UDC 532.5, 532.6

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DECAY FLOW OF LIQUID CRYSTALS IN THE PRESENCE OF MAGNETIC AND ELECTRIC FIELDS FOR VISCOSITY MEASUREMENTS

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In this paper we describe the investigation results of a Poiseuille's shear flow of nematic liquid crystal through the plane channels with the initial planar orientation perpendicular to the flow plane in the presence of electric or magnetic fields, needed for realization of three principal flow geometries. The obtained experimental data were analyzed in the framework of the simple model describing slipping and anti-slipping action of the surface layers on the effective shear viscosity. That made it possible to estimate the possibility and errors of measurements of Miesowicz viscosities in microliter samples of liquid crystals, which is important for characterization of newly synthesized liquid crystals.

Key words: *decay flow, Miesowicz viscosities, electric and magnetic fields.*

DOI: 10.18083/LCAppl.2020.3.49

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ЗАТУХАЮЩИЙ ПОТОК ЖИДКИХ КРИСТАЛЛОВ В ПРИСУТСТВИИ МАГНИТНОГО И ЭЛЕКТРИЧЕСКОГО ПОЛЕЙ ДЛЯ ИЗМЕРЕНИЙ ВЯЗКОСТИ

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В данной работе представлены результаты исследований Пуазейлевского сдвигового течения нематического жидкого кристалла через плоские каналы с исходной планарной ориентацией, перпендикулярной плоскости потока, в присутствии электрического или магнитного полей, необходимых для реализации трех основных геометрий потока. Полученные экспериментальные данные анализируются в рамках простой модели, описывающей влияние скольжения и противоскольжения поверхностных слоев на эффективную сдвиговую вязкость. Это позволило оценить возможность и погрешности измерения вязкости Месовича в микролитровых образцах жидких кристаллов, что важно для характеристики вновь синтезированных жидких кристаллов.

Ключевые слова: *затухающий поток, вязкости Месовича, электрическое и магнитное поля*

Introduction

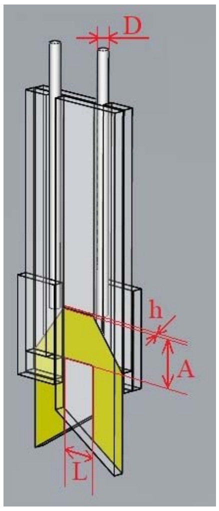
It is well known, that liquid crystals (LC) play a leading role in the modern display industry. Nowadays they are also applied in different photonic devices (modulators, shutters, deflectors, switchers etc.) designed for the control of light beams [1]. In most cases, the operation of the mentioned above devices is based on the changes of the initial orientational structure, induced by a proper surface treatment, and optical properties of LC layers under action of electric field. Dynamics of the given electro-optical effect is determined by a number of parameters like an electrical voltage, a thickness of LC layers, the type of surface orientation as well as elastic (Frank's moduli) and viscous (Leslie's coefficients) characteristics of a liquid crystal [2]. Optimization of viscous characteristics of LC materials is one of the most promising ways to decrease operating times of the mentioned above devices based on liquid crystals. Up to now, the rheological study of liquid crystals in shear flows of different types provides the most reliable data on anisotropic viscosities important for practical applications [2]. Moreover, precise measurements of such type [3] can be used for a determination of complete set of Leslie's coefficients, describing dynamical characteristics of any particular mode realized in the given device. Nevertheless, the information on the anisotropic shear viscosities has been obtained now only for a few LC materials, which is explained by a relatively large (about 10 ml) amount of LC needed for measurements.

Previously, we proposed [4] a new measurement method of anisotropic shear viscosities of liquid crystals based on the decay Poiusellue flow. It was performed in a thin plane capillary with the LC orientation stabilized by the inner surfaces of a capillary contrary to thick capillaries where the quasi-homogeneous orientation of LC samples was provided by magnetic fields [3, 5–7]. That made it possible to measure three principal viscosities of liquid crystals (Miesowicz viscosities) using three LC cells. The LC cells contained plane capillaries with different surface orientation of LC relative to the velocity and velocity gradient. Later we have shown [8] that two Miesowicz viscosities of LC with a positive value of a dielectric permittivity anisotropy can be measured at realization of the decay flow in the same capillary with additional application of electric field. It provided a rather small (smaller than 1 ml) amount of LC and more correct comparison of two anisotropic viscosities obtained in the similar conditions.

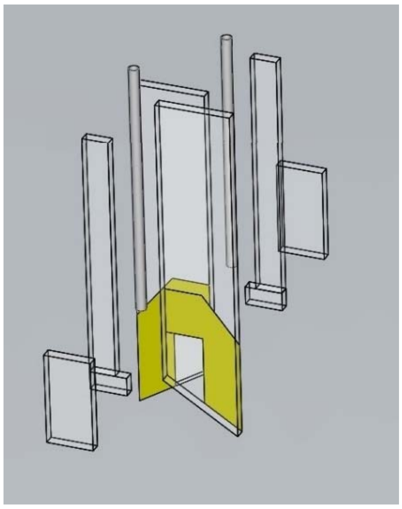
In this paper, we describe results of further investigation of a decay flow in a plane capillary with the orientation controlled by surfaces, electric and magnetic fields. The aim of such study is to estimate a possibility to measure three Miesowicz viscosities in LC sample of amount smaller than 0.1 ml, which is typical for newly synthesized liquid crystal materials.

Experimental

In our experiments we used two liquid crystal cells of the similar construction, shown in Fig. 1. It includes the plane capillary of length L = 4.3 mm and width A = 5 mm with slightly different gap values h = 80 μm and 70 μm for the cell 1 and 2, respectively, which was connected with two open vertical tubes of diameter D = 1,25 mm and length L = 50 mm.



a



b

Fig. 1. Construction of LC cell for viscosity measurements
a – Model of assembled cell, *b* – Exploded view of LC cell

The inner surfaces of the capillary glass substrate were preliminary coated by a thin layer of the azo dye (SD-1) and afterwards illuminated by a polarized UV light to provide a surface orientation of a liquid crystal normal to the flow direction (in accordance with the photo-alignment technique [9]). The transparent ITO electrodes on the glass substrate made it possible to apply a.c. electric voltage to a LC layer formed inside a capillary.

Additionally to electric field, we also used magnetic fields needed to realize three principal orientations of the director (**n**) relatively to the velocity (**v**) and the velocity's gradient (grad **v**) (geometry (1) -**n**⊥**v**, **n**∥grad **v**; geometry (2) – **n**∥**v**, **n**⊥grad **v**; and geometry (3) –

n⊥**v**, **n**⊥grad **v**) needed for determination of the anisotropic shear viscosities η_1, η_2 and η_3 (Miesowicz viscosities) [2]. It is known that the main problem arising at combined action of surfaces and fields during viscosity measurements is connected with experimental errors introduced by inhomogeneous orientation of LC in a layer [2]. In our investigation, we have tried to solve this problem by both the optimization of the geometrical sizes of the cell and the proper choice of the control experimental parameters. In particular, we used the electromagnet FL-1, which produced the magnetic field of induction $B = 1.8$ T (in the geometry 2), and the permanent magnet of induction $B = 0.3$ T in the geometry 3. The orientation of LC in the geometry (1) was tuned using a radio frequency generator, which provided a generation of voltage of the amplitude U_a in the range 0…200 V at the frequency, equal to 1 kHz). The corresponding estimates for different experimental geometries will be presented below.

In our experiments we used a standard nematic liquid crystal 5CB (*4-cyano-4'-pentylbiphenyl*) with the clearing temperature $T_c = 35 \pm 0.2$ °C. All measurements in the nematic phase were carried out at room temperature $T = 24.0 \pm 0.5$ °C.

In the method of decay Poiseuille flow [4], the motion of liquids through a capillary is induced by the difference of a hydrostatic pressure:

$$\Delta P = \rho g \Delta H, \qquad (1)$$

proportional to the density ρ of a liquid and the difference of heights ΔH of meniscuses in the vertical tubes. The difference ΔH decreases with time from the initial value ΔH_0 to zero at approaching to the equilibrium state. In the cases of isotropic Newtonian liquids as well as conventionally anisotropic liquids with a constant value of the shear viscosity coefficient η, the dependence ΔH(*t*) is described by the simple exponential law:

$$\Delta H(t) = \Delta H_0 e^{-t/\tau} \qquad (2)$$

with the decay time τ expressed as:

$$\tau = 2\frac{\eta}{K}, \qquad (3)$$

where $K = K_v \rho g$, K_v – the viscosimeter constant expressed as:

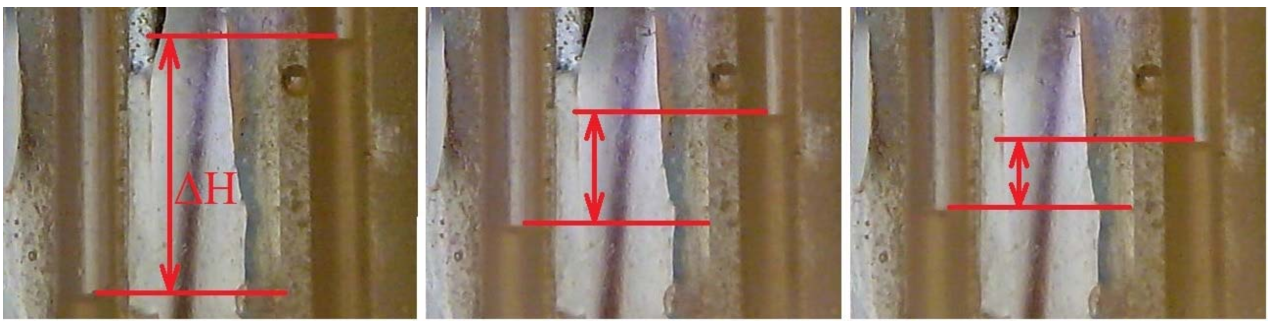
$$K_v = \frac{4}{3}\left(\frac{h^3 A}{L\pi D^2}\right) \qquad (4)$$

So, experimental measurements of a decay time τ can be used to calculate the shear viscosity coefficient in accordance with the next expression:

$$\eta = \frac{\tau K}{2} \qquad (5)$$

In our experiments, the initial difference of meniscus heights ΔH_0 (typically 5…12 mm), was produced by tilting the cell and quickly restoring the vertical position of the cell after some time (about 1…2 min). The motion of the meniscuses was registered by a digital camera. The example of snapshots of meniscuses obtained at different times is shown in Fig. 2. The time dependences ΔH(*t*) was extracted via processing of the digital images.

In the case of a liquid crystal, decay flow can produce the time dependent changes in the initial orientation of LC. The effective coefficient of the shear viscosity obtained in accordance with the above presented equations should reflect such changes in the declination of this parameters from the principal Miesowicz viscosities. Below we will consider such problem for different flow geometries.



a *b* *c*

Fig. 2. The snapshots of meniscuses obtained at different times (*t*);
a) *t* = 0 s, *b*) *t* = 60 s, *c*) *t* = 120 s

Results and discussion

The example of the time dependence of the normalized parameter $\Delta H(t)/\Delta H_0$ in isotropic phase (T = 41 °C), needed for a calibration of the LC cells, is shown in Fig. 3. The processing of the experimental results by the exponential law (2) provided determination of a decay time in isotropic phase (τ_{iso} = 21 s and 40 s for the cells 1 and 2). Usage of this values as well as the results of independent measurements of the shear viscosity (η_{iso} = 0.0205 Pa·s [10]) and density (ρ_{iso} = 999 kg/m^3 [11]) at the same temperature, made it possible to determine values of the constant K_{iso}. The values were equal to $1.95\cdot10^{-3}$ Pa and $1.03\cdot10^{-3}$ for the cells 1 and 2, correspondingly (in accordance with the expression 3). The correspondent value K_N of this parameter in the nematic phase needed for calculation of the effective shear viscosities was determined as:

$$K_N = K_{iso}\left(\frac{\rho_N}{\rho_{iso}}\right) = 1.40\ Pa, \qquad (6)$$

where the density of 5CB in the nematic phase (ρ_N = 1.022 kg/m^3 at T = 25 °C) slightly differs (about 2 %) from the value of this parameter in the isotropic phase.

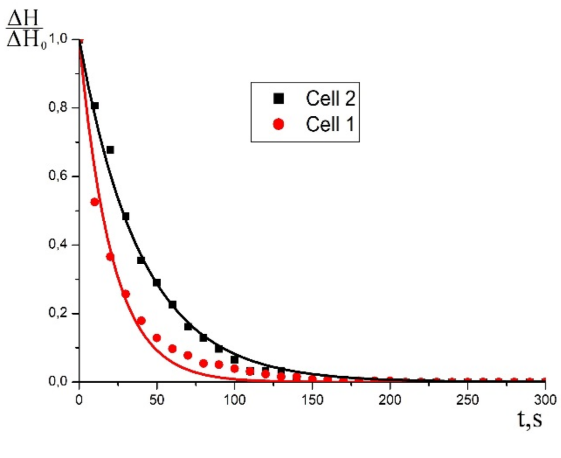


Fig. 3. Time dependences of the normalized parameter $\Delta H(t)/\Delta H_0$ for two cells in isotropic phase (T = 41 °C). Solid lines represent exponential approximations according to (2)

It is well known that in a general case, nematic liquid crystals belong to non-Newtonian liquids showing complicated behavior in shear flows, including the appearance of a number of hydrodynamic instabilities. For LC with a high positive value of the dielectric permittivity anisotropy, electric fields can be considered as the most proper tool for controlling the orientation structure and physical properties of plane thin layers of such materials.

The time dependences of the normalized parameter $\Delta H(t)/\Delta H_0$ for the cell (1) in the nematic phase at different amplitudes of electric voltages U_a are shown in Fig. 4.

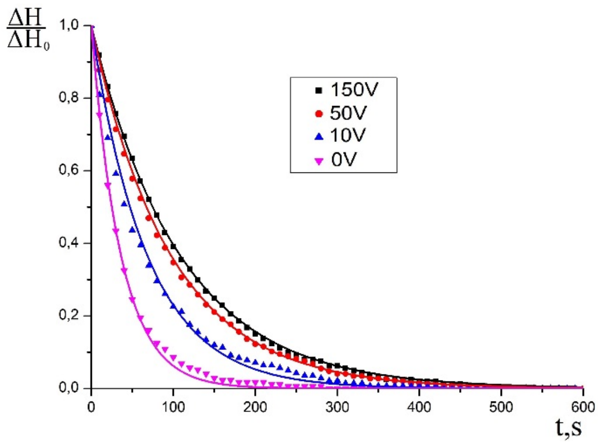


Fig. 4. Time dependences of the normalized parameter $\Delta H(t)/\Delta H_0$ for the cell 1 in the nematic phase for different voltage amplitudes. Solid lines represent exponential approximation according to (2)

The relaxation process slows down with increasing electric voltage. This can be explained by the field induced distortions of the initial planar sample, which lead to an increase in effective shear viscosity. The decay times at various voltages determined by approximating the experimental results are presented in Table 1.

Table 1. **The values of the characteristic decay time τ at different voltages U_a**

U_a, V	0	5	10	15	20	25	30
τ, s	36	45	53	60	70	76	82
U_a, V	60	70	80	90	100	150	200
τ, s	94	97	98	100	102	108	112

The corresponding dependence of the effective shear viscosity η_{eff} on voltage is shown in Fig. 5. At high voltages $U_a \geq 100$ V the saturation of this dependence takes place. This means that the strong electric field suppresses the action of both surfaces and the flow effectively. Indeed, the action of surfaces is restricted by near surface layers with a thickness close to the electric coherence ξ_E with a planar orientation expressed as:

$$\xi_E = \frac{h}{U}\sqrt{\frac{K_{ii}}{\varepsilon_0 \Delta\varepsilon}}, \quad (7)$$

where $U = 0.77U_a$ – the effective value of a voltage, $\Delta\varepsilon$ – the dielectric permittivity anisotropy, K_{ii} – the Frank's elastic module correspondent to the given deformation (in the case of splay deformation described above $K_{ii} = K_{11}$).

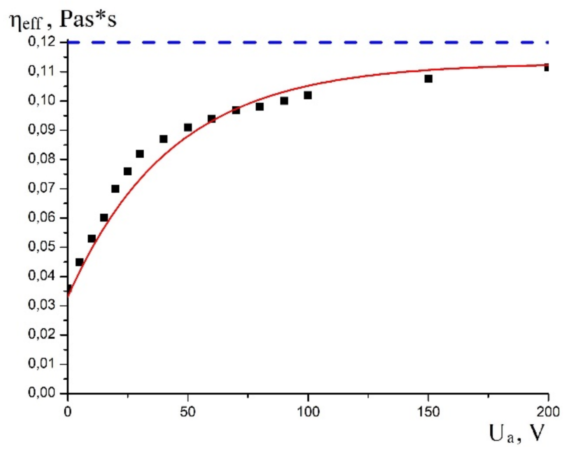


Fig. 5. Dependence of the effective shear viscosity η_{eff} on voltage U_a for the cell 1. The solid line is a simple exponential approximation

The estimate that was made in accordance with the expression (7) at $U_a = 100$ V and material parameters of 5CB $K_{11} = 6.2 \cdot 10^{-12}$ N [12], $\Delta\varepsilon = 11.5$ [12] results in the value $\xi_E \approx 0.3$ µm, which is essentially lower than the thickness of LC layer. At the same time, the calculated value of $\eta_{eff} = 0.111$ Pa·s at $U_a = 200$ V is about 10 % lower than the value $\eta_1 = 0.120$ Pa·s at the same temperature derived from the precise results of the independent measurements [10]. The first mechanism that can explain this difference is associated with the orientation of liquid crystal by the flow [2]. The action of the flow in the central region, excluding boundary layers, can be estimated by considering the flow-induced deviation of the angle θ from the quasi-homeotropic orientation structure stabilized by strong electric field ($E = U/h \approx 10^6$ V/m at $U_a = 100$ V). The value of the angle θ can be derived from the next expression [5]:

$$tg\theta = -\frac{\alpha_2 s}{\varepsilon_0 \Delta\varepsilon E^2}, \quad (8)$$

where α_2 – the Leslie's coefficient ($\alpha_2 = -0.0812$ Pa·s for 5CB [13]). The value of the velocity gradient *s* for a Pouseulie flow is expressed as:

$$s = \frac{dv_x}{dz} = G\frac{z}{\eta_1}, \quad (9)$$

where $G = \Delta P/L$ – the value of the pressure gradient. The maximal value s_{max} of the velocity gradient takes place near the surface ($z \approx h/2$) at the initial stage of the decay flow. For $\Delta P_0 \approx 100$ Pa corresponding to the typical values of $\Delta H_0 = 10$ mm one can get $G_0 \approx 25 \cdot 10^3$ Pa/m and $s_{max} \approx 10$ s^{-1} (at $\eta_1 \approx 0.1$ Pa·s [13]). So, the estimate of the maximal value of the declination angle θ_{max} that was made in accordance with the expression (8) gives

$$\theta_{max} \approx tg\theta_{max} \approx 0.01\, rad. \quad (10)$$

The expression for the dependence of the shear viscosity of oriented LC samples on polar θ and azimuth φ angles reads as [2]:

$$\eta(\theta,\varphi) = \eta_2 cos^2\theta + (\eta_1 + \eta_{12} cos^2\theta) \\ sin^2\theta cos^2\varphi + \eta_3 sin^2\theta sin^2\varphi. \quad (11)$$

Using this expression, it can be concluded that the flow-induced distortions in strong electric fields introduce only a small (about 0.1 %) error in the measured value of the shear viscosity.

It is of interest to estimate the errors introduced into the measured values of the effective shear viscosity by surface layers, defined in accordance with (11) by the changes in the polar angle from 0 (homeotropic orientation) to π/2 (planar orientation). For this, we replace the real flow with shear viscosity, which monotonically depends on coordinates, by a model system consisting of a central layer stabilized by an electric field with the effective viscosity equal to η_1, and two surface layers of thickness ξ_E with the effective viscosity approximately equal to the minimal Miesowicz viscosity η_1 (see Fig. 6). The results of the precise viscosity measurements [10] in 5CB revealed the strong anisotropy of shear viscosities (the ratio η_1/η_2 is about 5 at room temperatures). This means that despite their small thickness, the surface layers provide some slipping of the central part of the flow relatively to the boundaries. As it is well known (see, for example [13]), the slip phenomena can be taken into account by introducing the slip length *L* into the parabolic velocity profile $v(z)$ in the central part of the specific parameter:

$$v_x^{(1)}(z) = \frac{G}{2\eta_1}\left[z^2 - (\frac{h}{2} + L)^2\right] \quad (12)$$

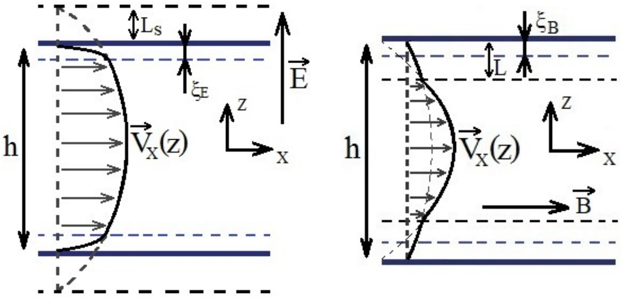


Fig. 6. Flow geometries in the presence of electric (*a*) and magnetic (*b*) fields

In accordance with (12), the slip length is equal to the distance between the real inner surface of the capillary and the imagine boundary at which the flow velocity extrapolated from the central region is equal to zero, as it is shown in Fig. 6, *a*. The expression for L can be obtained from an equality of the velocities $v_x^{(1)}(z)$ and $v_x^{(2)}(z)$, corresponding to the central region and surface layers at the boundaries between these regions (($v_x^{(1)}(z) = v_x^{(2)}(z)$ at $z = h/2 - \xi_E$)). Given the expression for $v_x^{(2)}(z)$:

$$v_x^{(2)}(z) = \frac{G}{2\eta_2}\left[z^2 - (\frac{h}{2})^2\right] \quad (13)$$

one can get in the case of strong field ($\xi_E/h << 1$) the next expression for L:

$$L = \xi_E\left(\frac{\eta_1}{\eta_2} - 1\right). \quad (14)$$

The slip length is proportional to the coherence length and goes to zero at $\eta_1 \to \eta_2$, which coincides with the result obtained for slipping of the pressure-driven flow in the case of strongly hydrophobic surfaces [14].

According to the equation (12), the linear flow velocity increases due to the slip phenomenon. This also leads to an increase in the volumetric flow rate $Q = dV/dt$ (V – the volume pumped through the channel). The final expression for this parameter, obtained by integration the velocity profile over the layer thickness in the strong field approximation, reads as:

$$Q = \frac{Gh^3A}{12\eta_1}\left[1 + 6(\frac{\xi_E}{h})(\frac{\eta_1}{\eta_2} - 1)\right]. \quad (15)$$

Equation (15) shows that the effective value of shear viscosity, obtained from the results of flow in a strong electric field, can be expressed as:

$$\eta_{eff} \approx \eta_1\left[1 - 6(\frac{\xi_E}{h})(\frac{\eta_1}{\eta_2} - 1)\right]. \quad (16)$$

Estimates made in accordance with expression (16) show that the declination of the effective viscosity from the Miesowicz coefficient η_1, induced by the surface layers, can be significant even in the case of relatively strong electric fields. In particular, for the ratio $\eta_1/\eta_2 \approx 5.5$ [10], the values of the relative declination:

$$\frac{\delta\eta}{\eta} = \frac{\eta_1 - \eta_{eff}}{\eta_1}\eta_{eff} \approx 6(\frac{\xi_E}{h})(\frac{\eta_1}{\eta_2} - 1) \quad (17)$$

are close to 10 % and 5 % for U_a = 100 V and 200 V, respectively. These estimates show that the deviation of the experimentally determined values of the effective shear viscosity at high voltages from the precise results of independent measurements of the Miesowicz viscosity η_1 can be partly referred to the influence of slipping. It is also obvious, taking into account (17), that alternative usage of strong magnetic field (B = 1.8 T) instead of an electric field for measurements of Miesowicz viscosity η_1 at the same cell gap may be ineffective. Indeed, in this case, the electric coherence length ξ_E is replaced by the magnetic coherence length ξ_B, defined as:

$$\xi_B = \frac{1}{B}\sqrt{\frac{K_{ii}}{\mu_0^{-1}\Delta\chi}}, \quad (18)$$

where $\Delta\chi$ – the diamagnetic susceptibility anisotropy, equal to = $1{,}43\cdot10^{-6}$ for 5CB [13].

So, with the value of B = 1.8 T, the magnetic coherence length ξ_B = 1.3 μm. Taking this value, it is possible to obtain from the equation (17) $\delta\eta/\eta$ = 0.44, which corresponds to the large (about 50 %) error.

The influence of magnetic field on the dynamics of a decay flow in the geometries 2 and 3 are illustrated by Fig. 7. The estimate of the influence of slipping on the effective viscosity for the geometry 2 can be made in the same way as described above for the geometry 1. In this case, the application of a magnetic field results in twist deformation and one has to replace the constants K_{11} and the ratio η_1/η_2 in equations (17), (18) by K_{22} and η_2/η_3. For 5CB the ratio η_2/η_3 is about -1.6. It means that contrary to the geometry (1), the surface layers provide an anti-slip effect, shown in Fig. 6, *b*, which results in increasing of the effective shear viscosity combined with the essential decreasing in the absolute declination value described by the equation (17). Some decreasing of this parameter also takes place due to the difference between $K_{22} = 3.9\cdot10^{-12}$ N and K_{11}. In particular, for B = 1.8 T, the estimate of the error results in the value $\delta\eta/\eta$ = -0.08, which corresponds to the typical errors at measurements of η_1 using strong electric fields in geometry 2. In particular, the approximation of the experimental dependence $\Delta H(t)/\Delta H_0$ shown in Fig. 7 by exponential law (2) provides the calculation of the effective shear viscosity using the measurements results in the isotropic phase. The obtained value of the effective viscosity η_{eff}= 0.020 Pa·s coincides with the results of precise measurements η_2 = 0.0220 Pa·s at the same temperature with an error equal to 10 %.

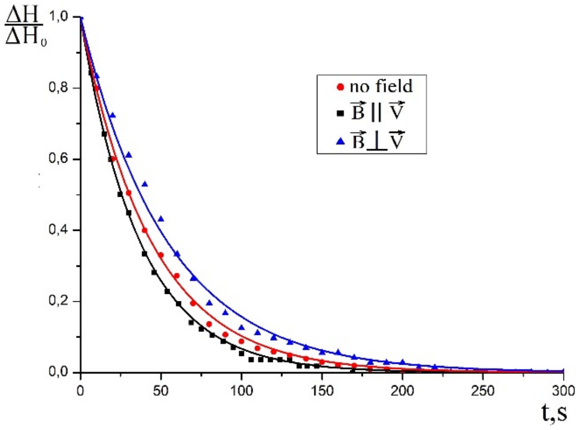


Fig. 7. Time dependences of the normalized parameter $\Delta H(t)/\Delta H_0$ for the cell 2 obtained in the absence and in the presence of a magnetic field in different geometries. Solid lines represent exponential approximations according to (2)

Our attempt to make similar measurements in the third principal geometry using the same cell as in the geometry 2 met some problems. It is known that the initial planar orientation, perpendicular to the flow plane, corresponding to the Miesowicz coefficient η_3, is stable at weak shear flows and undergoes to the twisted structure at certain critical pressure gradient G_c, which can be calculated from the following expressions [2]:

$$Er_c = \frac{G_c}{\eta_3}\sqrt{\frac{\alpha_2\alpha_3}{K_{11}K_{22}}}\left(\frac{h}{2}\right)^3, \qquad (19)$$

where Er_c= 17.1 – the theoretical estimate of the critical value of the Ericksen number, α_3 of the Leslie coefficient equal to 0.0036 Pa·s for 5CB. An estimate made in accordance with (19) for the thickness h = 80 µm, gives the value $G_c = 2.5\cdot10^3$ Pa/m, which results in the critical pressure difference ΔP_c = 10 Pa, applied to the cell. This means that the uniform planar orientation perpendicular to the flow plate can be realized only at a final stage of the decay flow, when the difference of the meniscuses heights does not exceed 1 mm, which prevents the correct determination of η_3. Some increase in the critical parameters Er_c and G_c can be achieved due to the additional action of the magnetic field, which stabilizes the initial planar orientation. The field induced changes in Er_c is described by the expression [2]:

$$Er_c = F + bF^{2/3} \qquad (20)$$

$$F = \left(\frac{h}{2}\right)^2 \frac{\mu_0^{-1}\Delta\chi B^2}{K}, \qquad (21)$$

where b = 2.55 and K – the elastic module in one constant approximation. Calculation by expression (20) for the value of the magnetic field induction B = 0.3 T used in our experiments, gives Er_c = 43. So, the application of the magnetic field makes possible to increase the critical value of the pressure gradient approximately in 2.5 times which should increase the effective shear viscosity. The time dependences of the parameter $\Delta H(t)/\Delta H_0$, obtained in the absence and in the presence of the magnetic field in geometry 3, shown in Fig. 7, correspond to the above estimates. In particular, both values of the effective shear viscosity are intermediate between the values of Miesowicz viscosities η_2 and η_3. Nevertheless, even in the case of the magnetic field, the obtained value η_{eff} = 0.028 Pa·s is about 20 % lower than η_3. This problem can be overcome by further optimizing the experimental cell and using more powerful fields.

Conclusion

The results of the experimental investigation of a decay flow of the nematic liquid crystal 5CB under the combined action of surfaces and electric (or magnetic) fields are presented. The analysis of the effective shear viscosity as a function of electric voltage revealed that even in the case of strong electric fields, the value of this parameter was lower than the corresponding Miesowicz viscosity η_1. The simple model predicted the slip of the central flow region relative to surfaces was used to explain the difference mentioned above. In general, two Miesowicz viscosities η_1 and η_2 were determined with an error of about 10 %. At the same time, further optimization of the experimental cell is needed for measuring Miesowicz viscosity η_3. The obtained results can be used at elaboration of micro-viscosimeters intended for measurements of the anisotropic shear viscosities of liquid crystals.

Acknowledgements: *This work was supported by Ministry of Education and Science of Russian Federation (Grant № FSFZ-2020-0019). The reported study also was funded by Russian Foundation for Basic Research (RFBR and DFG project № 20-52-12040).*

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Поступила 1.09.2020 г.
Received 1.09.2020
Принята 9.09.2020 г.
Accepted 9.09.2020