, but does not describe the motion along the coordinate y . The velocity V of the soliton motion can be found from the following transformation [7]:

$$\psi_0 = \Phi(y' - Vt') \exp[i(V/2)(y' - (V/2)t')]. \quad (27)$$

Then for the function ψ we obtain: (28)

$$\psi = \sqrt{\frac{2}{\alpha'}} \frac{\sqrt{\alpha' \Theta_0^2 - V^2/4}}{\sqrt{1+k^2}} k \operatorname{sn} \left(\frac{\sqrt{\alpha' \Theta_0^2 - V^2/4}}{\sqrt{1+k^2}} \frac{y'}{k}, k \right) \exp[i(V/2)(y' - Vt') - i\omega t']$$

where $\omega = (\xi - V^2/4)$ is the frequency in the reference frame moving with the soliton, and ξ is the frequency in the stationary (laboratory) frame of reference.

From the relation (28) for $k \rightarrow 1$, we obtain a spatially localized solution that describes the wave of a stationary profile moving along the coordinate y with the velocity V : (29)

$$\psi = \sqrt{\frac{\alpha' \Theta_0^2 - V^2/4}{\alpha'}} \operatorname{th} \left(\frac{\sqrt{\alpha' \Theta_0^2 - V^2/4}}{2} (y' - Vt') \right) \exp[i(V/2)(y' - Vt') - i\omega t']$$

Thus, the spatially localized solution of Eq. (19) is the two-parametric soliton, one of the parameters of which is the velocity V of the center of this soliton, and the other parameter is its eigen frequency ω in the reference frame moving with the soliton.

The relationship (29) is represented graphically in Figure 5a.

a

B

Figure 5: The spatially localized solution of equation (19) is a wave of stationary profile moving along the coordinate y (a) and motion of the orientational inflection (b). $E=1$ V/ μ m, $P_S=50$ nC/cm², $M=12 \cdot 10^3$ erg/cm³, $K=5 \cdot 10^{-12}$ N, $\gamma_\psi=0.1$ P, $\Theta_0=23^\circ$, $\varphi_0=30^\circ$.

Taking into account the normalization (20), the soliton motion velocity can be written as:

$$V = 2\Theta_0 \sqrt{\alpha' K} / (\gamma_\psi d) = (\Theta_0 / \gamma_\psi) \sqrt{2K(M + P_s E \cos \phi_0)} \cdot (30)$$

Let $\omega = 0$, that is, the frequency $\xi = V^2 / 4$. Then the reorientation time of the director caused by the orientation inflection motion (Figure 5b), taking into account the normalization (20) has the following form:

$$\tau_c = \frac{\gamma_\psi d^2}{K \xi} = \frac{2\gamma_\psi}{\Theta_0^2 (P_s E \cos \phi_0 + M)} \cdot (31)$$

If we take as in the previous case the initial azimuth angle of the director's orientation $\phi_0 = 30^\circ$, $P_s = 50 \text{ nC} / \text{cm}^2$, $M = 12 \cdot 10^3 \text{ erg} / \text{cm}^3$, $E = 1 \text{ V} / \mu\text{m}$, $\Theta_0 = 22^\circ$, $\gamma_\psi = 0.1 \text{ P}$ (value of the coefficient γ_ψ is defined in p. 4), the elasticity coefficient describing the deformation on the angle ψ , $K = 5 \cdot 10^{-12} \text{ N}$, then the velocity of the soliton center $V = 0.65 \text{ cm} / \text{s}$, and the director reorientation time in the soliton mode τ_c , obtained from the relation (31), is about $70 \mu\text{s}$, which is close to the experimentally observed values (see further).

Compositions of Helix-Free FLCs

For the first time a helix-free FLC was considered in [16]. In the present work five compositions of helix-free FLCs were explored, and spatially non-uniform structures with periodic deformations of smectic layers were observed in the FLC layer. All compositions were differed from each other by the coefficient of the rotational viscosity γ_ϕ , varying in the range from 0.15 to 1.5 Poise (Table 1).

Table 1: Physical parameters of the investigated compositions of helix-free FLCs.

FLC	$\Theta_0, ^\circ$	$P_S, \text{nC} / \text{cm}^2$	γ_P, P
HF-32B	21.7	40	0.7
HF-32C	22	40	1.0
HF-32D	23	42	0.15
HF-32E	22	45	1.5
HF-32F	21	42	0.75

The sequence of phase transitions for the used FLC is as follows:

Here, C_r is the crystalline phase, $Sm-C^*$ is the chiral smectic C phase (ferroelectric), $Sm-A^*$ is the chiral smectic A phase (paraelectric), and I is the isotropic (liquid) phase.

We note that the magnitude of the spontaneous polarization P_S , the tilt angle of molecules in smectic layers Θ_0 , and the temperature range for the ferroelectric phase existence for all used FLC compositions are differed insignificantly. The period of smectic layers deformation for all used FLC compositions, except HF-32F, is $2 \div 3 \mu\text{m}$, and for HF-32F, the pitch is $5 \div 6 \mu\text{m}$. The deformation pitch was measured for electro-optic cells $13 \mu\text{m}$ thick with the homeotropic orientation of the director.

Dynamics of the Reorientation of the Flc Director and the Influence of Viscosity

The transition to the Maxwellian mechanism of energy dissipation means that the viscoelastic tension at the termination

of motion does not retain the value that corresponds to the instantaneous value of elastic deformation (as in the ordinary theory of elasticity), and does not go to zero (as in the theory of a viscous liquid), but decreases to zero gradually, after a certain period of time τ_m - the Maxwellian relaxation time.

In the case when the frequency of the external force change corresponds to the relation $f \sim 1 / \tau_m$, the fluid must undergo a viscous flow in addition to the elastic deformation, that is, a combination of viscous flow and elastic shear deformation takes place. Such properties can be simultaneously characterized by the viscosity coefficient γ and some shear modulus μ . In this intermediate case, the viscosity is related to the shear modulus by the following relationship [3]:

$$\gamma \sim \tau_m \mu. \quad (32)$$

FLC can be considered as a very viscous liquid with a viscosity coefficient γ_ϕ that behaves as a solid state for a sufficiently small time, that is, it is elastically deformed. In this case τ_m is the time during which the shear tensions in FLC acquired as a result of the action of an external electric field disappear after the cessation of this action.

If the field change frequency is $f \sim 1/\tau_m$, then the FLC viscosity is also related to the shear modulus by the relation (32). In this case, if the time τ_m does not depend on the field change frequency, then the dissipative coefficient is the viscosity γ_ϕ , which also does not depend on the frequency. Accordingly, the time of the director reorientation $\tau_R \sim \gamma_\phi / (P_s \cdot E)$ is independent of the frequency. Conversely: the time τ_m depends on the frequency when the viscosity depends on the field change frequency. In this case, the dissipative coefficient is the viscosity at the shear tension γ_ψ .

For the electro-optical cells of the thickness 1.7 μm , where FLC with the rotational viscosity γ_ϕ from 0.7 to 1.0 P was used as a modulating medium, the electro-optical response time $\tau_{0,1-0,9}$ does not depend on the field change frequency if this frequency does not exceeds $70 \div 100$ Hz (Figure 6).

Figure 6: Frequency dependence of the electro-optical response time for the FLC cells 1.7 μm thick with the compositions: curve 1 - HF-32B ($\gamma_\phi = 0.7$ P), curve 2 - HF-32C ($\gamma_\phi = 1.0$ P). Control voltage - pulses of a rectangular shape with the amplitude of ± 1.5 V.

In this frequency range, τ_{rel} – the time of FLC director relaxation to the unperturbed state after the electric field is turned off also does not depend on the field change frequency (Figure 7).

The relaxation time was measured both by the electro-optical channel (Figure 7) and by the dielectric one. When the electro-optical channel was used, the rectangular-shaped voltage pulses (unipolar or bipolar) were applied to cell electrodes, and the pulse duration was equal to a pause between pulses. The relaxation time was measured as the time of disappearance of all the director's orientation distortions after the field was switched off, that is, the time during which the light transmission of the electro-optical cell was returned to a level corresponding to the state without an electric field. By the dielectric channel, the relaxation time was measured as the time during which the surface charge on cell electrodes (proportional to the current integral through a cell) decreased to zero. In both cases, the character of the frequency dependence of the relaxation time did not change.

Figure 7: Frequency dependence of the director relaxation time for the electro-optical cells 1.7 μm thick with the compositions: HF-32B (curve 1), and HF-32C (curve 2). Control voltage - pulses of a rectangular shape with the amplitude of ± 1.5 V.

The transition to the Maxwellian relaxation time occurs when the director relaxation time begins to depend on the frequency of the field change (Figure 7). The change in the energy dissipation channel, that is, the transition to the shear viscosity γ_ψ , is accompanied by a strong frequency dependence of the electro-optical response time $\tau_{0,1-0,9}$ (Figure 6). It's seen that in the frequency range of the control voltage from 100 to 200 Hz, the time $\tau_{0,1-0,9}$ decreases almost twofold.

If the shear viscosity prevails, the director reorientation is due to the motion of soliton waves. The time of reorientation τ_R and, correspondingly, the time of the electro-optical response $\tau_{0,1-0,9}$ are determined by the wave velocity (ratio (30)), and the dependence of the response time on the frequency of the electric field variation is rather weak (Figures 6 and 8).

For the electro-optical cell with the composition HF-32B (the rotational viscosity coefficient $\gamma_\phi = 0.7$ P) as a modulating medium, the maximum modulation frequency of the light was 3.5 kHz at the control voltage amplitude of ± 1.5 Volts (Figure 6, curve 1, and Figure 8b).

The shear viscosity of HF-32B can be estimated using the corresponding dependence $\tau_{0,1-0,9}(f)$. The increase in the electro

optical response time $\tau_{0,1-0,9}$ (Figure 6) is associated with the simultaneous presence of two dissipative coefficients γ_φ and γ_ψ , and the time $\tau_{0,1-0,9}$ increases by no more than 10%. This allows to suppose that the shear viscosity value γ_ψ in the case under consideration does not exceed 0.1 P, because $\gamma_\varphi = 0.7$ P.

a

b

Figure 8: Oscillograms of the control voltage (green color - channel CH3) and electro-optical response (yellow - CH1). The thickness of the electro-optical cell with FLC composition HF-32B is 1.7 μm . Control voltage: meander with the amplitude ± 1.5 V, zero voltage level - the arrow with digit 3. The frequency of the control voltage is 2 kHz (a) and 3.5 kHz (b). The upper level of the electro-optical response is the closed state, the lower one is the transmission state, and zero level of the light transmission is the arrow with digit 1.

The electro optical response of the cell with the more viscous composition HF-32C (rotational viscosity coefficient $\gamma_\varphi = 1.0$ P) does not differ qualitatively from the response of the cell with HF-32B ($\gamma_\varphi = 0.7$ P), but there are quantitative differences.

First, the value of the control voltage frequency, up to which the dissipative coefficient in this material is the rotational viscosity (when the time $\tau_{0,1-0,9}$ does not depend on the frequency), decreased to 50 Hz (Figure 6, curve 2).

Secondly, to both directions the frequency interval has widened, in which the predominant dissipative coefficient is the shear viscosity - from 100 Hz to 7 kHz; accordingly, the maximum light modulation frequency in the soliton mode increased up to 7 kHz (Figure 6, curve 2 and Figure 9 b).

Thirdly, the FLC fast-ability increased because the electro-optical response time $\tau_{0,1-0,9}$ decreased from 80 to 40 μ s at the light modulation frequency of 100 Hz (Figure 6 and Figure 9a).

In addition, in the soliton mode at the maximum modulation frequency of 7 kHz, the time $\tau_{0,1-0,9}$ does not exceed 25 μ s at the control voltage amplitude of ± 1.5 Volts (Figure 9 b). Thus, the use of more viscous FLC composition made it possible to significantly expand the frequency interval of the soliton mode existence, and, as the result, to increase the speed and increase the maximum frequency of modulation of light radiation by the factor of two.

a

b

Figure 9: Oscillograms of the control voltage (green color - channel CH3) and electro-optical response (yellow - CH1) for the FLC composition HF-32C. The electro-optical cell thickness is 1.7 μm . Control voltage: meander with the amplitude of ± 1.5 V, zero voltage level - the arrow with digit 3. The frequency of the control voltage is 100 Hz (a) and 7 kHz (b). The upper level of the electro-optical response is the closed state, the lower one is the transmission state, and the zero level of the light transmission is the arrow with digit 1.

On the other hand, when the rotational viscosity coefficient γ_ϕ achieves 1.5 P (the composition HF-32E), the frequency dependence of the electro-optical response time $\tau_{0,1-0,9}$ weakens significantly, the transition to the Maxwellian energy dissipation mechanism does not occur, and the soliton mode is not observed (Figure 10). Weakening the frequency dependence of the time $\tau_{0,1-0,9}$ and the absence of the soliton mode is the consequence of the fact that the time of the HF-32E director relaxation to the unperturbed state after electric field switching off is practically independent on the field change frequency (Figure 11).

The lifetime of a molecule in the time equilibrium position between two discontinuous changes in the equilibrium orientation is defined [17] as $\tau \sim \tau_0 \exp(W/kT)$, where W is the activation energy, T is the temperature, $\tau_0 \sim 10^{-12}$ s is the mean period of molecular vibrations. Every jump occurs, when an activation energy transmitted to a molecule is sufficient to break its bonds with neighboring molecules and to go into the environment of other molecules. Under the influence of an external force, which retains its direction longer than the time τ ,

a preferential movement of molecules along the action of this force appears that is a fluidity – a property inverse to viscosity.

Figure 10: Frequency dependencies of the electro-optical response time for the FLC composition HF-32E. Curve 1 – the amplitude of bipolar voltage of a rectangular shape (meander) is ± 1.5 V; curve 2 – the voltage amplitude is ± 12 V. The thickness of the electro-optical cell is $1.7 \mu\text{m}$.

Figure 11: Frequency dependence of the director's relaxation time in the FLC composition HF-32E. The electro-optical cell thickness is $1.7 \mu\text{m}$. Control voltage – pulses of a rectangular shape with the amplitude of ± 1.5 V.

The decrease in the coefficient γ_ϕ to 0.15 P (the FLC composition HF-32D) leads to the fact that in weak electric fields (less than $1 \text{ V} / \mu\text{m}$) the mechanism of fluidity begins to predominate, the elastic properties of FLC manifest weakly, and

as a consequence, the soliton mode is not observed (Figure 12, curve 1).

Figure 12: Frequency dependencies of the electro-optical response time for the FLC composition HF-32D. Curve 1 – the amplitude of bipolar voltage of a rectangular shape (meander) is of ± 1.5 V; curve 2 – the voltage amplitude is of ± 9 V. The thickness of the electro-optical cell is $1.7 \mu\text{m}$.

However, when the electric field strength increases to $3 \div 5 \text{ V} / \mu\text{m}$, the shear elastic deformations again prevail, and the soliton mode appears (Figure 12, curve 2). In this case, increasing the time $\tau_{0,1-0,9}$ is due to the simultaneous presence of two dissipative coefficients γ_ϕ and γ_ψ , and the time $\tau_{0,1-0,9}$ increases almost doubling regardless of the electric field strength, whereas for the more viscous FLC (γ_ϕ in the range from 0.5 to 1.0 P), the increase $\tau_{0,1-0,9}$ does not exceed 10% (Figure 6).

Field and Temperature Dependences of the Electro-Optical Response

The transition to the soliton mode (to the Maxwellian mechanism of energy dissipation) can occur not only with an increasing the frequency of the electric field change, but also with increasing the field strength at a fixed frequency. The transition is accompanied by a sharp decrease in the electro-optical response time when a certain threshold value of the field strength is reached (Figure 13). The threshold value of the field

strength decreases with increasing the frequency, and the time $\tau_{0,1-0,9}$ decreases (Figure 13, curve 2).

Figure 13: Field dependences of the electro-optical response time for the cell 1.7 μm thick with the FLC composition HF-32B. The frequency of the control voltage (meander) is 200 Hz (curve 1) and 3 kHz (curve 2).

The pitch of smectic layers deformation for the composition HF-32B is about two micrometers. Such a pitch can be increased to $5 \div 6 \mu\text{m}$ almost without the change of the spontaneous polarization, the angle of inclination of molecules in smectic layers and the rotational viscosity coefficient as one can see from the comparison with parameters of the composition HF-32F (Table 1).

An increase in the pitch of layer deformation results in a significant increase (more than three times) in the threshold value of the electric field, upon which the transition to a soliton mode occurs (Figure 14). Almost immediately after the transition to a soliton mode, a part of the dependence $\tau_{0,1-0,9}(E)$ appears, where the electro-optical response time $\tau_{0,1-0,9}$ does not depend on the electric field strength. This means that the dissipative coefficient – the shear viscosity also ceases to depend on the field strength.

Figure 14: Field dependences of the electro-optical response time for the cell 1.7 μm thick with the FLC composition HF-32F. The control voltage frequency is 200 Hz (curve 1) and 3 kHz (curve 2).

The transition to the Maxwellian mechanism of energy dissipation changes drastically also the character of the temperature dependence of the electro-optical response time $\tau_{0,1-0,9}$ of helix-free FLCs. If the FLC behaves as a viscous liquid (the dissipative coefficient is the rotational viscosity), then the time $\tau_{0,1-0,9}$ decreases continuously with increasing the temperature up to the temperature of the transition into the paraelectric phase Sm A* (Figure 15, curve 1), and total time changing is more than an order of magnitude.

Simultaneous presence of two dissipative coefficients γ_ϕ and γ_ψ weakens the temperature dependence of the electro-optical response time: a temperature interval appears where the time $\tau_{0,1-0,9}$ is almost constant (Figure 15, curve 2). The predominance of shear viscosity in a soliton mode results in weakening of the temperature dependence of the electro-optical response time in a wide temperature range (Figure 15 curve 3).

Figure 15: Temperature dependences of the electro-optical response time for the cell 1.7 μm thick with the FLC composition HF-32B. The amplitude of the control voltage (meander) is ± 1.5 V. The control voltage frequency is 50 Hz (curve 1), 140 Hz (curve 2) and 3.5 kHz (curve 3).

The higher the frequency of light modulation (respectively, and the frequency of the control voltage), the wider the temperature range, in which the time $\tau_{0,1-0,9}$ is almost constant. At the same time, the temperature range, in which the time $\tau_{0,1-0,9}$ is practically constant, expands both to the high and low temperatures. Thus, for the HF-32C FLC at the maximum possible light modulation frequency of 7 kHz, the time $\tau_{0,1-0,9}$ is practically independent on the temperature in the range from 10 to 52 $^{\circ}\text{C}$ (Figure 16).

Figure 16: Temperature dependence of the electro-optical response time for the FLC cell 1.7 μm thick with the FLC composition HF-32C. The control voltage amplitude (meander) is ± 1.5 V. The frequency is 7 kHz.

Hysteresis-Free Modulation Characteristic

A periodic and continuous change in the position of FLC director, and therefore of the main optical axis of the refractive index ellipsoid, along each smectic layer, is the cause of the electrically controlled birefringence of helix-free FLCs, and this birefringence depends on the control voltage frequency (Figure 17). Thus, for the FLC composition HF-32C the birefringence Δn varies from 0.19 to 0.11 in the frequency interval from 80 Hz to 6.5 kHz.

The static section of the dependence $\Delta n(f)$ at low frequencies ($f < 100$ Hz) corresponds to the turnabout of the ellipsoid of refractive indices in all smectic layers by 180° . At the transition to the Maxwellian mechanism of energy dissipation, the additional angle of inclination of the FLC director changes at each point of a smectic layer and, accordingly, the position of the ellipsoid of refractive indices along the layer changes, and this is accompanied by a decrease of the FLC effective birefringence.

Figure 17: The frequency dependence of the birefringence of the FLC composition HF-32C. The electro-optical cell thickness is $1.7 \mu\text{m}$. The amplitude of the bipolar control voltage of a rectangular shape (meander) is $\pm 1.5 \text{ V}$. On the inset: a low-frequency part of this dependence.

The maximal change Δn , i. e. its decrease in 1.7 times in the relation to the molecular value, occurs in a soliton mode at the frequency of the order of $5 \div 6 \text{ kHz}$ (Figure 17). At higher frequencies of the control voltage, the birefringence does not change - the high-frequency static section of $\Delta n(f)$ appears. This corresponds to the maximum light transmission of the electro-optical cell.

A continuous change in the position of FLC director along the smectic layers makes it possible to obtain a practically hysteresis-free dependence of the electro-optical cell transmission on the control voltage amplitude $I(U)$ both at increasing and at decreasing this amplitude (Figure 18).

Dependence $I(U)$ is hysteresis-free for both positive and negative values of the control voltage, if its frequency corresponds to the frequency interval of the soliton mode existence: for the composition HF-32C it is from 100 Hz to 7 kHz. If the frequency of the control voltage does not correspond to this frequency interval, then there is a difference in the light transmission of an electro-optical cell with increasing and decreasing the voltage amplitude.

a

b

Figure 18: The light transmission dependence on the control voltage amplitude (meander) at increasing (curve 2) and decreasing this amplitude (curve 1) at the voltage frequencies 100 Hz (a) and 6 kHz (b). The electro-optical cell thickness of the FLC composition HF-32C is 1.7 μm .

On the other hand, the control voltage frequency must not correspond to the static (low-frequency and high-frequency) sections of the dependence $\Delta n(f)$. The latter circumstance does not make it possible to obtain the hysteresis-free $I(U)$ dependence at the control voltage frequency above 6 kHz, since at higher frequencies the high-frequency static section of the dependence $\Delta n(f)$ begins (Figure 17).

Conclusion

1. The appearance of structurally stable waves of a stationary profile (dynamic solitons) in spatially inhomogeneous structures with periodic deformations of smectic layers in helix-free FLCs is associated with the transition to the Maxwellian mechanism of energy dissipation and is caused by the nonlinear process of interaction of the spontaneous polarization with the alternating electric field.
2. The soliton mode, which arises at the transition to the Maxwellian mechanism of energy dissipation due to the shear viscosity predominance, manifests itself with increasing the frequency of the electric field change if the coefficient of rotational viscosity of the helix-free FLC belongs to the range from 0.15 to 1.0 P.
3. The reorientation of the FLC director is carried out due to the motion of soliton waves, and this provides a frequency of light modulation of the order of 7 kHz at the electric field strength of 1 V / μm (at the control voltage amplitude ± 1.5 Volts), and the electro-optical response time $\tau_{0,1-0,9}$ in this case is about 25 μs .
4. The shear viscosity predominance weakens the temperature dependence of the electro-optical response time $\tau_{0,1-0,9}$ over a wide temperature range (from 10 °C to 52 °C at the modulation frequency of 7 kHz). The higher the frequency of light modulation (respectively, and the frequency of the control voltage), the wider the temperature range in which the time $\tau_{0,1-0,9}$ is almost constant.
5. A continuous change in the position of the director along smectic layers of the helix-free FLCs makes it possible to obtain the hysteresis-free dependence of the light transmission of an electro-optical cell on the control voltage both with increasing and decreasing its amplitude if the voltage frequency belongs to the frequency interval of a soliton mode existence and does not correspond to the static sections of the frequency dependence of the FLC birefringence. Due to this, the maximal frequency of the hysteresis-free modulation characteristic reached 6 kHz.

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