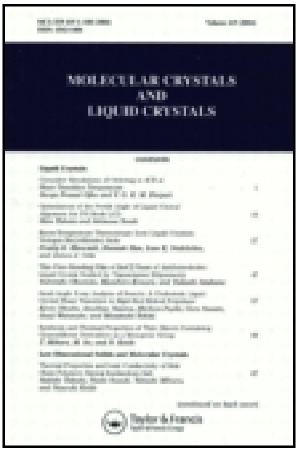

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Photoalignment in Rheology of Liquid Crystals

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Photoalignment in Rheology of Liquid Crystals

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The application of photoalignment technique for rheological study of liquid crystals is considered. Such technique can be used for the initial preparation and rotation of mono domain samples of liquid crystals. It provides new possibilities in measurements of anisotropic viscosities of liquid crystals. Three shear and the rotational viscosities of nematic liquid crystals can be determined at additional usage of the electric field.

Keywords Photoalignment; pure twist deformation; rotational viscosity; shear viscosity

Introduction

Photoalignment (PA) technique was proposed initially as a new technology of surface treatment which provides the given surface orientation of liquid crystals (LC) with the well defined direction and anchoring strength [1]. It is based on various physical (chemical) transformations taking place in the thin layers of dyes or photopolymers under the action of light or UV irradiation in the absence of liquid crystals. As a result such layers become anisotropic and induce a preferred orientation of surface LC layers after filling an optical cell with a liquid crystal. Recently it was shown that PA technique can also be applied to change the surface orientation in the cells already filled with LC [2]. It can be achieved, for example by illumination of polarized light with the orientation of the polarization plane different from that used for the initial PA treatment of surfaces. This effect can be also controlled by additional action of "in-plane" electric field [3]. Such phenomenon has already found application in the new "rewritable" technology [2,4].

This paper is devoted to the perspectives of usage of PA technique for basic studies of rheological properties of liquid crystals stabilized by surfaces. In particular, we will consider the advantages of such approach for two types of experiments providing determination of viscous properties of nematic liquid crystals.

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1. Decay Flow of Nematic Stabilized by Surfaces: New Possibilities for the Shear Viscosity Measurements

It is well known that investigations of nematics under Poiseuille flow can be considered as the most reliable method for determination of anisotropic shear viscosities and Leslie's coefficients. These viscous parameters play the key role in operating times of liquid crystal displays (LCD) and other LC devices. In particular, in some modern types of displays the initial homeotropic orientation is realized [5]. In this case a dynamic optical response is determined not only by the rotational viscosity coefficient γ_1 but also by a so-called "back flow viscosity" η_b, which can be extracted from measurements of anisotropic shear viscosities in Poiseuille flow. Typically, strong magnetic fields are applied to create homogeneous orientation in an NLC layer for such measurements [6]. It enables to neglect the possible action of surfaces. Nevertheless, for the precise measurements of such type [7] large amount (about 10 cm^3) of LC is needed. It prevents the routine laboratory measurements for newly synthesized liquid crystal materials.

Previously we proposed alternative approach for the measurement of anisotropic shear viscosities of LC in decay Poiseuille flow [8]. In this method the given orientation of LC samples is provided by a proper surface treatment. It gives a possibility to determine anisotropic shear viscosities of LC in the samples of small volume (less than 1 cm^3) and applicable for a rapid analysis of new LC materials. In general case the construction of LC cells includes the wedge like channel with regions of different orientations (homeotropic and planar, for example). The optical response induced by a decay flow in a homeotropic part of the cell is used to extract the decay time of the flow, proportional to the effective value η_{eff} of the shear viscosity. The latter becomes close to the given principal viscosity of the planar layer in the case when the hydrodynamic resistance of the homeotropic part of the channel R_h exceeds essentially the resistance of the planar part R_p. In our experiments [8] we used three different LC cells to provide determination of the three principal shear viscosity coefficients (Miesovicz's viscosities). Each cell has to be calibrated by usage isotropic liquid of the known shear viscosity. Evidently, it restricts the application of such technique for routine viscosity measurements.

The situation can be partly improved by additional usage of electric field as an additional factor for LC orientation. It provides a simple way for reorientation of LC samples inside the unique cell and makes possible to determine two Miesovicz's viscosities at least for LC with high positive values of dielectric permittivity anisotropy [9]. The obtained experimental results confirmed that such possibility is easily realized at moderate electric voltages (less than 100 V).

In this paper we describe the further development of the mentioned above approach based on PA technique which can be used for determination of the three principal shear viscosity coefficients by study the decay Poiseuille flow in the unique LC cell. The two types of corresponding experimental geometry are realized by application of PA technique on the preliminary stage (before filling the cell with LC) and at the secondary stage (after filling LC cell) as a tool for reorientation of nematic samples. The third geometry can be obtained by electric field application.

The general construction of a LC cells used in the described experiments (Fig. 1) close to that reported previously [8,9]. It includes two channels with homeotropic (h) and planar (p) orientations. In the latter case the initial planar orientation on the opposite inner surfaces of the channel was achieved by preliminary polarized UV irradiation of azo-dye layer (SD1, Dainippon Ink and Chemicals [1]).

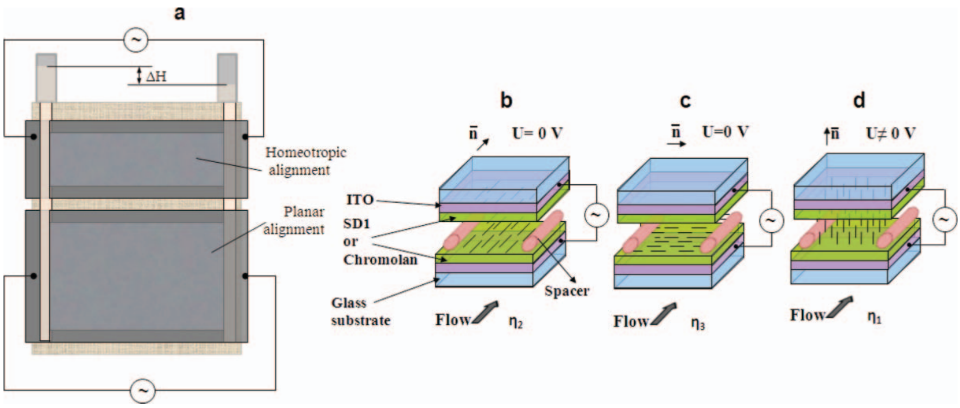


Figure 1. Typical construction of LC cells (a) and experimental geometries corresponding to the three shear viscosities η_1 (d), η_2 (c) and η_3 (b).

It results in the given initial orientation of a mono domain sample of a nematic liquid crystal and make possible to change this orientation after filling the channel with the help of secondary PA treatment (rewritable technology). The open ages of the channels are connected in a parallel schema which provides the same pressure difference ΔP applied to the each channel. The latter difference is proportional to the difference of levels ΔH in the open cylindrical tubes and can be expressed as:

$$\Delta P(t) = \rho g \Delta H(t), \tag{1}$$

where ρ—a density of LC, g—a free fall acceleration. In our experiments the initial difference of levels ΔH(0) of about 5 mm was created by an application of a moderate air pressure difference (not more than 100 Pa) to the open tubes for relatively short time (about 20 s). After drop off the difference of air pressure decay flows were induced in both channels under the action of decreasing difference of a hydrostatic pressure, defined by (1). For a decay flow of a conventional liquid with a constant shear viscosity η_{eff} the time variations of the level (pressure) difference are described by the simple exponential law [8]:

$$\Delta H(t) = \Delta H(0) \exp\left(-t/\tau\right); \tag{2}$$

$$\Delta P(t) = \Delta P(0) \exp\left(-t/\tau\right), \tag{3}$$

where the decay time τ is proportional to the shear viscosity η_{eff}. So, determination of the decay time can be used to calculate the effective shear viscosity coefficient accordingly to the next expression:

$$n_{eff} = K\tau/2, \tag{4}$$

where K—the coefficient, which depends on geometrical sizes of the channels and open tubes. In particular, when the hydrodynamic resistance of the "homeotropic" channel R_h exceeds essentially the resistance R_p of the "planar" channel the latter defines mainly the total resistance of the channels. In this case the parameter K is expressed as:

$$K = \frac{4}{3}\left(\frac{d_p^3 A_p}{L_p \pi D^2}\right)\rho g, \quad (5)$$

where, L_p, A_p, and d_p–the geometrical dimensions of a "planar" channel, D – the inner diameter of the open tubes. The analogous expression is valid for a "homeotropic" channel with the dimensions L_h, A_h, and d_h We used two LC cells with the essential different values of d_p. The dimensions of the channels were: $L_p = 15.0$ mm, $A_p = 15.0$mm, and $d_p = 80\mu$; $L_h = 15$ mm, $A_h = 3.0$ mm, and $d_h = 60\ \mu$ for the cell 1; $L_p = 19.5$ mm, $A_p = 21.0$ mm, and $d_p = 140\ \mu$, $L_h = 19.5$ mm, $A_h = 5.0$ mm, and $d_h = 60\ \mu$ for the cell 2 respectively.

It is simple to check that the inequality $R_h \gg R_p$ is fulfilled for the both cells. The inner diameter of the open tubes D = 2.33 mm was the same in all experiments described below. Usage of the well known nematic- 4′-pentyl-4-cyanobiphenyl (5CB) provides a comparison of our date with the results of independent experiments [10].

The decay time τ can be determined both by direct observation of the meniscus motion and by registration of the dynamic optical response I(t) for polarized light, passing through "homeotropic" channel [8]. In the latter case the phase delay δ(t) between an the extraordinary ray and the ordinary one extracted from I(t) dependence also has to be described by the simple exponential law:

$$\delta(t) = \delta(0)\exp(-t/\tau_\delta), \quad (6)$$

where the relaxation time of the phase delay $\tau_\delta = \tau/2$. So a proper analysis of light intensity changes I(t) induced by a decay flow obeys to calculate the decay time τ of the flow. In particular, for $\delta(0) > \pi$ a monotonic decreasing of the phase delay predicted by (6) results in appearance of local extremes in I(t) dependence as it follows from the well known expression:

$$I(t) = I_0 \sin^2[\delta(t)/2]. \quad (7)$$

It is simple to show [9] using (6) and (7) that the relaxation time of the phase delay τ_δ is connected with the time interval Δt between the last minima and maxima on I(t) dependences by the next expression:

$$\tau_\delta = \Delta t/\ln 2. \quad (8)$$

So, the effective shear viscosity η_{eff} can be expressed as:

$$n_{eff} = K\tau_\delta = K\Delta t/\ln 2. \quad (9)$$

In our experiments we realized both possibilities for determination of a decay time and effective shear viscosity of nematic liquid crystals.

The optical schema of the experimental set up is shown in Fig. 2. It includes a semiconductor laser ($\lambda = 650$ nm), two crossed polarizers oriented at 45^0 relatively to the flow direction, a liquid crystal cell, a photodetector connected with personal computer via ADC, a digital video camera providing snapshots of moving meniscus. Two ac generators were used to apply voltages of frequency 1 kHz to the "planar" (U_p) and "homeotropic" (U_h) channels. The first voltage U_p induced electric field of strength $E = U_p/d_p$ in the planner channel which results in variations of the effective shear viscosity. The second voltage U_h was used to avoid a possible hydrodynamic instability of a homeotropic layer of nematics

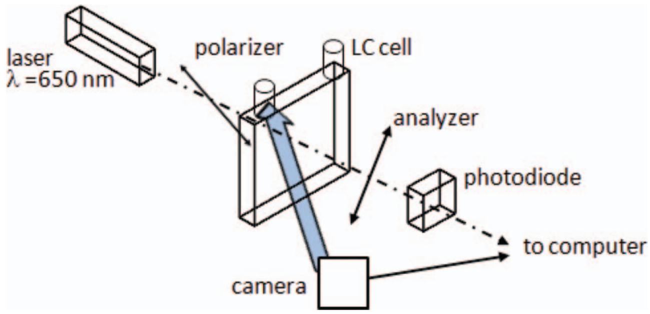


Figure 2. Optical schema of the experimental set up.

under applied pressure gradient [11]. All experiments were fulfilled at room temperature $T = 23 \pm 0.5^{\circ}C$.

The example of snapshots of the meniscus position taken at different times is shown in Fig. 3.

Processing of the digital photos makes possible to obtain the dependences $\Delta H(t)$ which are shown in Fig. 4 for two experimental geometries (see Fig. 1) corresponding to the initial planar orientation in the direction of flow (Fig. 1(a)) and in the direction, normal to the flow plane (Fig. 1(b)). The transformation of the geometry from the first to the second case was made by usage PA technique as it was mentioned above. One can see that experimental dates are well described by the exponential law (2) for both cases. Increasing of the decay time under such transformation ($\tau_3/\tau_2 = 1.19$) can be explained by corresponding increasing of the shear viscosity coefficients ($\eta_3/\eta_2 = 1.63$ [10]).

The time dependences of the intensity I(t) of polarized light passing through a homeotropic channel (cell 2) are shown in Fig. 5 for the cases of zero ($U_p = 0$) and strong ($U_p = 100$ V) electric fields.

In both cases relatively weak ($U_h = 5$V) electric field was applied to the homeotropic LC layer. Increasing of the time interval Δt with voltage ($\Delta t(100V)/\Delta t(0V) = 2.8$) can be assigned to the corresponding increasing of the shear viscosity coefficient (the maximal value of a ratio $\eta_1/\eta_3 = 3.46$) under corresponding transformation of a geometry, shown

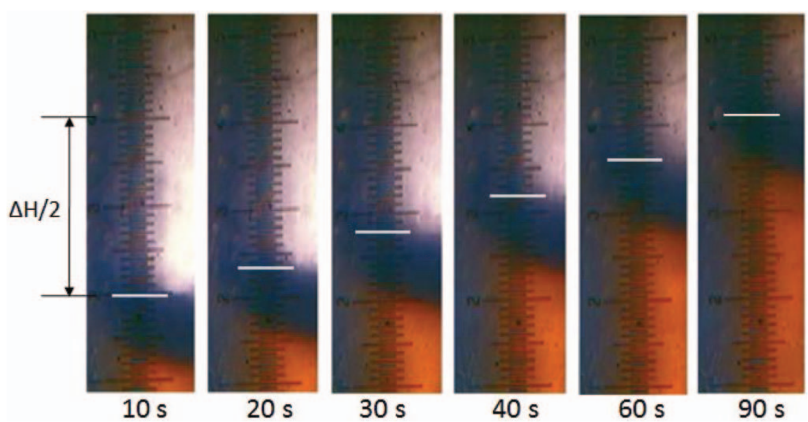


Figure 3. Snapshots of the meniscus position taken at different times, the cell 2, $U_p = 0$.

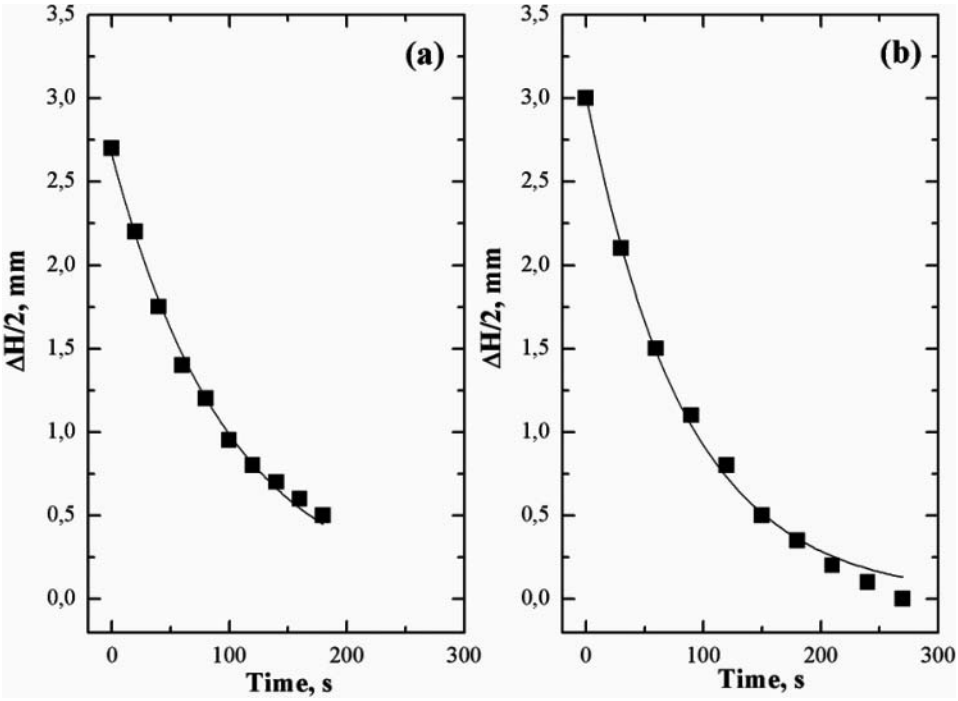
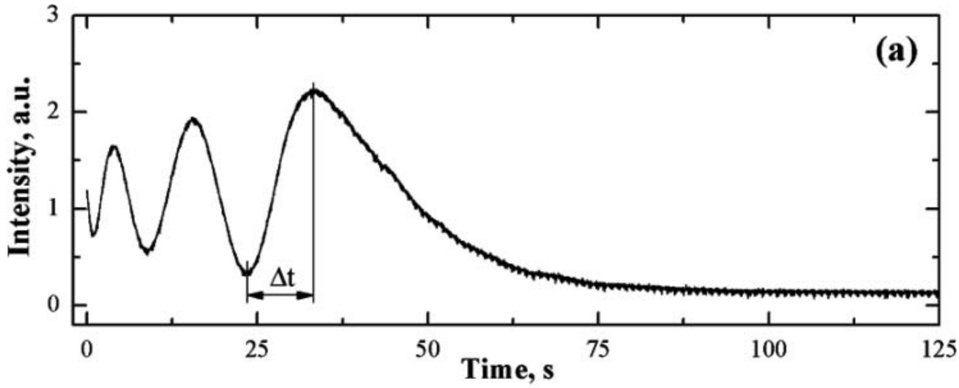


Figure 4. The difference of levels ΔH(t) as a function of time for the initial geometry (Fig. 1(b)) and for the second geometry (Fig. 1(c)), obtained by secondary illumination of the cell 1 by polarized blue light ($\lambda = 450$ nm). Solid lines correspond to the simple exponential law (3).

in Fig. 1. It is worthwhile to notice that both methods of determination of decay time give the similar values of this parameter. It confirms the simple hydrodynamic model used above. The dependence of effective shear viscosity on voltage determined from I(t) analysis using calculated value of constant K is shown in Fig. 6 for the cell 2. One can see that saturation takes place at relatively low voltages (about 50 V). The obtained results are in a general agreement with the independent data on anisotropic shear viscosities of 5 CB. Some difference can be assigned to an inaccuracy in a calculation of the constant K, which can be determined more precisely by calibration of a measuring cell using isotropic liquids [8].

2. Special Geometry for 3D Study of Nematic Liquid Crystals: Rotational Viscosity Measurements

The rotational viscosity coefficient is known as a key parameter defining operating times of LC devices. Some different techniques were proposed to measure it. Direct measurements of a mechanical moment induced in bulk samples by rotating magnetic fields provide the best precision [12]. Nevertheless usage of strong magnetic fields, finest mechanical assumption and large amount of liquid crystals restricts an application of such experimental set-ups for routine laboratory measurements. From this point of view, it is very desirable for practical application to extract the rotational viscosity coefficient from experiments with thin layers of LC. Unfortunately back flow effects introduce essential errors in most cases. It is well



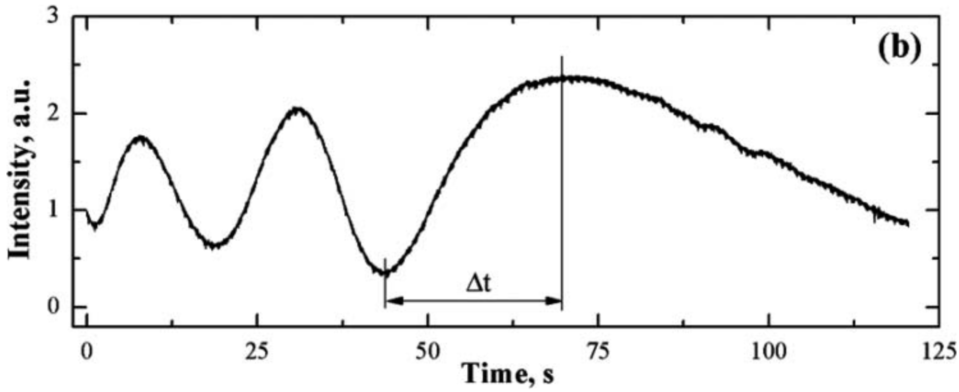


Figure 5. Time dependences of the intensity I(t) of polarized light passing through a homeotropic channel (cell 2) for different voltages U_p applied to the planar layer: $U_p = 0$ V (a), $U_p = 150$ V (b).

known that only pure twist deformation doesn't induce molecular mass motion and so the back flow effects can be ignored. Previously such type of deformation under the action of magnetic fields applied to the thin layers of LC was registered only by conoscopic observations [13]. Thus, it's important to replace the action of a magnetic field by the electric one and usage of optical birefringence measurements instead of a conoscopic study.

The pronounced photoalignment technique allowed constructing non-traditional optical geometry which was previously [14,15] suggested to study of both static and dynamic properties of liquid crystal layers. The key advantages of the cell are very high sensitivity of optical response to the small variations of the twist angle and possibility to create homogeneous "in-plane" electric field. Two pairs of transparent glass substrates were formed to realize a narrow channel of a rectangular cross-section (Fig. 7).

It gives opportunity to observe orientational structure of LC in the cell from both *x* and *z* directions. The upper and bottom surfaces were treated in a standard manner to provide homeotropic orientation and so to avoid arising of disclination lines.

The inner polished edges was covered by sulfuric azo-dye SD1 and illuminated by polarized UV light to produce well defined planar boundary orientation. After it the cell was filled by nematic mixture ZhK 616 (NIOPiK production).

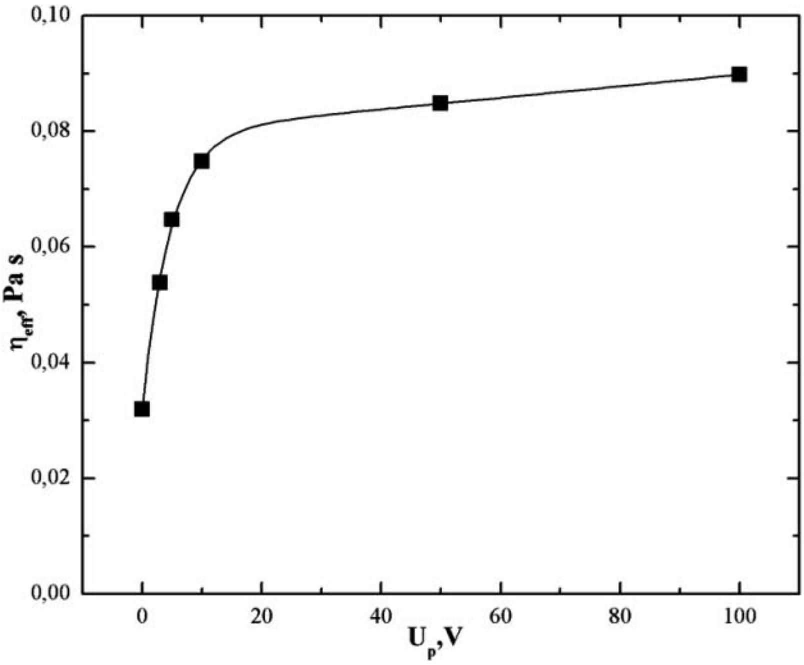


Figure 6. Dependence of the effective shear viscosity $\eta_{\rm eff}$ on voltage U_p, applied to the planar layer (cell 2).

Observation of the channel form x direction corresponds to traditional propagation of light in cells normally to LC layer (Mauguin regime), where application of electric field varies the light intensity from bright to dark state (Fig. 8(a) and Fig. 8(b)).

Contrary to *x* direction, light beams propagating in the plane of LC layer along *z* direction show strong birefringence (B-like effect) due to variation of twist angle. The parameters of the channel (b = 130 μm, d = 1 mm) and symmetric boundary conditions

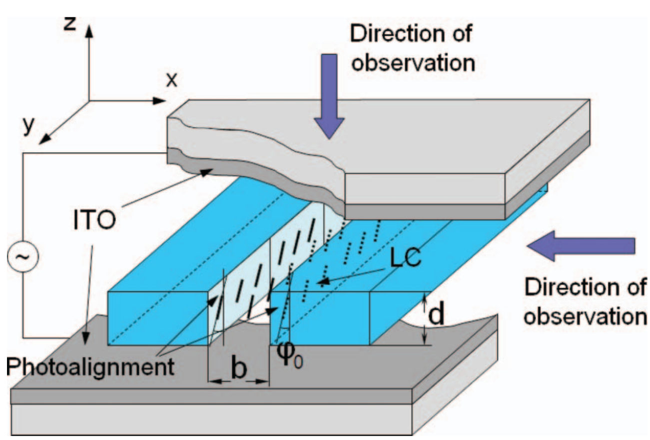


Figure 7. General scheme of LC cell and distribution of NLC director orientation in the channel: $\phi 0 = 21^{\circ}$, b = 130 μm; d = 1 mm.

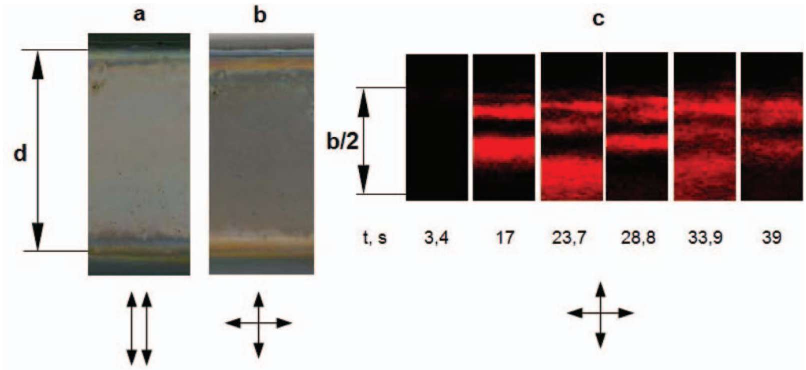


Figure 8. Microscopic images of the channel obtained from x direction (parallel (a) and crossed (b) polarizers) and from z direction (c) after turning off the electric field (in a red color).

with relatively small angle $\varphi_0 = 21^\circ$ of the cell allowed to register the slow variation of interference stripes after switching off the electric field. The set of the instant images of the channel obtained from *z* direction after switching off the electric field is shown in Fig. 8(c).

Such dynamical variations contain information about rotation viscosity of nematic liquid crystals. As there is no back flow effects the rotation viscosity coefficient γ_1 can be determined from I(t) dependences obtained by processing of digital images.

After switching off the electric field, electric field torque is equal to zero. So one can apply simple dynamical equation:

$$K_{22}(\partial^2\varphi/\partial x^2) - \gamma_1(\partial\varphi/\partial t) = 0. \quad (10)$$

The initial condition can be written as:

$$\varphi(x, t = 0) = \varphi(x). \quad (11)$$

where $\varphi(x)$ corresponds to the electrically induced stationary deformation.

Usually such problems are solved via Fourier expansion:

$$\varphi(x, t) = \sum \varphi_n(t) \cos(q_n x). \quad (12)$$

where

$$q_n = (\pi + 2n\pi)/b \quad (13)$$

- is a wave vector.

So the solution of the equation (10) has a well known form:

$$q_n(t) = q_n(0) \exp(-t/\tau_n). \quad (14)$$

where $\varphi_n(0)$ are defined by Fourier transformation of (11) and the relaxation times τ_n depends on the number of a harmonic:

$$\tau_n = \gamma_1/K_{22}q_n^2. \quad (15)$$

At the final stage of the relaxation process the slowest harmonic with the time [16]:

$$\tau_0 = \frac{\gamma_1 b^2}{K_{22}\pi^2}, \tag{16}$$

defines the dynamics of the director reorientation. The corresponding variations of the azimuthal angle are expressed as:

$$\varphi(x, t) = \varphi_0 - \varphi(0)\, e^{\frac{-t}{\tau_0}} \cos\frac{\pi x}{b}. \tag{17}$$

In our geometry the phase delay δ is defined as:

$$\delta(x, t) = \frac{2\pi\Delta n}{\lambda}[\varphi(x, t)]^2, \tag{18}$$

where Δn is the optical anisotropy, d is the thickness of the cell, λ is a wavelength.

The intensity of polarized light passing in *z* direction is expressed as:

$$I(x, t) = I_0 \sin^2\frac{\delta(x, t)}{2}. \tag{19}$$

The latter equation together with (17) and (18) explains the time variations of the interference stripes, shown in Fig. 8(c) and makes possible to determine the relaxation time τ_0.

In the case of large enough phase delay one can obtain the relaxation time τ_0 and the rotational viscosity coefficient (using the value of K_{22}) by measuring the time intervals between interferential maximum or minimum:

$$\tau_0 = \frac{t_m - t_n}{\ln\frac{\sqrt{\delta_m} - \sqrt{B}\varphi_0}{\sqrt{\delta_n} - \sqrt{B}\varphi_0}}, \tag{20}$$

where $B = 2\pi d\Delta n/\lambda$; t_m, t_n–time coordinates of extremums and $\delta_m[\delta_n] = 2\pi m[2\pi n]$— for maxima; $\delta_m[\delta_n] = 2\pi(m\text{-}1)[2\pi(n\text{-}1)]$-for minima.

The results of such calculations are presented in the Table 1.

The average value γ_{1av} of rotational viscosity coefficient is in agreement with independent measurements [17].

The comparison of experimental results and theoretical calculations is shown in Fig. 9.

Table 1. The time coordinates of interferential maximum (t_m^{max}) or minimum (t_n^{min}) at different *x* and the calculated values of γ_1

	γ_1, P						
x, μm	t_2^{max}, t_1^{max}	t_3^{max}, t_2^{max}	t_3^{max}, t_1^{max}	t_2^{min}, t_1^{min}	t_2^{min}, t_2^{max}	t_3^{min}, t_2^{max}	γ_{1av}, P
28	2,36	2,3	2,33	2,36	2,31	2,31	
24	2,59	2,13	2,38	2,45	2,23	2,21	2,36
18	2,47	2,28	2,38	2,49	2,67	2,21	

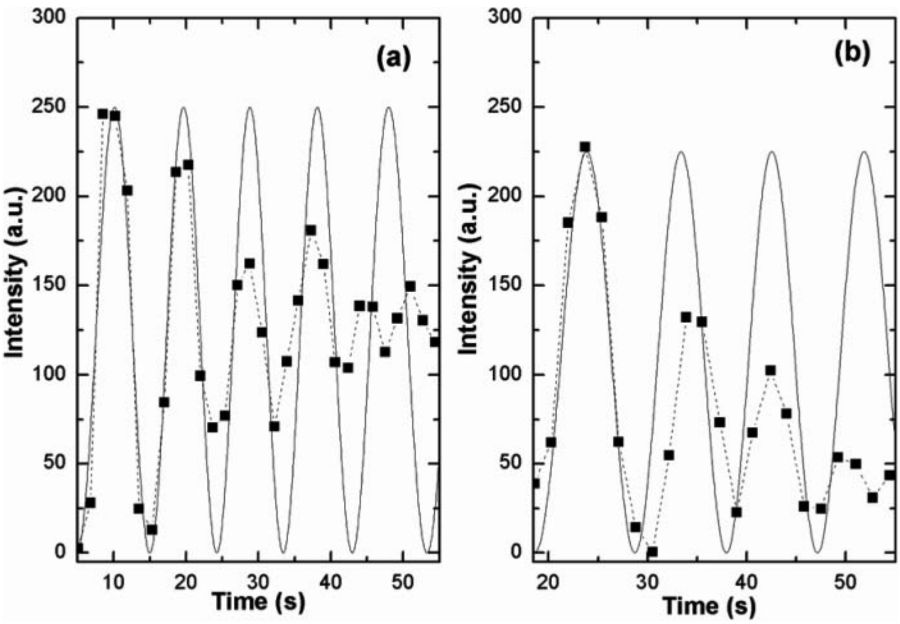


Figure 9. Time dependence of light intensity at different x coordinates after turning off the electric field (solid lines—the theoretical curves corresponding to the averaged value of $\gamma 1$): a) x = 28 μm, b) x = 18 μm.

Conclusion

Usage of photoalignment technique (PA) opens new perspectives in rheological investigations of liquid crystals. Firstly, it enables azimuth rotation of LC samples and measurements of anisotropic shear viscosities of nematics. A combination of optical rewritable technology and electric fields provides determination of the three principal viscosities (Miesovicz's viscosities) by using the unique measuring cell. Secondly, PA technique is very attractive for a proper surface treatment in LC cells of complicated geometries. In particular, such cells can be used for registration of small azimuth rotation of LC director and determination of the rotational viscosity coefficient. The proposed decisions increase a reliability of obtained information about anisotropic viscous properties of nematics. They also enable to elaborate compact and simple devices for routine viscous measurements of newly synthesized liquid crystal materials.

Acknowledgments

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