

Review

Helix-Free Ferroelectric Liquid Crystals: Electro Optics and Possible Applications

Alexander Andreev, Tatiana Andreeva, Igor Kompanets * and Nikolay Zalyapin

P. N. Lebedev Physical Institute, Moscow 119991, Russia; ALA-2012@yandex.ru (A.A.); rybusenok@yandex.ru (T.A.); nikolay.zal@gmail.com (N.Z.)

* Correspondence: kompan@sci.lebedev.ru; Tel.: +7-903-124-3235

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Featured Application: As the materials for fast low-voltage displays and light modulators.

Abstract: This is a review of results from studying ferroelectric liquid crystals (FLCs) of a new type developed for fast low-voltage displays and light modulators. These materials are helix-free FLCs, which are characterized by spatially periodic deformation of smectic layers and a small value of spontaneous polarization (less than 50 nC/cm²). The FLC director is reoriented due to the motion of solitons at the transition to the Maxwellian mechanism of energy dissipation. A theoretical model is proposed for describing the FLC deformation and director reorientation. The frequency and field dependences of the optical response time are studied experimentally for modulation of light transmission, scattering, and phase delay with a high rate. The hysteresis-free nature and smooth dependence of the optical response on the external electric field in the frequency range up to 6 kHz is demonstrated, as well as bistable light scattering with memorization of an optical state for a time exceeding the switching time by up to 6 orders of magnitude. Due to the spatially inhomogeneous light phase delay, the ability of a laser beam to cause interference is effectively suppressed. The fastest FLCs under study are compatible with 3D, FLC on Silicon (FLCoS), and Field Sequential Colors (FSC) technologies.

Keywords: liquid crystal; ferroelectric; helix-free; soliton; transparent mode; light scattering; speckle suppression

1. Introduction

It is known that the minimum-time optical response is achieved in some smectic liquid crystals, called smectics C*, which possess ferroelectric properties and high sensitivity to electric fields [1–3]. The principle of electro-optical modulation in ferroelectric liquid crystals (FLCs), like in widely used nematic liquid crystals (NLCs), is the electrically controlled birefringence or light scattering.

A distinctive feature of smectic crystals is the layered structure formed as a result of ordering the centers of mass of FLC molecules along the direction of orientation of their long axis (director), with a pitch of the order of the molecules' length. Among the smectic crystals, the helix FLCs are the most well known, in which the polar axes of various smectic layers are rotated relative to each other, forming a helix (spiral) twist of the FLC director in the absence of an external electric field (Figure 1). In each layer, the position of the director \mathbf{n} is determined by the polar angle Θ_0 and the azimuth angle φ , which varies from 0 to 2π at a distance equal to the pitch p_0 of the helix. Under the action of the electric field E , which is parallel to smectic layers (along the coordinate x), the vector P_s of spontaneous polarization is oriented in all layers along the field direction. As a result, the director acquires one direction in the entire volume of the FLC layer—the direction of the FLC main optical axis.

When a sign of the field E changes, the orientation of the vector P_s changes by 180° , and the long axes of molecules unfold along the cone with the generatrix $2\Theta_0$, resulting in a change of the angle φ by 180° . The reorientation of the director determining the main optical axis of the ellipsoid of the FLC refractive indices results in a change in the angle between the polarization plane of the incident light (I_0) and the main optical axis of the ellipsoid. This leads to the modulation of the phase delay between the ordinary and extraordinary rays, or to light intensity modulation, if the electro-optic cell is placed between crossed polarizers [1–3]. Under certain conditions, electrically controlled light scattering on so-called transient domains is observed in the helix FLC due to the formation of a spatially inhomogeneous structure of refractive index gradients in the FLC layer [4,5].

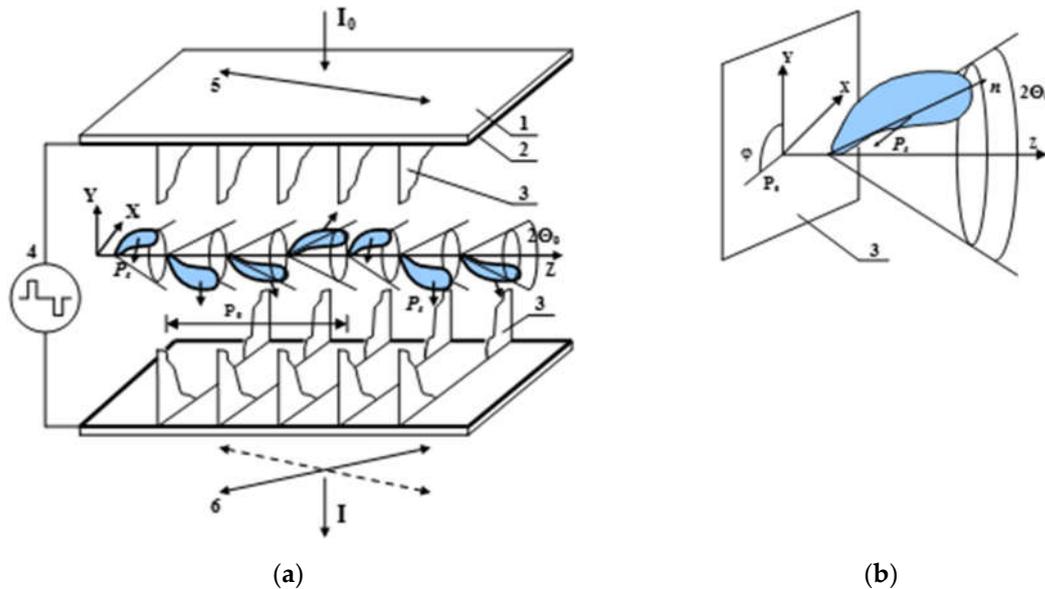


Figure 1. The helix ferroelectric liquid crystal (FLC)-based electro-optical cell with planar orientation of a layer (a) and mutual location of the spontaneous polarization vector P_s , a smectic layer, and FLC director n (b). I_0 and I are the intensities of the incident light and the light passing through an FLC cell, correspondingly. 1—glass substrates with conductive covers; 2; 3—smectic layers; 4—generator of bipolar pulses; 5 and 6—polarizers; p_0 —helix pitch; Θ_0 —angle of molecule tilt in smectic layers.

Helix-free (no spiral) FLCs are also known [6]. In them, the helicoidal twist of the director in the FLC volume is compensated or suppressed by the interaction of chiral optically active additives with opposite signs of optical activity. The coincidence of the same signs of spontaneous polarization in chiral additives makes it possible to obtain a value of spontaneous polarization of 100 nC/cm^2 or higher for FLCs with a compensated helicoid.

In contrast to NLC, the electro-optical effect in FLC is linear relative to the field [7,8], and since FLC reacts to the sign of the applied electric voltage, the values of the on and off times of the optical response are the same and are proportional to

$$\tau_R \sim \gamma_\varphi / P_s \times E, \tag{1}$$

if the dissipative coefficient is the rotational viscosity γ_φ . The FLC is returned to its original state by a reverse-polarity pulse, i.e., forcedly, not as a result of relaxation due to elastic forces (like in the NLC). Therefore, the optical response at switching on and off is symmetrical and very short (in the sub-millisecond region), especially for FLC with low viscosity and large spontaneous polarization. However, it is impossible to significantly reduce τ_R by increasing the spontaneous polarization, since this usually leads to an increase in the FLC rotational viscosity. This circumstance limits the frequency of light modulation if the applied voltage does not exceed several volts.

In the research carried out at the Lebedev Physical Institute (LPI), it was shown that the nature of the FLC director reorientation in an electric field depends essentially on the coefficient responsible for the energy dissipation in an FLC layer: the rotational or shear viscosity [9]. If the alternating

electric field of the frequency f acts on the FLC, and the period of its variation is large ($\tau_m \cdot f \ll 1$) in comparison with the Maxwellian relaxation time τ_m [10], then the FLC behaves as a liquid with the viscosity γ_φ . On the contrary, at sufficiently high frequencies ($\tau_m \cdot f \gg 1$), the FLC behaves as an amorphous solid, and the dissipative coefficient is the shear viscosity (denoted γ_ψ).

The predominance of shear viscosity leads to a change in the character of the motion of the helix FLC director in weak electric fields: the reorientation occurs due to the motion of 180°-domain walls [9,11,12]. Such a reorientation process made it possible to obtain a modulation frequency of light radiation of the order of 3 kHz with an electric field strength of about 1 V/ μm . The time of the electro-optical response was 50 ÷ 70 μs . As a disadvantage, some distortion of the spectral composition of the modulated radiation and the residual light scattering caused by the presence of a helix was noted.

The character of the electric field action is practically the same for known helix-free FLCs. In them, the azimuth angle φ of the director's orientation in the volume of a layer is a value that is practically constant in all smectic layers; i.e., there is a so-called spatially homogeneous structure. The magnitude of the spontaneous polarization is much higher than 50 nC/cm². The dissipative coefficient is the rotational viscosity γ_φ , and the director reorientation time, as for the helix FLC, does not depend on the frequency of the electric field change [8].

A new type of helix-free FLC specially developed at the LPI has a different character of interaction with the electric field. This material is a spatially inhomogeneous structure with periodic deformation of smectic layers (with a pitch from 1.5 to 5 μm) in the absence of an electric field and a relatively small value of spontaneous polarization (less than 50 nC/cm²). In such an FLC, the director reorientation in the alternating electric field is due to the motion of structurally stable localized waves of a stationary profile—dynamic solitons that arise upon the transition to the Maxwellian mechanism of energy dissipation—and the electro-optical response depends essentially on the frequency of the electric field change [13–16]. Note that a director reorientation through solitons was proposed for the first time in [17].

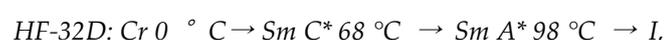
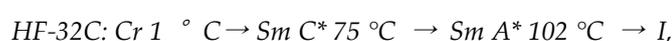
The new materials provided unique parameters of radiation modulation unattainable for LC analogs. For example, in a transparent mode, in an electric field of the order of 1 V/ μm (at a control voltage of ± 1.5 V), experimental samples of electro-optical cells with the novel helix-free FLC show a modulation characteristic with the fastest optical response (about 25 microseconds) and the highest modulation frequency (up to 7 kHz), including hysteresis-free characteristics with a continuous gray scale up to 6 kHz. In a bistable light-scattering mode, a state with intensive scattering can be turned on and off for a few tens of microseconds and be memorized for several tens of seconds or until a pulse of opposite polarity appears [16].

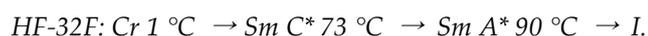
We will now consider in more detail the physical properties, light modulation characteristics, and possible applications of the new helix-free FLC based on the results attained from our research during the last few years.

2. Ferroelectric Liquid Crystals Compositions Used and Their Basic Properties

The basis of the new FLCs is the same optically active additives used in known FLC and based on derivatives of terphenyl-dicarboxylic acid. The difference is only in the specific compositions of FLCs, which is the object of know-how. In spite of the great difference in the rotational viscosity coefficient γ_φ , which in various compositions changes by practically an order of magnitude (from 0.15 to 1.5 P), and in spite of the manifestation of essentially different optical properties (to be discussed below), the values of the spontaneous polarization P_s , the initial tilt angle of molecules in smectic layers Θ_0 , and the sequence of phase transitions for all new FLCs differ insignificantly (see Table 1).

The sequence of phase transitions for these compositions is the following:





Here, *Cr* 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.

Table 1. Investigated helix-free ferroelectric liquid crystals (FLC) compositions.

Compositions	Θ_0 , grad.	P_s , nC/cm ²	γ_{φ} , 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 new helix-free FLC materials exhibit the following basic properties in electro-optical cells, first discovered and investigated at the LPI:

- A periodic spatial deformation of FLC smectic layers with a pitch of $1.5\div 6\text{ }\mu\text{m}$ is observed in the absence of an electric field;
- The FLC director is reoriented as a result of the soliton waves motion;
- Certain FLC compositions show the fastest (among all LCs) optical response in a transparent mode with continuous gray scale;
- Certain FLC compositions show the fastest (among all LCs) optical response in the light-scattering mode with data storage;
- Certain FLC compositions provide a rapid change in the phase delay of the modulated radiation initiated by light scattering.

3. Periodic Deformation of Smectic Layers

The periodic deformation of smectic layers in new helix-free FLCs in the absence of an external electric field is the result of compensation of the space charge of the spontaneous polarization. In the case of homeotropic orientation of FLC molecules, where smectic layers are parallel to the electro-optical cell substrates, the periodic deformations are clearly observed (Figure 2) and look behind crossed polarizers like alternating bands with a pitch of $1.5\div 6\text{ }\mu\text{m}$, depending on the FLC molecular structure [15,16].

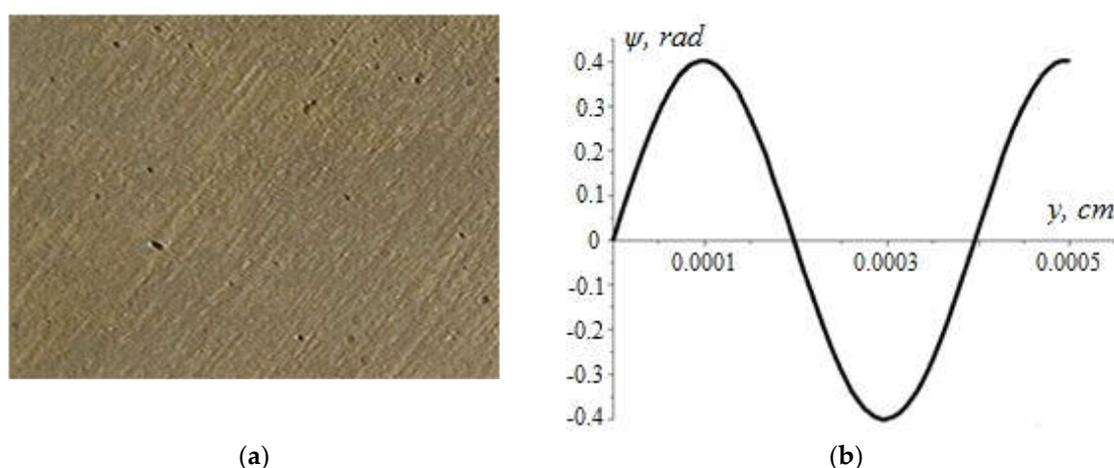


Figure 2. Alternation of light and dark bands, illustrating the spatially periodic deformation of smectic layers: (a) observed experimentally and (b) calculated (see Section 5).

A deformation occurs at a certain ratio between the values of FLC essential parameters, as follows:

- The rotational viscosity is in the range of $0.3 < \gamma_\varphi < 1.0$ P. If it is less, the transition to the shear viscosity γ_ψ is not achieved, and the soliton mechanism of reorientation of the FLC director is not realized; if it is more than 1 P, the optical response time increases significantly at higher frequencies, and the transition to the soliton mode is not observed;
- The magnitude of the spontaneous polarization P_s is less than 50 nC/cm². If it is greater, the saturation voltage increases, the ferroelectric domains begin to form, and residual light scattering takes place after the electric field is turned off.

The periodical deformation of smectic layers is responsible for the properties of the novel FLCs indicated above. In fact, the fast electro-optical response with continuous gray scale in a transparent mode, which could be used in display devices, is observed in the FLC compositions with a rather long deformation pitch of more than 3 μm . For example, for the composition HF32F, the deformation pitch is of 4 μm . Intensive light scattering in FLC, which could be used in polarization-free devices, manifests in compositions with rather short deformation pitch of $1.5 \div 2.0$ μm (this value for HF32 is about 1.5 μm). A fast-changing phase delay (initiated by light scattering switching on), which could be used for suppressing speckles in laser images, is realized more preferably in FLC compositions with a deformation period of about 3 μm . Although the periods of spatial deformation for HF32F and HF32B are different, other essential parameters of these compositions are almost the same (see Table 1).

4. Ferroelectric Liquid Crystals Director Reorientation by an Electric Field in the Soliton Mode

As shown in [15,16], the periodic deformation of smectic layers means that for FLC molecules initially inclined at the angle Θ_0 with respect to the normal to the FLC layer at a given point, it is energetically preferable to additionally deflect by some angle $\pm\Psi$ with respect to the direction z of substrate rubbing (Figure 3). Because of this, the position of the FLC main optical axis along the smectic layers changes, and the birefringence depends on the electric field frequency.

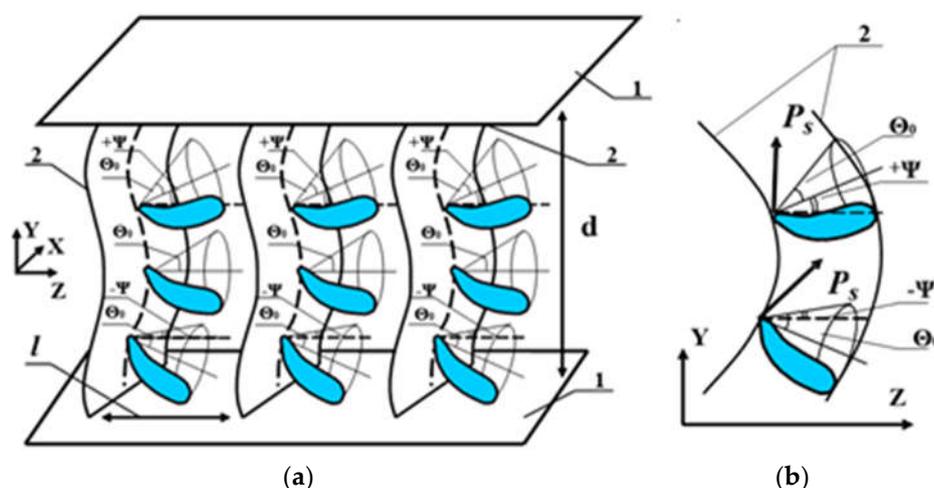


Figure 3. An electro-optical cell with helix-free FLC (a) and a fragment of a deformed smectic layer (b) 1—glass substrates with conductive covers; 2—smectic layers; Θ_0 —the angle of molecule tilt in smectic layers; ψ —angle of tilt of a smectic layer; P_s —vector of spontaneous polarization; d —FLC cell thickness; l —smectic layer thickness.

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

When after the electric field is turned off the director relaxation time begins to depend on the electric field frequency, the transition to the Maxwellian mechanism of energy dissipation takes place—that is, the transition from the rotation viscosity γ_φ to the shear viscosity γ_ψ .

To describe the nonlinear dynamic process of FLC director reorientation in an external electric field, we used the soliton approach [15–18] by which many nonlinear effects and processes have been described before, including those in ferroelectric liquid crystals [17, 19–21].

The soliton mode manifests in the new FLC compositions possessing coefficients of rotational viscosity in the range from 0.15 to 1 P, when the electric field frequency or, simultaneously, the frequency and the strength increase. At frequencies corresponding to the transition to the Maxwellian mechanism of energy dissipation, the viscosity γ_ψ for the shear strain becomes a dissipative coefficient, and the dynamic solitons move along the smectic layers. The director of the FLC is reoriented by the motion of soliton waves.

The appearance of the soliton waves is due to the joint influence of the medium nonlinearity and the presence of a dispersion of velocities of deformation waves (or displacements). They sharply increase the steepness of the wave front and cause the growth of gradients of the field variables. This leads to a spatial redistribution of the excitation energy and its localization (the wave period tends to infinity). The shape of dynamic solitons is uniquely related to the independent parameters—in particular, to the velocity of the soliton center [18]:

$$V = \frac{\Theta_0}{\gamma_\psi} \left(2K(P_s E \cos \varphi_0 + M) - \left(\frac{2K}{d\Theta_0} \right)^2 \right)^{1/2}, \quad (2)$$

where K is the FLC elastic modulus describing the deformation of the director with respect to the angle Ψ ; γ_ψ is the shear viscosity; M is the bending energy of smectic layers; and φ_0 is the initial azimuth angle of the director orientation.

The transition to the Maxwellian mechanism of energy dissipation and the soliton mechanism of director reorientation is accompanied by a strong frequency dependence of the electro-optical response time $\tau_{0.1-0.9}$. It can be seen from Figure 4 that in the frequency range from 100 to 200 Hz, this time for the composition HF-32B with $\gamma_\varphi = 0.7$ P decreases by almost half.

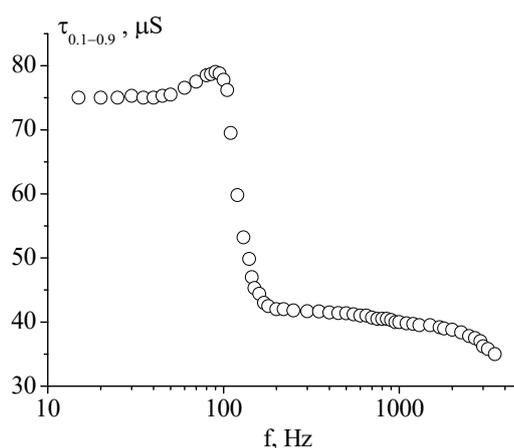


Figure 4. Frequency dependence of the optical response time for the HF-32B composition at $V = \pm 1.5$ V. The bipolar voltage is of the rectangular shape (meander). The thickness of the electro-optical cell is $1.7 \mu\text{m}$.

For more viscous FLC compositions, for example, for HF-32E with $\gamma_\varphi = 1.5$ Poise, the frequency dependence of the electro-optical response time $\tau_{0.1-0.9}$ is significantly weakened (Figure 5a). The reason for this weakening is that the relaxation time of the FLC director to the unperturbed state after the voltage turning off is practically independent of the frequency of the electric field change (Figure

5b). Consequently, the transition to the Maxwellian mechanism of energy dissipation does not occur, and the soliton mode does not manifest.

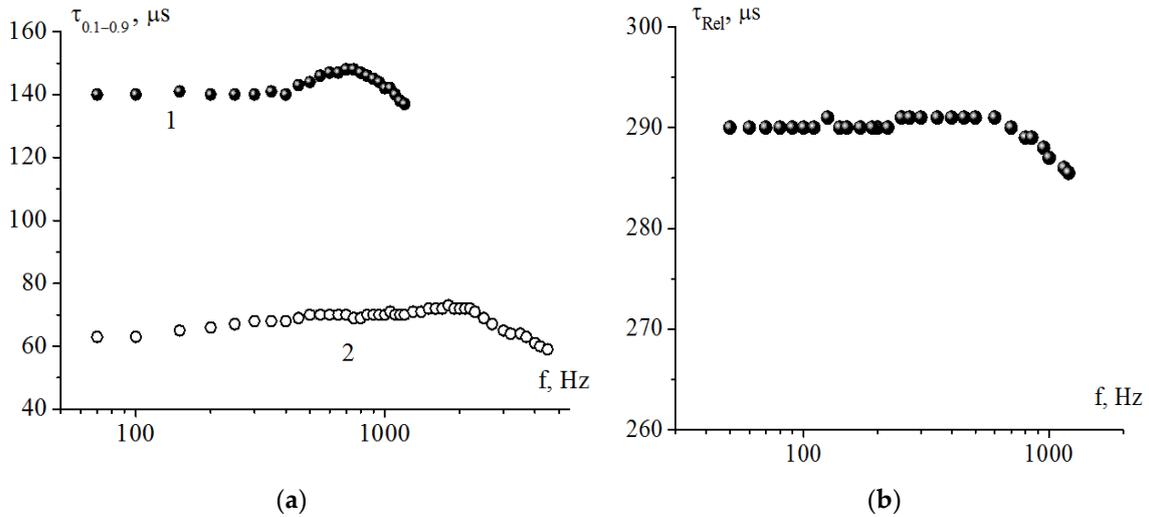


Figure 5. Frequency dependences (a) of the optical response time at ± 1.5 V (curve 1) and ± 12 V (curve 2) pulses acting and (b) of the director relaxation time after ± 1.5 V pulses turning off. The FLC composition is HF32E ($\gamma_\varphi = 1.5$ P). A bipolar voltage of rectangular shape (meander) was applied. The thickness of the electro-optical cell is $1.7 \mu\text{m}$.

A decrease in the rotational viscosity coefficient to 0.15 Poise (for the composition HF-32D with practically the same spontaneous polarization value) leads to the soliton mode not being observed at the control voltage amplitude ± 1.5 V (Figure 6, curve 1). The soliton mode appears when the electric field strength exceeds $5 \text{ V}/\mu\text{m}$ (Figure 6, curve 2). This is especially seen in the frequency range of a few hundred Hz, when the transition to the mechanism of energy dissipation takes place.

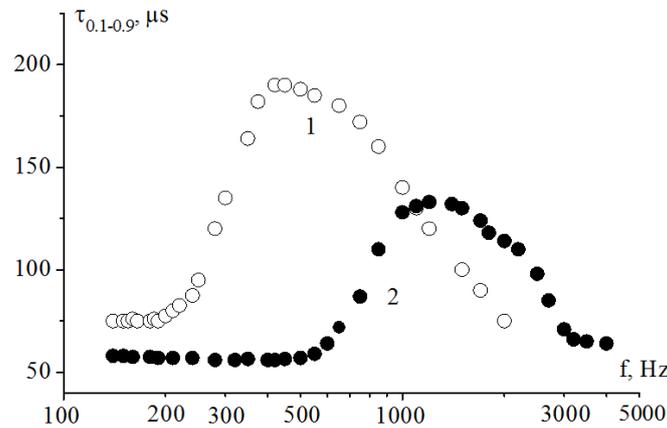


Figure 6. Frequency dependences of the electro-optical response time for the HF32D composition ($\gamma_\varphi = 0.15$ Poise). 1—amplitude of the bipolar voltage (meander) ± 1.5 V; 2—amplitude ± 9 V. The thickness of the electro-optical cell is $1.7 \mu\text{m}$.

5. Theoretical Model of the Ferroelectric Liquid Crystal Deformation and Director Reorientation

5.1. Deformation of Ferroelectric Liquid Crystal Smectic Layers

The bulk density of free energy associated with the periodic deformation of smectic layers can be written as [16,18,22]

$$F = \frac{1}{2}K\left(\frac{d\psi}{dy}\right)^2 + \frac{1}{2}M\left(\frac{l-l_0}{l_0}\right)^2. \tag{3}$$

Here, K is the elasticity coefficient describing the director’s deformation over the angle ψ (Figure 3) and M is the bending energy of smectic layers. We also take into account the smallness of the angles ψ and Θ_0 ($\psi, \Theta_0 \ll 1$), and the relative change of the smectic layer thickness $(l - l_0)/l_0 \cong (\psi^2 - \Theta_0^2)/2$ [23].

If to minimize Equation (3) we can find the distribution of the angle ψ along the coordinate y ($0 \leq y \leq d$, where d is the thickness of the electro-optical cell), then

$$\frac{d^2\psi}{dy^2} + \frac{M\Theta_0^2 d^2}{2K}\psi - \frac{Md^2}{2K}\psi^3 = 0, \tag{4}$$

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

The exact solution of Equation (4) is written in terms of the Jacobi elliptic sine:

$$\psi = \sqrt{2}\Theta_0 \frac{k}{\sqrt{1+k^2}} \operatorname{sn}\left(\Theta_0 d \sqrt{\frac{M}{2K}} \frac{(y'+C_1)}{\sqrt{1+k^2}}, k\right) \tag{5}$$

where $k = \sqrt{\frac{M\Theta_0^4 d^2}{8KC_2} - \frac{M\Theta_0^4 d^2}{8KC_2} - 1}$, $0 < C_2 < \frac{M\Theta_0^4 d^2}{8K}$, and $0 < k < 1$ is the modulus of Jacobi elliptic sine.

Using the Van der Pol method [24] and substituting the boundary conditions $\psi(y = 0) = 0$ and $d\psi/dy = 0$ at $\psi = \Theta_0$, we obtain an approximate solution to Equation (4):

$$\psi = \Theta_0 \sin\left(\frac{5}{8}\Theta_0 \sqrt{\frac{M}{2K}} y\right). \tag{6}$$

This analytical solution given by Equation (6) describes the structure of smectic layers deformed periodically in the direction y . Maxima and minima in the dependence $\psi(y)$ correspond to light and dark stripes in Figure 2a for a cell with FLC layer thickness of $5 \mu\text{m}$, $\Theta_0 = 23^\circ$, $M = 4 \times 10^3 \text{ erg/cm}^3$, and $K = 5 \times 10^{-12} \text{ N}$.

Reorientation of the FLC director due to the interaction of the alternating electric field E applied along the coordinate y (Figure 2b) with the spontaneous polarization P_s can occur both by changing the azimuth angle φ of the director orientation by 180° (if the director is reoriented along the generating lines of a cone with apex angle $2\Theta_0$) and by changing the distribution of ψ (deformation of smectic layers). In the first case, the dissipative coefficient is the rotational viscosity γ_φ , and in the second one, the viscosity γ_ψ of the shear deformation predominates [16,18].

5.2. Ferroelectric Liquid Crystal Director Reorientation

The bulk density of the free energy for the electrostatic interaction of the electric field and spontaneous polarization can be written as

$$\frac{1}{2}P_s E \cos\varphi_0 \left(\frac{l-l_0}{l_0}\right)^2, \tag{7}$$

where φ_0 is the initial azimuth angle of the director orientation. Now we minimize the expression in Equation (3) and record the equation of a balance of moments, describing the change ψ in the electric field E :

$$-\gamma_\psi \frac{\partial\psi}{\partial t} = K \frac{\partial^2\psi}{\partial y^2} + \frac{(P_s E \cos\varphi_0 + M)}{2}\psi(\Theta_0^2 - \psi^2) \tag{8}$$

where $K \partial^2 \psi / \partial y^2$ and $\gamma_\psi \partial \psi / \partial t$ are the elastic and viscous moments defining the director reorientation by the angle ψ ; K and γ_ψ are elasticity and viscosity coefficients; t is the time; and the electric field switches on at $t = 0$.

If we introduce variables

$$\alpha = \frac{d^2(P_s E \cos \varphi_0 + M)}{2K}, \quad y' = \frac{y}{d}, \quad t' = \frac{t}{t_0}, \quad \text{and} \quad t_0 = \frac{\gamma_\psi d^2}{K}, \tag{9}$$

then Equation (8) will be transformed to

$$\frac{\partial \psi}{\partial t'} + \frac{\partial^2 \psi}{\partial y'^2} + \alpha \Theta_0^2 \psi - \alpha \psi^3 = 0. \tag{10}$$

Since ψ in a general case can depend not only on the coordinates but also on the time, we have $\psi = \psi(y', t') = \psi_0 \exp(-i\xi t')$, where $\xi > 0$ is a constant, and ψ_0 is the amplitude, which is a slow function of time [24]. Thus, Equation (8) is converted to the following:

$$\frac{\partial \psi_0}{\partial t'} + \frac{\partial^2 \psi_0}{\partial y'^2} + \psi_0(\alpha \Theta_0^2 + i\xi) - \alpha \psi_0^3 = 0. \tag{11}$$

After substitution of $\psi_0 = \Phi \exp(-i\eta)$, where $\Phi = \Phi(y')$ and $\eta = \eta(t')$, Equation (11) is transformed to a system of two equations:

$$\begin{cases} \partial^2 \Phi / \partial y'^2 + \Phi \alpha \Theta_0^2 - \alpha \Phi^3 = 0, \\ \Phi(\partial \eta / \partial t') - \Phi \xi = 0. \end{cases} \tag{12}$$

Using the hyperbolic function sn , one can describe Φ as follows:

$$\Phi = \frac{\sqrt{2} \Theta_0 k}{\sqrt{1 + k^2}} sn \left[\frac{\Theta_0 \sqrt{\alpha} (y' + C_1)}{\sqrt{1 + k^2}}, k \right], \quad \text{and} \quad \eta = \xi t' + C_2. \tag{13}$$

Now the function $\psi = \psi_0 \exp(-i\xi t')$ using sn can be written as follows:

$$\psi = \frac{\sqrt{2} \Theta_0 k}{\sqrt{1 + k^2}} sn \left[\frac{\Theta_0 \sqrt{\alpha} (y' + C_1)}{\sqrt{1 + k^2}}, k \right] \exp(-2i\xi t' - iC_2). \tag{14}$$

For the extremely nonlinear situation at $k \rightarrow 1$ when the wave period tends to infinity, this relation (using the hyperbolic function th) results in the spatially localized waves of a stationary profile—solitons.

$$\psi = \Theta_0 th \left(\frac{\Theta_0 \sqrt{\alpha} (y' + C_1)}{2} \right) \exp(-2i\xi t' - iC_2) \tag{15}$$

From the boundary and initial conditions, we take $C_1 = C_2 = 0$ and $\eta = \text{const}$. Then,

$$\psi = \Theta_0 th \left(\frac{\Theta_0 \sqrt{\alpha} y'}{2} \right) \exp(-2i\xi t'). \tag{16}$$

This equation defines the width of the soliton localization region and its amplitude. However, it does not describe the soliton movement along the coordinate y , namely, its velocity V . This is defined through the transformation

$$\psi_0 = \Phi(y' - Vt') \exp[i(V/2)(y' - (V/2)t')]. \tag{17}$$

Then, the function ψ is

$$\psi = \sqrt{\frac{2}{\alpha}} \frac{\sqrt{\alpha\Theta^2 - V^2/4}k}{\sqrt{1+k^2}} \operatorname{sn} \left[\frac{\sqrt{\alpha\Theta^2 - V^2/4}y'}{\sqrt{1+k^2}}, k \right] \exp \left(i \frac{V}{2}(y'-Vt') - i\alpha t' \right), \quad (18)$$

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

From Equation (18) at $k \rightarrow 1$, we finally obtain a spatially localized solution—the wave of the stationary profile moving with velocity V along the coordinate y :

$$\psi = \sqrt{\frac{\alpha\Theta^2 - V^2/4}{\alpha}} \operatorname{th} \left[\sqrt{\frac{\alpha\Theta^2 - V^2/4}{2}}(y'-Vt') \right] \exp \left(i \frac{V}{2}(y'-Vt') - i\alpha t' \right). \quad (19)$$

So, the spatially localized solution of Equation (8) is a two-parameter soliton: the first parameter is the velocity V of its center motion, and the second one is its eigenfrequency ω in the system of reference moving with the soliton. Such a solution is presented in graphical form in Figure 7.

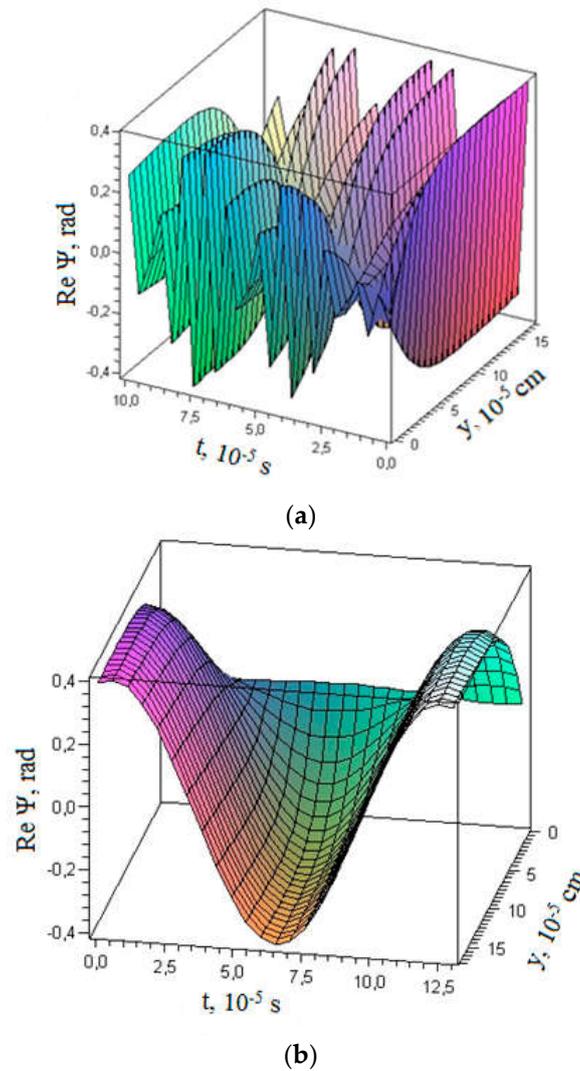


Figure 7. Graphical presentation of the solution of Equation (18), which describes the change of the angle ψ in the electric field E (a) and motion of the orientation bend in the FLC layer (b). $E = 3 \text{ V}/\mu\text{m}$, $P_s = 50 \text{ nC}/\text{cm}^2$, $M = 4 \times 10^3 \text{ erg}/\text{cm}^3$, $K = 5 \times 10^{-12} \text{ N}$, $\gamma_\psi = 0.2 \text{ P}$, $\Theta_0 = 23^\circ$, and $\varphi_0 = 30^\circ$.

As seen from Figure 7, the transition to the Maxwellian mechanism of the energy dissipation when the dissipative coefficient is the shear viscosity γ_ψ results in the appearance of a soliton, which is a wave packet with a periodic wave localized therein. The maximum speed of soliton motion found from Equations (9) and (19) is

$$V = 2\Theta_0 \sqrt{\alpha K / (\gamma_\psi d)} = (\Theta_0 / \gamma_\psi) \sqrt{2K(M + P_s E \cos \varphi_0)}, \quad (20)$$

and the time of director reorientation due to the movement of the orientation bend (Figure 7b) is

$$\tau_c = \frac{\gamma_\psi d^2}{K \xi} = \frac{2\gamma_\psi}{\Theta_0^2 (P_s E \cos \varphi_0 + M)}. \quad (21)$$

If $\varphi_0 = 30^\circ$, $P_s = 50 \text{ nC/cm}^2$, $M = 4 \times 10^3 \text{ erg/cm}^3$, $K = 5 \times 10^{-12} \text{ N}$, $E = 3 \text{ V}/\mu\text{m}$, $\Theta_0 = 23^\circ$, and $\gamma_\psi = 0.2 \text{ P}$, then the speed of the soliton center motion is $V = 0.65 \text{ cm/s}$, and the director reorientation time τ_c is of about $\approx 150 \mu\text{s}$.

6. Light Modulation in an Electro-Optic Cell with Helix-Free Ferroelectric Liquid Crystal (Experimental Results)

6.1. Modulation of Light Transmission

The modulation of light transmission was observed when an electro-optic cell of $1.7 \mu\text{m}$ thickness (achromatic for visible light) was placed between crossed polarizers and bipolar voltage of the rectangular shape (meander) was applied. The FLC director reorientation due to motion of solitons made it possible to reduce the optical response time in weak fields to $25 \mu\text{s}$ and to reach the light modulation frequency of 7 kHz at the control voltage of $\pm 1.5 \text{ V}$ [3] (Figure 8).

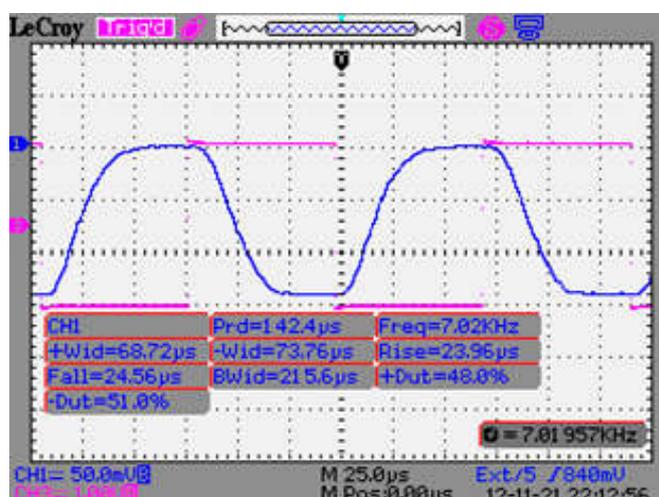


Figure 8. Oscillogram of the bipolar control voltage (zero level—digit 3) and optical response (zero level—digit 1) for an electro-optic cell with composition HF-32C at the voltage amplitude of $\pm 1.5 \text{ V}$ and frequency of 7.0 kHz . The upper level of the optical response is the closed state; the lower one is the light transmission state.

The transition to the soliton mode can occur not only with increasing frequency of the electric field change, but also with increasing field strength at fixed frequency. This transition is accompanied by a sharp decrease in the optical response time when a certain threshold value of the field strength is reached (Figure 9). With increasing frequency, the threshold value of the field for the transition to the soliton mode decreases, and the time of the optical response $\tau_{0.1-0.9}$ decreases (Figure 9, curve 2) at a lower frequency.

After the transition to the soliton mode, a section in the dependence $\tau_{0.1-0.9}(E)$ appears where the optical response time is independent of the electric field strength (Figure 9). This means that the FLC dissipative coefficient is the shear viscosity.

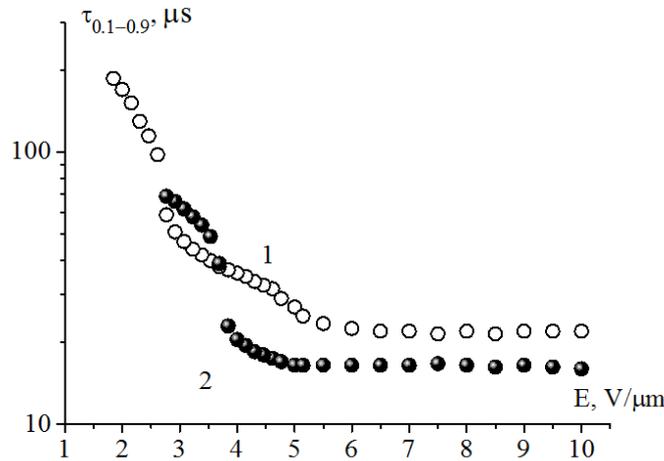


Figure 9. Field dependences of the optical response time for a cell with the HF32F composition. The thickness of the electro-optical cell is 1.7 μm . The voltage frequencies (meander) are 200 Hz (curve 1) and 3 kHz (curve 2).

Constant change in the director position along the FLC smectic layers provides the substantially hysteresis-free dependence of the light transmission of the electro-optical cell on the control voltage amplitude when both increasing and decreasing in a wide frequency range. Experiments have shown that the hysteresis-free modulation characteristic $I(V)$ for both positive and negative voltage occurs if the following two conditions are satisfied: First, the frequency control voltage corresponds to the frequency interval of existence of the soliton mode (for the HF-32C composition, it extends from 100 Hz to 7 kHz). Second, this frequency does not correspond to the static portions (Figure 10) of the frequency dependence of the FLC birefringence $\Delta n(f)$. This means that the frequency of the hysteresis-free modulation does not exceed 6 kHz (Figure 11). This maximum value was realized experimentally [25].

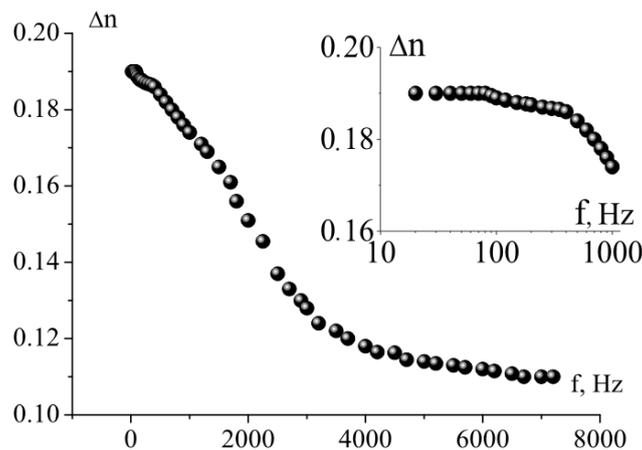


Figure 10. Frequency dependence of the birefringence of the HF-32C composition. Inset: a low-frequency part of this dependence. The thickness of the electro-optical cell is 1.7 μm . The amplitude of the bipolar control voltage (meander) is ± 1.5 V.

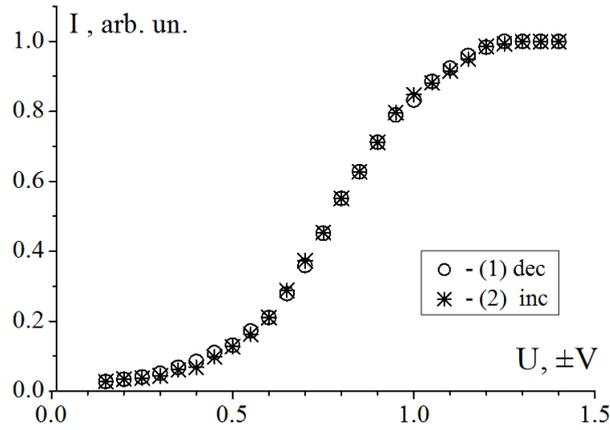


Figure 11. Dependence of the light transmission on the electrical voltage for the frequency 6 kHz at decreasing (1) and increasing (2) voltage. The thickness of the electro-optical cell with the HF-32C composition is 1.7 μm .

Note that the halftone modulation characteristic in such a uniquely wide frequency range is realized only on the basis of the physical properties of new FLC materials. In addition, unlike the bistable (two-level) characteristic of the known FLCs, possible applications of novel helix-free FLC do not require additional electronic modulation of a signal, which reduces the frame rate of color image formation.

It is important that the predominance of shear viscosity in a soliton mode results in weakening the temperature dependence of the electro-optical response time in a wide temperature interval (Figure 12). The higher the frequency of light modulation (and the frequency of the control voltage), the wider the temperature interval in which the time $\tau_{0.1-0.9}$ is almost constant. It shifts to both high and low temperatures, and for the HF-32C composition, the time $\tau_{0.1-0.9}$ at the maximum possible light modulation frequency of 7 kHz is from 10 to 52 $^{\circ}\text{C}$.

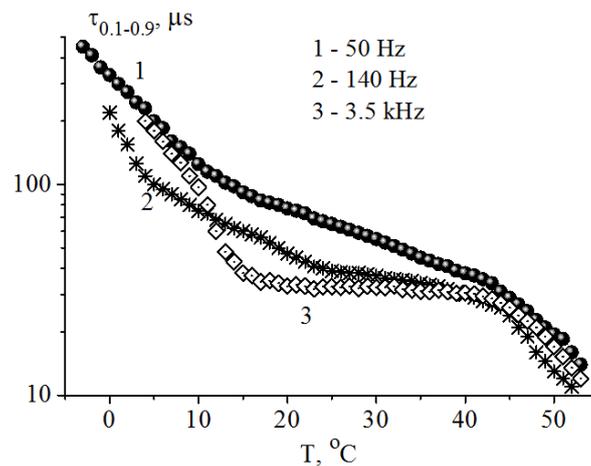


Figure 12. Temperature dependences of the electro-optical response time for the 1.7 μm thick FLC cell with the composition HF-32B. The amplitude of the control voltage (meander) is ± 1.5 V. The control voltage frequencies are 50 Hz (curve 1), 140 Hz (curve 2), and 3.5 kHz (curve 3).

The experimental results show that new helix-free FLCs are very promising materials for the next generation of LC displays, especially for fast displays using FLC on Silicon and 3D technologies, as well as the progressive Field Sequential Colors (FSC) technique, which allows us to form brighter images (because of the absence of RGB filters) by a display with a 3-times-smaller number of pixels [26,27].

6.2. Modulation of Light Scattering

The deformation of a single-domain structure of the FLC caused by a pulse of the electric field can be accompanied under certain conditions by a short “flash” of light scattering. Such scattering was observed for the first time in 1984 and was called “transient” [4], but it was considered parasitic and was not studied in detail. Intense transient scattering in the helix-free FLC was first studied in [30], where it was also proposed to be used in high-speed modulators of a three-dimensional display with a volumetric screen (volumetric display).

Transient light scattering in new helix-free FLCs, including bistable scattering, was studied in detail in [5, 16]. This scattering occurs on the boundaries of spontaneously ordered regions which are formed in the process of the appearance of waves of a stationary profile, i.e., solitons. Scattering occurs after changing the electric field sign and disappears when the motion of solitons reorients the director in all smectic layers—that is, a new homogeneous structure of the FLC layer is no longer formed. A change in the electric field direction induces transient domain formation again, and the process is repeated.

The frequency dependence of the optical response time for light scattering is similar to that described in Section 4. The transition to the Maxwellian mechanism of energy dissipation is accompanied by a strong frequency dependence of the response time $\tau_{0.1-0.9}$ similar to that shown in Figure 4. Some increase in the time $\tau_{0.1-0.9}$ is due to the simultaneous presence of two dissipative coefficients γ_φ and γ_ψ . After the transition to the soliton mode, the time $\tau_{0.1-0.9}$ is determined by the velocity of soliton wave motion, so the frequency dependence of the response time is practically absent.

During operation in a light-scattering mode, the polarizers are not required, and this increases the light transmission of the electro-optical cell by up to 80%. Basically, it is limited by the transparency of conductive coatings on glass substrates. If the pulse duration is less than the minimum time required for a complete disappearance of the transition domains, the light transmission of the cell decreases.

The maximum efficiency of light scattering corresponds to the regular structure of the scattering centers in the form of circular domains that are fairly uniformly distributed throughout the volume of the FLC layer. A decrease in the cell thickness shifts the maximum corresponding to a regular scattering structure toward shorter pulse durations, but the contrast ratio also decreases.

For a certain experimentally chosen relationship between the amplitude and duration of the alternating impulses of the control voltage, and also between the elastic deformation energy of smectic layers and the FLC spontaneous polarization, the process of light scattering on the dynamic domain structure at the transition to the Maxwellian energy dissipation becomes bistable with a maximum light transmission above 80% and a contrast ratio of about 200:1 (Figure 13). Both optical states (with or without scattering) could be turned off for a few tens of microseconds and be memorized for a few tens of seconds, or until a pulse of opposite polarity was applied. The maximum light-scattering modulation frequency was about 5 kHz [16].

A change in the duty cycle between the control voltage pulses (the duration is maintained) leads to a change in the ratio between the lifetimes of both optical states. When an alternating pulse duration is reversed (the duty cycle is maintained), and the duration of a pulse switching on the scattering becomes equal to the duration of a pulse switching off the scattering (and vice versa), the ratio of the lifetimes of states with the maximum light transmission and with the maximum light scattering is reversed.

There is a limit on the minimum duration of voltage pulses which turn on and turn off the scattering process. It should not exceed the characteristic reorientation time of the director caused by the motion of the orientation inflection [16]. From Equation (21), this is about 150 μs for the parameters indicated there.

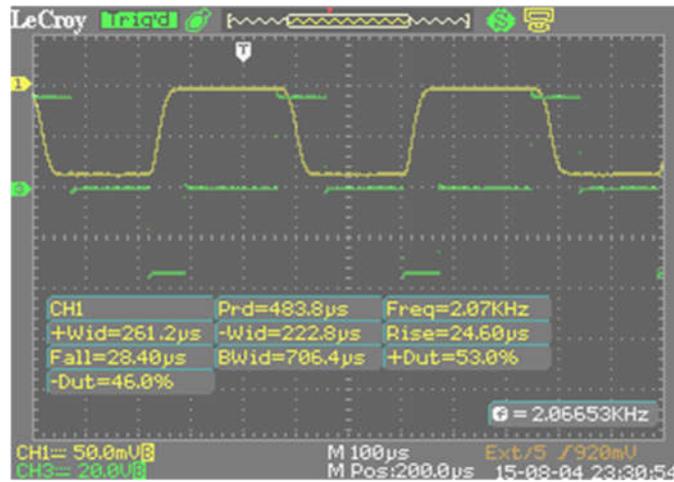


Figure 13. Oscillograms of the control voltage (zero level—digit 3) and optical response (zero level—digit 1) for the bistable switching mode. The upper level of the optical response is the scattering state, the lower level is the nonscattering state. The thickness of the FLC cell with the HF-32 composition is 13 µm. Control voltage—bipolar pulses of ±35 V. The frequency of light modulation is 2 kHz.

There is an optimal relationship between the period of deformation of FLC smectic layers and the electro-optical cell thickness when, at certain electric field strength, the velocity of the soliton wave motion is maximal (the optical response time $\tau_{0.1-0.9}$ is minimal) and the light modulation frequency is also maximal.

Depending on the time of the electric field action (duration of voltage pulses) and the cell thickness, there may occur several maxima of light scattering that may be treated as the scattering efficiency C (or contrast ratio) (Figure 14). The emergence of the second and third maxima of light scattering efficiency occurs with increasing the cell thickness up to 16–20 µm [16].

The maximum efficiency of light scattering and the maximum light transmittance without scattering are achieved at different durations of control voltage pulses. An increase in the pulse duration results in increasing the domain wall length and leads to irregular scattering structures. As a result, the density of scattering centers reduces; this is a reason for the light scattering efficiency decrease (Figure 14).

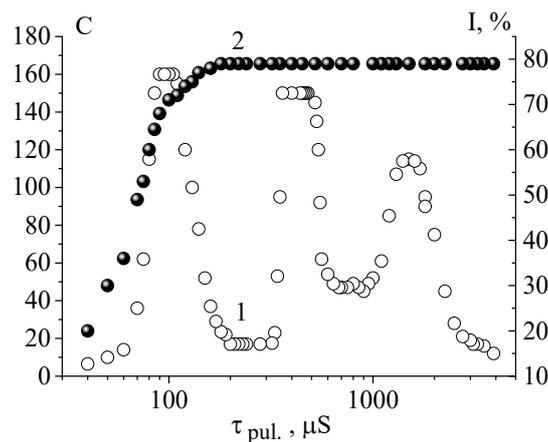


Figure 14. Dependences of light scattering C (curve 1) and light transmission I without scattering (curve 2) on the duration of bipolar voltage pulses with a fixed amplitude (±50 V). The thickness of an FLC cell with the HF-32 composition is 18 µm.

After switching off the electric field, the transmitted optical radiation does not change in spectral composition, and there is no residual light scattering in the FLC layer (due to the absence of a helix). Since the magnitude of the spontaneous polarization does not exceed 50 nC/cm², ferroelectric domains do not arise. Therefore, in the absence of an electric field, scattering and diffraction centers

are absent. The saturation voltage is rather low and, consequently, the control voltage of an electro-optical cell is also quite small (less than 50 V).

The main possible applications of light-scattering FLC compositions are the following: polarizer-free visible and infrared optical shutters, energetically effective screens of electronic books, 3D visualizers (volumetric screens) of volumetric displays, etc. [16,27].

6.3. Spatially Inhomogeneous Modulation of Light Phase Delay

When the duration of voltage pulses supplied simultaneously to the electro-optical cell corresponds to different maxima of light scattering efficiency, the transitions between light-scattering modes (which correspond to light scattering maxima) result in the most chaotic changes in the position of a scattering indicatrix. As a result of a short-term switching on of light scattering (less than for 50 μ s), structures with an almost random distribution of refractive index gradients are formed in the entire volume of the FLC. They cause a spatially inhomogeneous phase modulation of the light beam (over its cross section) passing through the electro-optical cell.

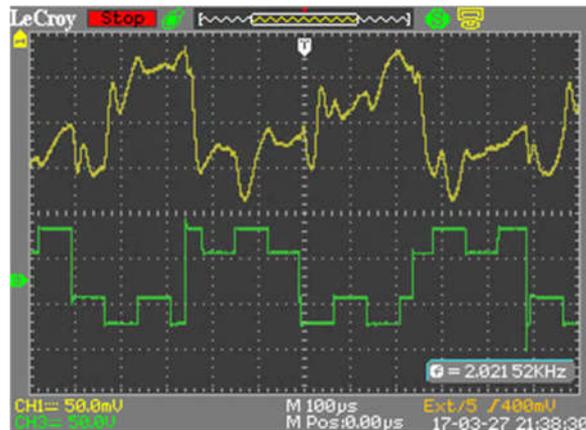
Spatially inhomogeneous modulation with a phase delay of the order of and more than π allows one to destroy the phase relations in a laser beam passing through the electro-optical cell and, further, to suppress the speckle noise in images formed due to the ability of laser rays to interfere.

This approach was used to develop the first electro-optical despeckler, the device that reduces the contrast of speckles and, due to this, suppresses the speckle noise in laser images. In [28,29], helix FLCs were used for this aim, and an alternating electric field was applied across the cell simultaneously at low and high frequencies, which caused spatial deformations of the helicoid in an FLC layer. Unfortunately, such a despeckler had serious drawbacks. First, the deformed helix structure of molecules changed the spectral composition of the laser radiation. Besides this, after the electric field was switched off, the residual scattering caused by the helices remained. Also, the light modulation frequency at the electric field strength of ~ 2 V/ μ m was limited by the value of 500 Hz, which hampers possible applications of this despeckler.

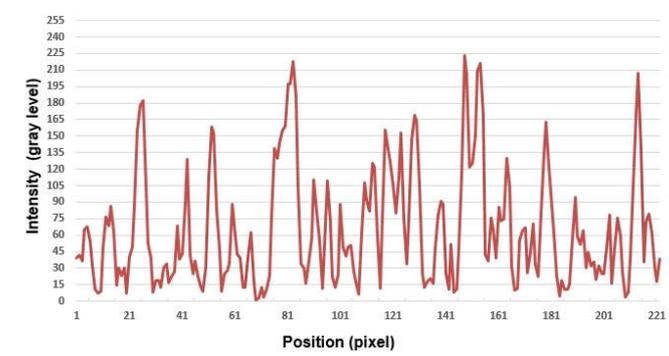
These drawbacks were eliminated after using new helix-free FLC in the experimental samples of a despeckler. Figure 15 illustrates the operation of one of the best samples. Signals of the control voltage and the modulated optical response are shown in Figure 15a. To create the different nonrepeating distributions of refractive index gradients in the volume of the FLC layer, two-frequency voltage pulses (meander) were supplied to the electro-optical cell, and pulses of low frequency (2 kHz) were modulated by pulses of high frequency (10 kHz).

In Figures 15b,c, the speckle intensity distributions in the cross section of the laser beam behind the FLC electro-optical cell are presented for when the control voltage on the cell electrodes is not applied and is applied, respectively. The optical data were recorded using a CCD (charge-coupled device) camera and were processed using a special software product. The reduction in the contrast of the speckle pattern was calculated from the data in Figures 15b,c as the ratio R between the contrast C_0 of a speckle pattern measured without the despeckler sample and the contrast C_1 of a speckle pattern measured with the despeckler sample: $R = 10 \log_{10} (C_0/C_1) = 10 \log_{10} (0.82/0.07) = 10.2$ dB [30,31].

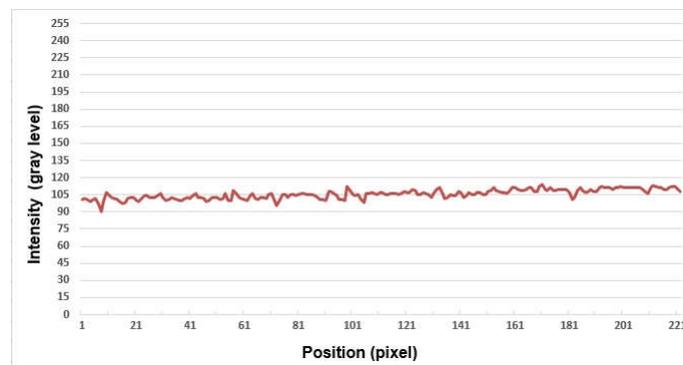
The experiments confirmed the simplicity and high efficiency of the electro-optical despeckler prototype based on an electro-optical cell with the helix-free FLC. Such a device can be widely used in holography and in laser projection displays.



(a)



(b)



(c)

Figure 15. Illustration of the operation of an experimental sample of a despeckler using the novel helix-free FLC: (a) Oscillograms of the control voltage (zero level—digit 3) and phase delay modulation (zero level—digit 1); value of a low-frequency signal (meander) 2 kHz and amplitude ± 50 V; frequency of the modulating signal is 7 kHz and amplitude ± 15 V; electric field strength is 3.25 V/ μm ; (b) Radiation intensity distribution in the cross section of a laser beam transmitted through the electro-optical despeckler sample when the control voltage is zero; contrast of the speckle pattern $C_0 = 0.82$; (c) Radiation intensity distribution in the cross section of a laser beam in the presence of the control voltage; contrast of the speckle pattern $C_1 = 0.07$. The thickness of the FLC cell is 20 μm . The wavelength of the laser radiation is 0.65 μm .

7. Conclusions

Based on our research results, we considered in detail the physical properties, light modulation characteristics, and possible applications of novel helix-free FLC developed and fabricated at the Lebedev Physical Institute, Moscow. They are distinguished by the spatially periodic deformation of smectic layers, a small value of spontaneous polarization, rather high viscosity, and the soliton

mechanism of FLC director reorientation upon transition to the Maxwellian mechanism of energy dissipation.

For FLC with a viscosity in the range from 0.15 to 1.0 P, the frequency and field dependences of the electro-optical response time were studied under modulation of light transmission, light scattering, and light phase delay.

When modulating the light transmission in an electric field of the order of 1 V/ μm (at the control voltage of ± 1.5 V), the smallest response time (25 μs) and the largest modulation frequency interval (7 kHz, including 6 kHz for hysteretic-free modulation with continuous grayscale) were realized experimentally for the first time.

A theoretical model was proposed that described satisfactorily the deformation of smectic layers in the absence of the external electric field and the soliton mechanism of FLC director reorientation under an alternating electric field.

In certain compositions of the new FLC, intensive light scattering on transient domains was realized, including the discovery for the first time of bistable scattering with a memory time that can exceed the optical switching time (tens of microseconds) by 6 orders of magnitude (up to tens of seconds).

Initiated by the short-time switching on of light scattering, the spatially inhomogeneous phase light modulation due to random small-scale gradients of the FLC refractive index was studied. This modulation is capable of destroying the phase relations in a laser beam and of suppressing the speckle noise in images formed by the beam. Based on this, experimental samples of simple and effective electro-optical despecklers were studied for the first time.

Possible applications of the novel helix-free FLC are the fastest display devices (including displays using FLCoS, 3D, and FSC technologies), spatial light modulators, light-scattering polarizer-free modulators (including infrared devices and optical-state-memorizing ones), energetically effective screens of electronics books, 3D visualizers (volumetric screens) of volumetric displays, coding–decoding devices, etc.

8. Patents

Two U.S. patents were issued based on results of the research described above, namely

- Patent No. US 9,532,037 B2: Fast-Acting Low Voltage Liquid Crystal Stereo Glasses. Inventors: Kompanets I.N., Andreev A.L., Ezhov V.A., Sobolev A.G. Date of Patent: 27 December 2016 (PCT Pub. Date: 22 May 2014).
- Patent No. US 9,709,877 B2: Video Projector Employing Ferroelectric Liquid Crystal Display. Inventors: Kompanets I.N., Andreev A.L. Date of Patent: 18 July 2017 (PCT Pub. Date: 5 December 2013).

Author Contributions: conceptualization, A. Andreev and Igor Kompanets; methodology, A. Andreev; software, N. Zalyapin; theoretical model, A. Andreev and T. Andreeva; investigation, A. Andreev and N. Zalyapin; resources, Igor Kompanets; data curation, N. Zalyapin; writing and editing, Igor Kompanets; supervision and project administration, Igor Kompanets; funding acquisition, Igor Kompanets.

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Conflicts of Interest: The authors declare no conflict of interest.

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