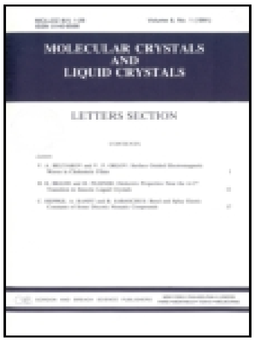

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Photo Controlled Surfaces in Rheology of Liquid Crystals

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The modified method for shear viscosity measurements of nematic liquid crystals (NLC) oriented by surfaces is described. The main modification is concerned with usage of photoalignment technique for preparation of mono-domain samples with an orientation controlled by a polarization state of the secondary light irradiation. It provides the azimuthal rotation of a sample at any desirable angle. Thus different geometries of shear flows can be realized after filling the measuring cell with a liquid crystal. The method is experimentally approved at investigation of Poiseuille decay flows of 4-pentyl-4-cyanobiphenyl (5CB) through the channels with a rectangular cross section. The proposed construction of the cell with different surface treatment provides measurements of the three principal viscosities coefficients (Miesowicz' viscosities) with additional usage of electric field. A small amount (less than 0.2 ml) of a liquid crystal needed for measurements and simple measuring procedure makes the method to be useful for rheological studies of newly synthesised liquid crystal materials.

Keywords nematic liquid crystals; photoalignment technique; anisotropic shear viscosities; Poiseuille flows

Introduction

It is well known that liquid crystals (LC) play the key role in modern display industry [1]. Moreover, the alternative applications of liquid crystals like those for fiber optics or sensors of different types [2] are in a progress now. In general, it stimulates elaboration of new liquid crystal materials with improved physical properties. The operation of a majority of liquid crystal devices (liquid crystal displays, for example) is based on variation of optical properties under the action of electric fields. That's way the material parameters of LC, like refractive indexes, dielectric permittivity anisotropy ($\Delta\varepsilon$), Frank's elastic moduli K_{11}, K_{22} and K_{33}, rotational viscosity coefficient γ_1 which describe a number of electro-optical effects were under a special attention. As a result a lot of experimental methods and set-ups were proposed for determination of the parameters mentioned above [2–4]. Most of

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them demand rather small amount of LC under investigation which is important for newly synthesized materials.

Contrary, the current situation with viscosity measurements in liquid crystals looks not so optimistic. For most practical application nematic liquid crystals can be considered as an incompressible uniaxial media in which velocity gradients induce rotation of the local optical axis (director n) and vice versia. It results in arising of five independent Leslie viscosity coefficients α_i, entering into hydrodynamic equations. Combinations of these parameters describe the energy dissipation for different types of liquid crystal motion. In the simplest case of a pure rotation of the LC director n in the plane of LC layer without fluid motion (T-deformation) the unique coefficient of rotational viscosity $\gamma_1 = \alpha_3 - \alpha_2$ is enough for such description. This parameter is the most important for the characterization of LCD dynamic behavior. However, in general case orientational motion is connected with the shear flow (back-flow effect [5]) which modifies dynamic response in electro-optical modes of different types [1, 6, 7]. In particular, for a frequently used twisted nematic (TN) mode—which includes twist, bend and splay deformations—back flow results in some increase of the response time [1]. By contrast, in the optically compensated bend (OCB) mode the back-flow effect plays the main role, providing several times faster response times in comparison with the TN mode [6]. The latter is also true for electrically induced deformations in vertically aligned cells at negative sign of $\Delta\varepsilon$ (VAN-mode). Correct theoretical description of the mentioned above optical modes involves not only the rotational viscosity coefficient but, additionally other combinations of Leslie coefficients. The information about the latter parameters is also of a practical importance for better understanding mechanic-optical phenomena in liquid crystals which can be used in LC sensors [2] and optofluidic devices [7].

Nevertheless, one can find only restricted number of LC materials with the known values of Leslie coefficients. Among different experimental methods rheological investigations of liquid crystals in shear flows provide the most reliable information about dissipative parameters of liquid crystals.

It is well known that in a general case liquid crystals show non-Newtonian behavior. In particular, the apparent shear viscosity measured at Poiseuille flows through a capillary may depend on a number of factors such as a shear rate, a diameter of a capillary and initial boundary orientation [8, 9]. But in well oriented samples (usually by magnetic fields) at low velocity gradients liquid crystals can be considered as conventional Newtonian liquid with a shear viscosity dependent of orientation [10]. It makes possible to extract the complete set of Leslie's coefficients via precise measurements of shear viscosities at different experimental geometries [2]. Experiments of such type performed for plane flows of both Couette [11–16] and Poiseuille [17–20] types have shown that only strong magnetic fields (of induction about 1T) can suppress the orientational action of surfaces and flows to guarantee Newtonian like behavior of liquid crystals. It results in relatively large amount (about 10 ml) of liquid crystals needed for correct measurements which prevents usage of such technique for a study of newly synthesized LC materials.

Some time ago we proposed the alternative experimental decision for measurements of anisotropic shear viscosities of liquid crystals [21]. The main idea was to use stabilizing action of surfaces instead of magnetic fields to provide the given orientation of LC. The experiments with a decay Poiseuille flow through capillaries of a rectangular cross section confirmed an applicability of such approach. The main advantages are connected with rather small amount (about 1 ml) of LC and essential simplification of the experimental set-up which makes it to be attractive from the point view of practical application. Nevertheless, in this case a number of calibrated LC cells with different orientation have to be prepared

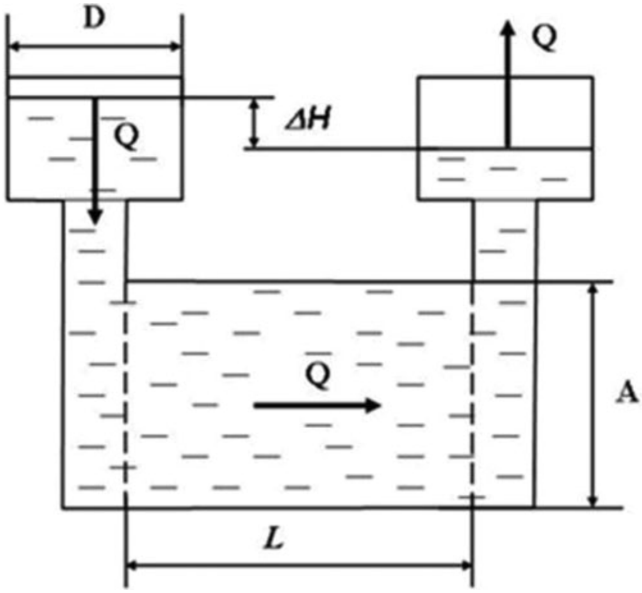

Figure 1. The schema of a decay flow.

to determine different viscosity coefficients. It complicates, to some degree a measurement procedure in the comparison with that for the case of magnetically controlled orientation of liquid crystals inside the flow channel. Some improvement can be achieved by additional usage of electric fields for stabilization LC structure [22]. It provides determination the maximal and minimal values of anisotropic shear viscosity for liquid crystals with a positive sign of dielectric permittivity anisotropy via a study of the decay flow through the unique channel.

In this paper we present the modified method and the results of measurements of anisotropic shear viscosities of liquid crystals. The modification involves the usage of LC cells with the inner surfaces coated by a nanometer layers of photosensitive azosulfidic dye. It provides a possibility to control orientation of LC layer via usage of polarized light. Thus different experimental geometries, needed for anisotropic viscosity measurements can be realized for the same measuring cell. It results in determination of the three principal shear viscosity coefficients (Miesovich viscosities) of liquid crystals with a positive sign of dielectric permittivity anisotropy.

Theoretical Backgrounds

In the simplest case a decay shear flow arises in the channel of the length L, width A and gap d via the difference of levels ΔH in the open tubes of diameter D connected by the rectangular channel (Fig. 1). The flow is produced by the instant pressure difference (1) slowly decreasing with time:

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

where ρ - a density of a liquid, g – a free fall acceleration. The fluid volumetric flow rate $Q = dV/dt$ is the same in different cross sections of the hydrodynamic circuit and can be expressed as:

$$Q = (S/2)(d\Delta H/dt), \tag{2}$$

where $S = \pi D^2/4$ – the cross section of the open tube.

Obviously, a certain time is required for complete cessation of the LC motion which depends on the shear viscosity. Actually such a decay flow takes place not only in liquid crystals, but also in isotropic fluids. In the latter case a Newtonian-like behavior with a constant value of shear viscosity is realized. By contrast, there is a connection between the velocity gradients and the LC alignment in the liquid crystalline media. Thus the shear flow can change the initial LC orientation and non-Newtonian behavior takes place (the effective shear viscosity depends on time).

The detailed theoretical description of a decay Poiuseulle flow of liquid crystals both for linear and non-linear regimes was presented earlier [2, 23]. Thus, in this paper we focus on some final results important for practical realization of the modified method for shear viscosity measurements. They can be summarized as follows:

1. For conventional Newton–like liquid with a constant shear viscosity both the difference of levels ΔH in the open tubes and the hydrostatic pressure difference ΔP decays exponentially with time:

$$\Delta H(t) = \Delta H(0) \exp(-t/\tau); \tag{3}$$

$$\Delta P(t) = \Delta P(0) \exp(-t/\tau). \tag{4}$$

2. The characteristic decay time τ is proportional to the shear viscosity coefficient η:

$$\tau = (1/K)\eta, \tag{5}$$

where K – the constant defined by geometrical sizes of the measuring cell and the density of a liquid as follows:

$$K = \frac{2}{3}\left(\frac{d^3 A}{L\pi D^2}\right)\rho g, \tag{6}$$

3. Equations (3) and (4) are also hold (or approximately hold) for nematic liquid crystals if the orientational structure inside the channel does not vary (or slightly vary) with time. In this case the parameter η can be considered as an effective shear viscosity coefficient dependent on orientation. For monodomain samples of LC this dependence is expressed by the well-known equation [2]:

$$\eta(\theta, \phi) = \eta_2 \cos^2\theta + (\eta_1 + \eta_{12}\cos^2\theta)\sin^2\theta\cos^2\phi + \eta_3 \sin^2\theta \sin^2\phi, \tag{7}$$

where, angles θ and ϕ define the orientation of a director respectively to the direction of a velocity v and a velocity gradient ∇v,

$$\begin{aligned} \eta_1 &= \eta(\pi/2, 0) \\ \eta_2 &= \eta(0, \pi/2) \\ \eta_3 &= \eta(\pi/2, \pi/2) \end{aligned} \tag{8}$$

where η_1, η_2, η_3 – the three principal viscosity coefficients (so-called Miesowicz viscosities), corresponding to the main geometries for viscosimetric measurements, η_{12} – the coefficient which enters into viscosity in combination with the principal viscosity coefficients.

4. The initial homeotropic orientation shows the highest sensitivity to the action of a shear flow. In particular, there is no threshold flow velocity gradient, so small declinations from initial orientation are possible even at weak flows. Such declinations can be used to control time variations of a pressure gradient G(t) applied to LC layer via polarized light passing through the layer. In this case the optical phase difference δ(t) is proportional to G(t) and exponentially decays with time:

$$\delta(t) = \delta_0 \exp(-t/\tau_\delta), \tag{9}$$

where the relaxation time of the phase difference:

$$\tau_\delta = \tau/2 = \eta_1/2K. \tag{10}$$

The time dependence of intensity I(t) of light, passing through the layer (cross polarizers are oriented at 45^0 relatively to the flow direction) is expressed by the well-known expression:

$$I(t) = I_0 \sin^2 \delta(t), \tag{11}$$

where I_0 – input light intensity. So, processing of I(t) dependence provides, accordingly to (9–11), determination of τ_δ and an effective viscosity coefficient η.

5. A number of orientational instabilities can arise in moderate and strong flows of LC [2]. It restricts the range of pressure gradients where the simple model of a decay flow holds. For homeotropic orientation a stabilizing electric field can be effectively used to suppress the instability connected with an escape of a director from the flow plane [2, 23].

Experimental

1. Methods of Surface Treatment and Rotation of LC Sample

In these experiments, we used two types of planar cells. The cell 1 (of LC thickness layer 20 μm), was made by according to standard ORW technology [27]. Namely it consists of two substrates with inner surfaces treated by different procedures. The first substrate was treated accordantly with traditional rubbing technique to provide planar orientation of LC with high value of an anchoring strength (strong anchoring). The second substrate was coated by photoalignment layer (SD-1) and illuminated by UV polarized irradiation ($\lambda = 365$ nm) which made possible to obtain a planar orientation with anchoring strength dependant on the irradiation dose D. It also provides easy axis rotation to the given angle α due to the secondary irradiation by polarized light ($\lambda = 450$ nm) of new polarization state after filling the cell with LC. Both substrates of the cell 2 (of LC thickness layer 80 μm), were treated by the above mentioned photoalignment technique. To change the orientation of the surface monodomain samples, we used different ways. The first one included heating of the cell up to the temperature T≈50°C corresponding to an isotropic phase of LC (5CB, the clearing temperature $T_{NI} = 35$°C) to delete optical phase delay, the secondary irradiation

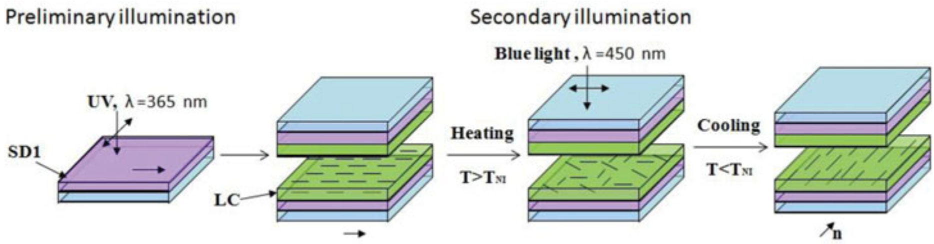

Figure 2. The scheme of treatment of LC cell corresponding to the first way.

by polarized light and cooling to a nematic phase. The corresponding scheme is shown in Fig. 2.

The second way was based on the secondary irradiationof cell filled with LC in the presence of strong electric field U needed to suppress the optical phase delay in nematic phase and frequency f = 10 kHz. The corresponding scheme of the experimental set-up

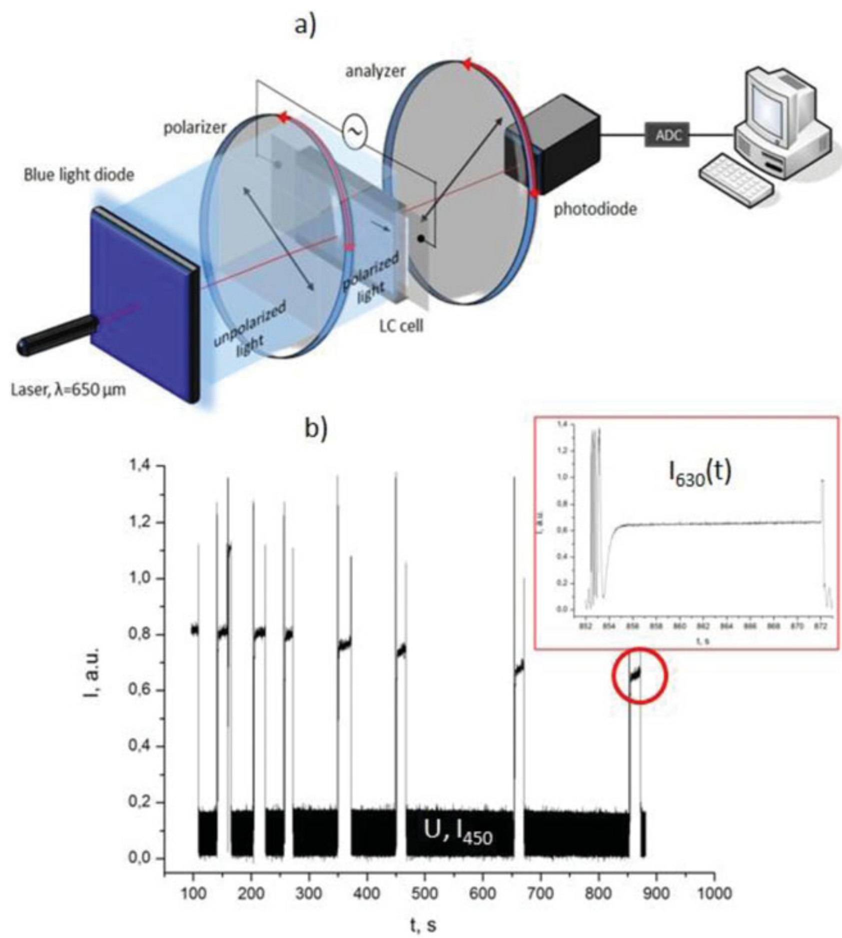

Figure 3. The experimental set-up (a) and time dependence of light intensity I_{630} (b).

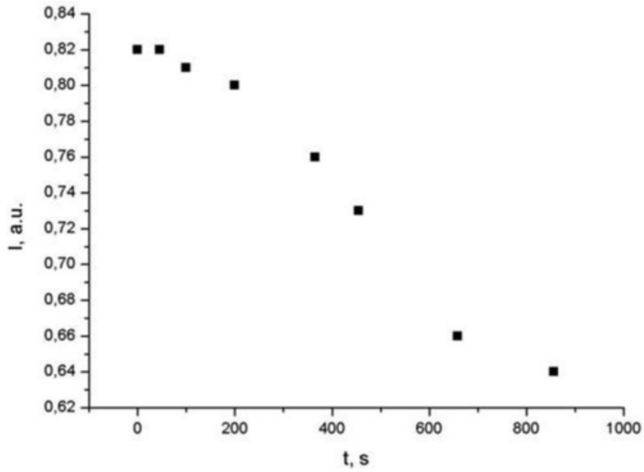

Figure 4. The time dependence of light intensity I_{630} induced by rotation of easy axis on photosensitive substrate of the cell 1to angle $\alpha = 45°$, the applied voltage U = 50 V.

is shown in Fig. 3a. It includes a blue light source for the secondary irradiation, as well as a semiconductor laser with a wavelength $\lambda = 630$ nm and a photodiode for registering changes of the intensity of the transmitted light laser I_{630}.

During the experiment, the combined influence of electric field and polarized light was stopped for a short time period appropriate for the relaxation of the bulk orientation of the LC layer under the action of surfaces. The optical response of LC cell on turning on and turning off blue light and electric field is shown in Fig. 3b. The time dependences of the laser light intensity associated with the rotation of the easy axis of LC on photoalignment surfaces can be obtained by using data on I_{630} registered during turning off light irradiation and electric field.

The results obtained for the two cells and different procedures of treatment differ relatively to the values of a characteristic time t_r needed to change surfaces orientation. In particular, the time of reorientation of easy axis to the angle $\alpha = 45°$ for cell 1 treated accordantly with the first way was about 10 minutes. At the same time, the values of t_r obtained by the second way showed strong dependence on the reorientation of the sample relatively to the direction of radiation, changing from 5 minutes (irradiation from the photosensitive substrate) to 120 minutes (irradiation from the rubbed substrate). The time dependence of light intensity $I_{630}(t)$ is shown in Fig. 4.

Similar effects were observed at study of the cell 2 treated by the ways described above. In this case, it was needed to perform the secondary illumination by changing (after 15 minutes) the position of the cell relatively to the input irradiation. It provided rotation of the monodomain sample. The intensity of transmitted laser light as function of the exposure time t is shown in Fig. 5.

2. *Experimental Results of Viscosity Measurements*

The measuring cells had two channels with homeotropic and planar orientations, two cylindrical tubes for supplying identical pressure difference for each channel. This allows creating a decay Poiseuille flow. This flow can be registered by direct observation for

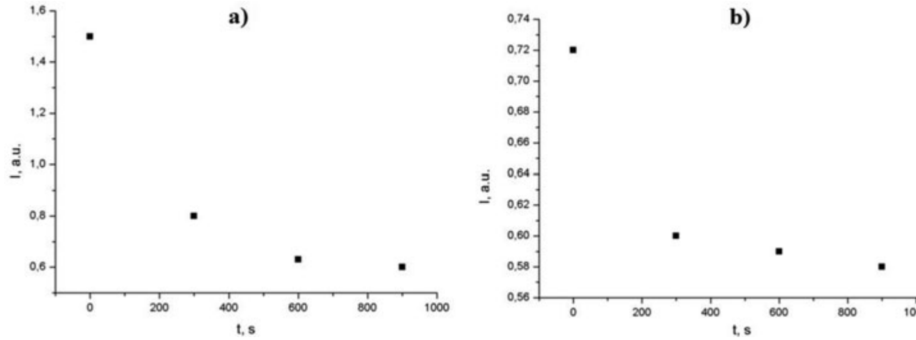

Figure 5. The time dependence of I_{630} for the cell 2 irradiated from the first (a) and second (b) substrates; U = 80 V, $\alpha = 45^o$.

meniscus in the tubes or by processing of light intensity which passes throw homeotropic channel of the cell [31]. The construction of the measuring cell is shown in Fig. 6.

It includes two channels of a rectangular cross section with a big aspect ratio (A/d>>1) providing simple plane flows through the channels. The channel is formed by glass substrates with an essentially different treatment of the inner surfaces. Namely, for the homeotropic channel the standard spin coating technique was used to get a thin polyimide film which provides, after baking and further rubbing, the homeotropic boundary conditions for near surface layers of a liquid crystal [1]. For the second channel the opposite inner surfaces of the substrates were spin coated by 0.5 wt.% solution of sulfonic azo dye (SD1, Dai-Nippon Ink and Chemicals, Japan) in DMF (N-dimethylformamide), with the further evaporation of the solvent and the treatment by polarized UV irradiation.

Such standard procedure [24] provides a planar surface orientation of a liquid crystal in the direction, normal to the polarization plane of the input electromagnetic wave. In our case the same direction of the surface orientation on both substrates was created which resulted in a mono domain planar sample of a nematic liquid crystal after filling the channel. It is of importance that such sample can be rotated at any desirable azimuthal angle ϕ with the help of the secondary UV or light irradiation of both substrates, *after* filling the cell. In this case, light with polarization plane different from that for preliminary irradiation is used. It results in slow change of preferable overall orientation (an easy axis) of LC molecules absorbed by surface. Such so-called gliding of an easy axis depends on a number parameters like a preliminary and a secondary doses of irradiation, the initial angle (an azimuth ϕ_L) of a

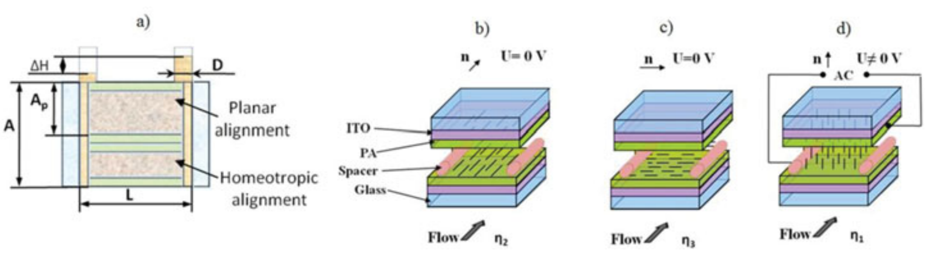

Figure 6. The construction of two channel LC cell (a) and experimental geometries corresponding to the three shear viscosities η_1 (d), η_2 (c) and η_3 (b).

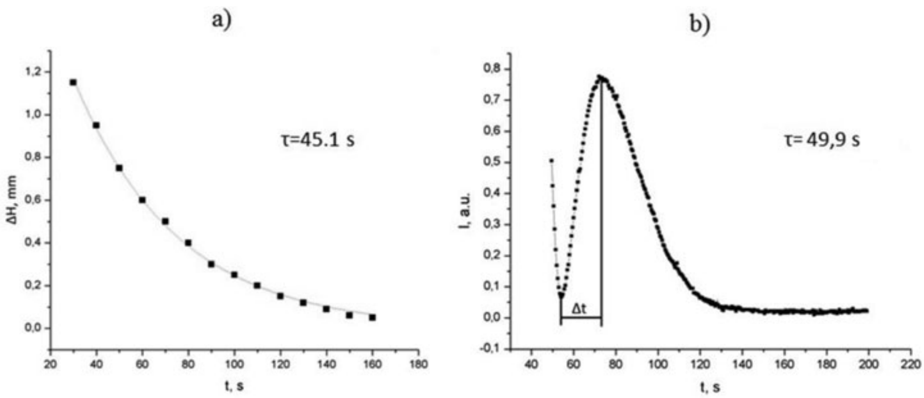

Figure 7. Dependences of: a) level difference on time; b) light intensity on time.

polarization plane of the secondary irradiation respectively to that at preliminary irradiation, electric field strength for a case of a combined action of light and field [25,26]. It demands some time (ranging from some seconds to some hours) to reach new stationary direction of surface and bulk orientation, corresponding to the rotation of the sample by the angle $\phi \approx \phi_L$. In general, this procedure can be considered as modification of previously proposed optical rewritable technology [27], where only one photosensitive substrate was used.

The decay time τ of a flow was determined by processing the digital images of the moving meniscus in the open tubes taken off at different times. This method was chosen as the simplest one, as it demands only a digital webcam connected with a computer. As it was shown previously [21] the similar information can be obtained by processing the intensity of light I(t) passed through homeotropic LC cell (or part of the cell), which is connected by parallel schema with the channel under investigation. In the latter case equations (9)–(11)

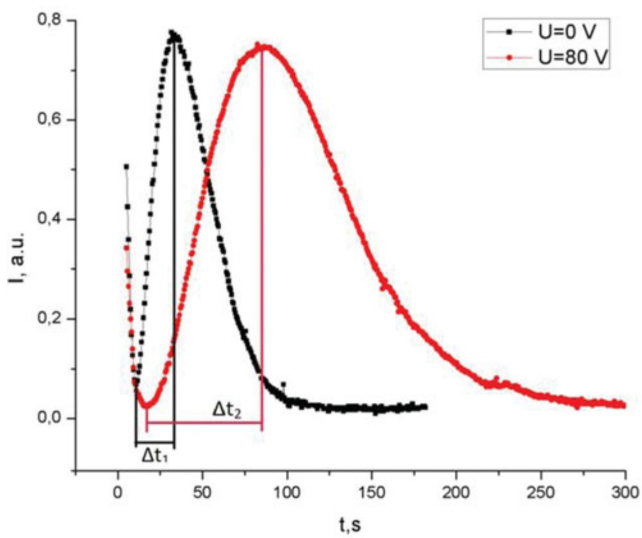

Figure 8. Time dependences of light intensity on different voltages.

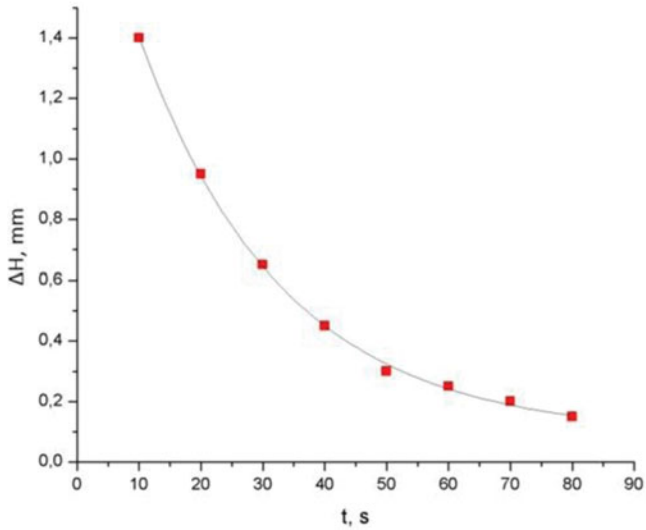

Figure 9. The difference of levels $\Delta H(t)$ as a function of time for an isotropic phase of 5CB (T = 51°C); the curve lines correspond to the simple exponential law (3).

are applicable to determine the decay time τ. It was checked experimentally that the results obtained by the two alternative methods are close to each other (Fig. 7). In experiments described below we used the well-studied nematic 5CB. It provides a comparison of our date with the results of independent experiments [28].

The time dependences of light intensity at different voltages are shown in Fig. 8.

The values of Δt at different voltages are proportional to the decay flow τ and can be used to determine anisotropic shear viscosity.

These experiments were made at the same temperature T = 25 ±0.2°C to provide correct comparison with the independent viscosity data. The experimental dependencies are well described by a simple exponent law (3) for all geometries under investigation. It confirms realization of a simple plane flow of liquid crystals with an effective shear viscosity independent on time. The data obtained for an isotropic phase (see Fig. 9) can be used for calibration of measuring cell.

The comparison of our data with the results obtained from precise viscosity measurements of Kneppe and Shneider [28] is presented in the Table. We neglected the changes of a LC density with temperature, which, accordingly to (5) can modify the calibration constant K. Accordingly to the density data for 5CB [29] such simplification results only in a small error (about 1%) at viscosity calculations.

Table

θ	ϕ	Principal viscosity coefficient	τ,s	η_i, Pa.s this work	η_i, Pa.s [28]
$\pi/2$	0	η_1	264	0.114	0.113
0	$\pi/2$	η_2	205	0.0230	0.0213
$\pi/2$	$\pi/2$	η_3	307	0.0344	0.0342

As a result of comparison one can conclude that the difference between two sets of data does not exceed 7% for the minimal viscosity coefficient and essentially lower for alternative experimental geometries. It confirms the simple theory of a linear decay flow mentioned above and makes possible to propose modified method for anisotropic shear viscosities measurements with a reasonable accuracy. The measuring procedure is rather simple and no expensive experimental technique is required. It is applicable for both isotropic and anisotropic liquids. The obtained data can be used to estimate the value of a rotational viscosity coefficient from the next expression [2]:

$$(\eta_1 - \eta_2)\cos 2\theta_0 = \gamma_1. \tag{12}$$

At small values of the flow alignment angle θ_0 (tg $\theta_0 = \alpha_3/\,\alpha_2$) which usually takes place far from transition point the approximate expression:

$$\gamma_1 \approx \eta_1 - \eta_2 \tag{13}$$

is valid. As an example, for 5CB $\theta_0 \approx 12^0$ at room temperature [30] and equation (13) holds with accuracy of about 10%. From the results, presented in the Table it follows $\gamma_1 = 0.091$ Pa.s. The value is in a correspondence with independent data [30] on rotational viscosity coefficient of 5CB.

Conclusion

In conclusion, the simple experimental method is proposed for measuring anisotropic shear viscosities of liquid crystals. It is based on a possibility to change a surface orientation of a liquid crystal via secondary treatment of photosensitive layers contacted with LC by polarized light irradiation. Such approach provides rotation of the planar monodomain sample of a liquid crystal at the given azimuthal angle to realize different geometries in experiments with shear flows of liquid crystals. The method is tested via study of decay Poiseuille flows of 5CB in the two channel cell applicable with different surface treatment. A reasonable agreement with the independent viscosity data is obtained. The proposed method does not demand usage of strong magnetic field and well suited for routine laboratory testing of small (less, than 0.2 ml) samples of newly synthesized LC materials.

Funding

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