

## NEW MAGNETIC METHODS FOR DETERMINATION OF ELASTIC CONSTANTS AND ROTATIONAL VISCOSITY COEFFICIENT IN NEMATIC LIQUID CRYSTALS

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*Se propune o metodă magnetică nouă pentru determinarea coeficientului de vâscozitate în cristale lichide nematice și a raportului constantelor elastice  $K_2/K_1$ . Aceasta constă în studierea fenomenelor de relaxare care au loc când o celulă cu cristal lichid nematic orientată homeotrop și o celulă cu nematic răsucit sunt supuse acțiunii unui câmp magnetic cu o intensitate mai mare decât valoarea critică a tranziției Freedericksz. S-au înregistrat variațiile transmisiei optice în funcție de timp la aplicarea respectiv decuplarea câmpului magnetic. Folosind un model teoretic, s-a estimat numărul de extincții, obținând un bun acord cu datele experimentale. S-au determinat câmpul critic al tranziției Freedericksz, timpul de relaxare, coeficientul de vâscozitate rotativ și raportul constantelor elastice  $K_2/K_1$  iar rezultatele au fost similare cu cele obținute prin alte metode.*

*A new magnetic method for determination of rotational viscosity coefficient in nematic liquid crystals and their elastic constant ratio  $K_2/K_1$  is proposed. It consists in studying the relaxation phenomena occurring when a homeotropically aligned nematic liquid crystal cell and a twisted nematic cell were subjected to a magnetic field with the strength higher than the critical value for Freedericksz transition. Changes in the light transmission were recorded as function of time when the magnetic field was switched on/off. Using a theoretical model the number of extinctions was estimated and good agreement with the experimental data was found. The critical field for Freedericksz transition, the relaxation time when logging on/off the field, the rotational viscosity coefficients and the ratio  $K_2/K_1$  were determined; the results were similar to those obtained by other methods.*

**Keywords:** nematic liquid crystals, light transmission, rotational viscosity, magnetic Freedericksz transitions.

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## 1. Introduction

The researches concerning the phenomena related to mechanical stability of liquid crystals are connected to elastic and viscous properties [1-3]. The rotational viscosity is one of the most important parameter of liquid crystal displays, as the switching time is proportional to it. When a liquid crystal display is subjected to an electric or magnetic field, viscous torques are exerted on the nematic director. Assuming a sample of volume  $V$  and an angular velocity  $\Phi$ , the rotation of the director is coupled by a torque  $M = \gamma\Phi V$  where  $\gamma$  is the rotational viscosity coefficient. Several methods have been proposed to determine this parameter; among them the most important are the following

- Mechanical methods using rotating electric or magnetic fields disturbing the equilibrium into a well oriented liquid crystal layer [4-6].

- Relaxation methods consist in inducing a nonequilibrium orientation of the director, followed by time-constant measurements [7-10]. The relaxation was determined by using optical methods or following changes of some electric parameters.

- Spin resonance or nuclear magnetic resonance allowing the determination of the angle between director and field direction [11-13]

It is the aim of this paper to present a new method for determining rotational viscosity of MBBA and the  $K_2 / K_1$  ratio for MLC-6601.

The article is organized as follows: First, we reviewed the theory connected to the dynamical behavior of a nematic liquid crystal acted by a magnetic field [17]. Second, the experimental materials and set-up are described. Finally the experimental results relating to relaxation time-constants when magnetic field was switched on/off are shown and some physical parameters, including rotational viscosity, are given.

## 2. Theory

The free energy density of a homeotropically oriented nematic cell subjected to a magnetic field (Fig. 1) is given by:

$$f = \frac{1}{2} (K_3 \cos^2 \theta + K_1 \sin^2 \theta) \theta_z^2 - \mu_0^{-1} \chi_a B^2 \sin^2 \theta - \frac{1}{2} \gamma \left( \frac{\partial \theta}{\partial t} \right)^2 \quad (1)$$

where  $\gamma$  is the rotational viscosity,  $\theta$  is the deformation angle due to the applied field and  $\theta_z = \frac{\partial \theta(z)}{\partial z}$ . In Eq.1,  $K_1$  and  $K_3$  are the splay and bend elastic constants, respectively. The last term in Eq.1 corresponds to viscous torques and is considered when dynamical problems are involved.

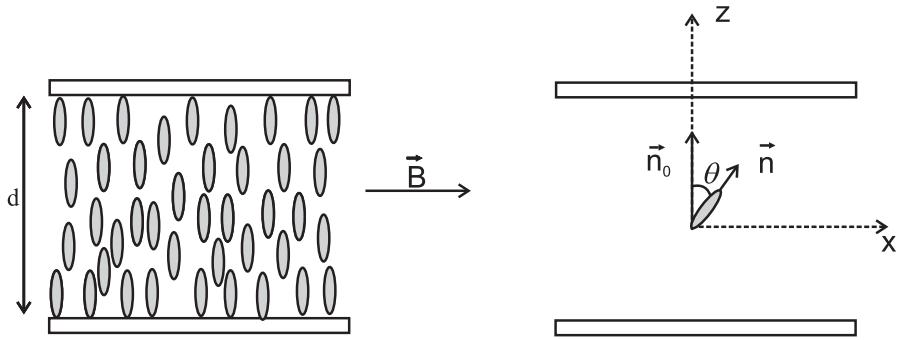


Fig.1 Director orientation in case of a homeotropically aligned nematic cell

The critical field for magnetic Fredericksz transition in homeotropically aligned cells is

$$B_c^2 = \frac{\mu_0 K_3}{\chi_a} \left( \frac{\pi}{d} \right)^2, \quad (2)$$

where  $d$  is the LC cell thickness,  $K_3$  the splay elastic constant,  $\chi_a = \chi_{\parallel} - \chi_{\perp}$  the anisotropy of the pure LC magnetic susceptibility, and  $\mu_0$  the magnetic permeability of the free space.

When the LC cell is subjected to a magnetic field higher than the critical one,  $B_c$ , some fluctuations in the light transmission are noticed. The quasi-periodically oscillations of the transmitted light are observed several seconds and finally the light intensity reaches a standing value. These oscillations appear due to the interference of extraordinary and ordinary rays passing through a birefringent slab. Such phenomena have been investigated in pure nematics by Pieranski et al. [14].

If we consider the light propagating along the Oz axis, the optical path difference between the ordinary and extraordinary rays is

$$l = \int_{-\frac{d}{2}}^{\frac{d}{2}} (n_{ef} - n_o) dz, \quad (3)$$

where  $n_o$  is the ordinary refractive index and  $n_{ef}$  is the effective refractive index and  $d$  the cell thickness. For the homeotropically aligned cell,  $n_{ef}$  is given by [18]

$$\frac{1}{n_{ef}^2} = \frac{\cos^2 \theta}{n_o^2} + \frac{\sin^2 \theta}{n_e^2}, \quad (4)$$

where  $n_e$  is the extraordinary refractive index and  $\theta$  is the LC director distortion angle of nematic director. (see Fig. 1))

As the difference  $n_e - n_o$  is usually small, it is convenient to introduce a new parameter

$$\xi = \frac{n_e - n_o}{n_o} \ll 1 \quad (5)$$

and the extraordinary refractive parameter may be expressed as

$$n_e = n_o(1 + \xi) \quad (6)$$

Considering (6) one obtains

$$\frac{1}{n_{ef}^2} = \frac{1}{n_o^2} \left[ \cos^2 \theta + \frac{\sin^2 \theta}{(1 + \xi)^2} \right] \quad (7)$$

Therefore

$$n_{ef} = n_o \frac{1 + \xi}{\sqrt{(1 + \xi)^2 \cos^2 \theta + \sin^2 \theta}} \quad (8)$$

After neglecting terms containing  $\xi^2$  the relationship (8) becomes

$$n_{ef} = n_o \frac{1 + \xi}{\sqrt{1 + 2\xi \cos^2 \theta}} \approx n_o(1 + \xi \sin^2 \theta) \quad (9)$$

It follows

$$n_{ef} = (n_e - n_o) \sin^2 \theta + n_o \quad (10)$$

and the path difference (3) is

$$l = \int_{-d/2}^{d/2} (n_e - n_o) \sin^2 \theta dz \quad (11)$$

We assume periodic distortions for the nematic director, such as

$$\theta = \theta_m \cos \frac{\pi z}{d}, \theta_m \ll 1 \quad (12)$$

where  $\theta_m$  is the maximum distortion angle in the middle of the cell.

When considering higher order distortions

$$\sin^2 \theta = \left( \theta - \frac{\theta^3}{6} \right)^2 \approx \theta^2 - \frac{\theta^4}{3} \quad (13)$$

and the phase difference is

$$\delta(B) = \frac{\pi d(n_e - n_o)}{\lambda} \left( \theta_m^2 - \frac{\theta_m^4}{4} \right) \quad (14)$$

where  $\theta_m = \theta_m(B)$ .

The transmitted light intensity through the nematic sample is given by

$$I = I_0 \sin^2[2\Phi(B)] \sin^2 \frac{\delta(B)}{2}, \quad (15)$$

where  $\Phi(B)$  is the angle between the molecular director and the direction of incident light polarization. From Eq. (15) it is obvious that the transmitted light intensity will pass through a sequence of minima and maxima when increasing the magnetic field above the critical field for Fredericksz transition. The minima are obtained when

$$\delta(B) = 2\pi N \quad (16)$$

where  $N$  is the extinction order and for high deviations we obtain:

$$N = \frac{d\Delta n}{2\lambda} \left( \theta_m^2 - \frac{1}{4} \theta_m^4 \right) \quad (17)$$

In order to obtain the maximum deviation angle we have to minimize the free energy of the system by solving the Euler-Lagrange equations for the free energy density described in Eq. (1) [10].

After obtaining the Euler-Lagrange equations and considering small deviations of nematic director one obtains the equation

$$(h^2 - 1)\theta_m - \left[ (h^2 - 1) + \frac{K_1}{K_3} \right] \frac{\theta_m^3}{2} = \frac{\gamma}{\mu_0^{-1} \chi_a B_c^2} \frac{d\theta_m}{dt} \quad (18)$$

where  $h$  is the reduced magnetic field, i.e.

$$h^2 = B^2 / B_c^2 \quad (19)$$

The solution of Ec. (18) is

$$\theta_m^2(t) = \frac{\theta_m^2(\infty)}{1 + \left[ \frac{\theta_m^2(\infty)}{\theta_m^2(0)} - 1 \right] \exp\left(-\frac{t}{\tau_A}\right)} \quad (20)$$

where  $\theta_m^2(\infty)$  is the maximum distortion angle when  $t \rightarrow \infty$  and  $\theta_m^2(0)$  is the distortion angle when  $t = 0$ .  $\tau_A$  is the relaxation time intervening when the magnetic field is switched on; this one depends on the rotational viscosity coefficient as follows

$$\tau_A = \frac{\gamma}{2\chi_a \mu_0^{-1} (h^2 - 1) B_c^2} \quad (21)$$

From the expression above, one can determine the rotational viscosity coefficient if the relaxation time  $\tau_A$  and the critical field are known:

$$\gamma = 2\chi_a \mu_0^{-1} (h^2 - 1) \cdot B_c^2 \cdot \tau_A \quad (22)$$

When using a twisted nematic cell, (Fig. 2) the free energy density has a more complex formula due to the molecular rotation along  $Oz$  axis:

$$f = \frac{1}{2} K_1 \cos^2 \theta \cdot \theta_z^2 + \frac{1}{2} K_2 \cos^4 \theta \cdot \varphi_z^2 + \frac{1}{2} K_3 [\sin^2 \theta \cdot \theta_z^2 + \sin^2 \theta \cdot \cos^2 \theta \cdot \varphi_z^2] - \frac{1}{2} \mu_0^{-1} \chi_a B^2 \sin^2 \theta - \frac{1}{2} \gamma \dot{\theta}^2 - \frac{1}{2} \gamma \cos^2 \theta \cdot \dot{\varphi}^2 \quad (23)$$

where  $\theta_z = \frac{\partial \theta}{\partial z}$ ,  $\varphi_z = \frac{\partial \varphi}{\partial z}$ ,  $\dot{\theta} = \frac{\partial \theta}{\partial t}$ ,  $\dot{\varphi} = \frac{\partial \varphi}{\partial t}$ .

The Euler-Lagrange equation for  $\theta$  considering small deviation angle is in this case:

$$\frac{\pi^2}{4d^2} \left[ \theta^3 \left( \frac{8}{3}(\nu+1) - \frac{10}{3}\mu - \frac{8}{3}h^2 \right) + \theta \left( 2\mu - \nu - 1 + 4h^2 \right) \right] + \theta_{zz} (1 + \nu \theta^2) + \nu \theta_z^2 \left( \theta - \frac{2\theta^2}{3} \right) = \frac{\gamma \dot{\theta}}{K_1} \quad (24)$$

where we used the following notation to simplify the expression form:

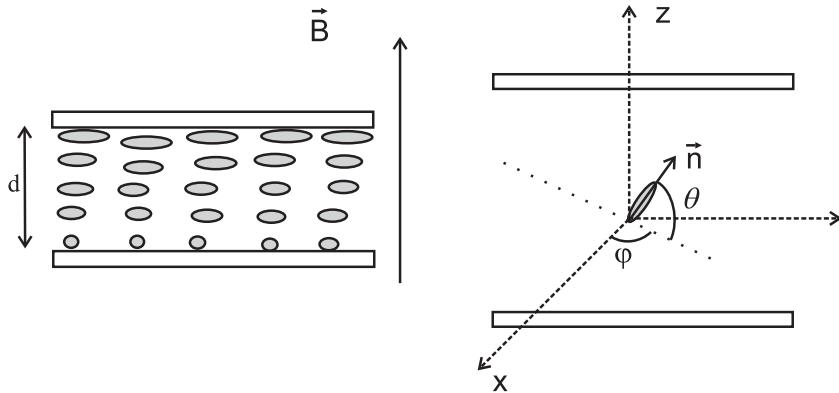


Fig. 2 The director orientation for a twisted nematic cell

$$\frac{K_3}{K_1} - 1 = \nu, \quad \mu = \frac{K_2}{K_1} \quad \text{and} \quad h^2 = \frac{B^2}{\frac{\pi^2}{d^2} \left( \frac{\mu_0 K_1}{\chi_a} \right)} = \frac{B^2}{B_{c, \text{planar}}^2} \quad (25)$$

Eq. (24) has the general solutions:

$$\theta(z, t) = \theta_m(t) \cos \frac{\pi z}{d} \quad (26)$$

where  $\theta_m$  is the maximum deviation angle and  $d$  is the cell thickness.

After integrating over the cell thickness and neglecting high order parameters, the Eq. (24) becomes:

$$p_1\theta_m - p_2\theta_m^3 = \frac{\gamma d^2}{K_1 \pi^2} \frac{d\theta_m}{dt} \quad (27)$$

$$\text{where } p_1 = \frac{\mu}{2} - \frac{\nu}{4} - \frac{5}{4} + h^2 \text{ and } p_2 = \frac{5}{8}\mu - \frac{1}{4} - \frac{h^2}{2}$$

By solving equation (27) one gets:

$$\theta_m^2(t) = \frac{\theta_m^2(\infty)}{1 + \left[ \frac{\theta_m^2(\infty)}{\theta_m^2(0)} - 1 \right] \exp\left(-\frac{t}{\tau_A}\right)} \quad (28)$$

where the relaxation time when the magnetic field is switched on is:

$$\tau_A = \frac{\gamma d^2}{2K_1 \pi^2} \cdot \frac{1}{\frac{1}{4} \frac{K_3}{K_1} - \frac{1}{2} \frac{K_2}{K_1} - h^2 + 1} \quad (29)$$

When the magnetic field is switched off, one has to consider  $h = 0$  in equation (24):

$$\frac{\pi^2}{4d^2} \left[ \theta^3 \left( \frac{8}{3}(\nu+1) - \frac{10}{3}\mu \right) + \theta(2\mu - \nu - 1) \right] + \theta_{zz}(1 + \nu\theta^2) + \nu\theta_z^2 \left( \theta - \frac{2\theta^2}{3} \right) = \frac{\gamma\dot{\theta}}{K_1} \quad (30)$$

Following the same procedure as in the previous case one must consider the solutions are  $\theta = \theta_m(t) \cos \frac{\pi z}{d}$ , integrate over the cell thickness and solve the new found equation leading to a final solution:

$$\theta_m^2(t) = \frac{\theta_m^2(0)}{q\theta_m^2(0) + (1 - q\theta_m^2) \exp\left(\frac{t}{\tau_B}\right)} \quad (31)$$

$$\text{where } q = \frac{\frac{5}{8} \frac{K_2}{K_1} - \frac{1}{2}}{\frac{1}{2} \frac{K_2}{K_1} + \frac{1}{4} \frac{K_3}{K_1} + 1}.$$

In this case one obtains for the relaxation time when the magnetic field is switched off:

$$\tau_B = \frac{\gamma d^2}{2K_1 \pi^2} \cdot \frac{1}{\frac{1}{4} \frac{K_3}{K_1} - \frac{1}{2} \frac{K_2}{K_1} + 1} \quad (32)$$

From the equations above we can determine the ratio:

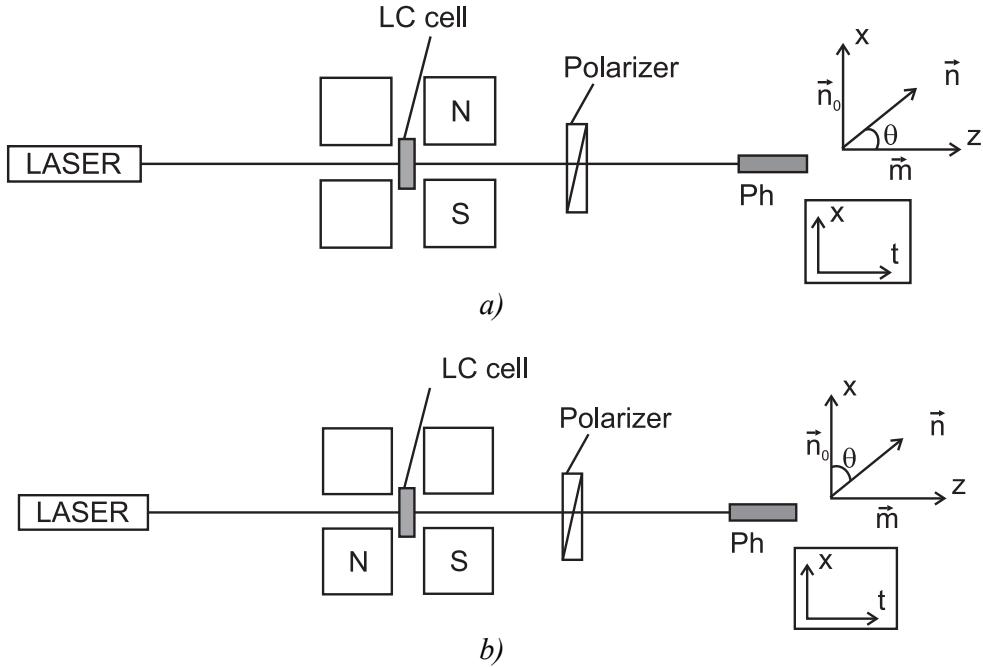


Fig.3 Experimental set-up for registering the dynamic behaviour of the nematic LC subjected to a magnetic field. a) homeotropically aligned cell, b) twisted nematic cell

$$B_{c_{planar}}^2 \cdot \alpha' = B^2 \cdot \frac{1/\tau_B}{1/\tau_B - 1/\tau_A} \quad (33)$$

where  $\alpha'$  is:

$$\alpha' = 1 + \frac{1}{4} \frac{K_3}{K_1} - \frac{1}{2} \frac{K_2}{K_1} \quad (34)$$

From Eq. (34) we obtain the value of the  $K_2 / K_1$  ratio:

$$K_2 / K_1 = 2 \cdot \left( \alpha' - \frac{1}{4} \frac{K_3}{K_1} - 1 \right) \quad (35)$$

### 3. Experimental

#### 3a) Experimental set-up

Liquid crystal cells, with Mylar spacers of 185  $\mu\text{m}$  thicknesses, were filled by capillarity with two nematic liquid crystal: homeotropically aligned MBBA and other one (180  $\mu\text{m}$ ) with MCL-6601 (Merck) with a twisted nematic alignments. Before filling the glass substrates were chemically processed to obtain

a homeotropic alignment of the liquid crystal molecules. For the twisted cell the glass substrates were chemically and mechanically processed in order to obtain the desired alignment.

The experimental set-up is shown in Fig. 3. The LC cell was placed in the middle of the electromagnet (E) provided with hollow poles. The He-Ne laser beam (632.8nm, 1mW) falls at normal incidence on the cell's glass plates. Thus, in this configuration, the magnetic field is perpendicular to the incident light. At the exit, the plane of polarization was determined by rotating a Glan-Thomson polarizer (P) to obtain the extinction of the transmitted light. The intensity of the transmitted light was recorded by means of a photomultiplier (Ph), connected to a system for registering the light intensity as function of time (I-t). Changes in the magnetic field strength were achieved by a DC power supply which allowed both the current adjustment and change of the polarity. The experimental procedure was the following [15,16].

First the polarizer (P) was adjusted to obtain total extinction of the laser beam; then, the LC cell was placed in its proper position, as shown in Fig. 3a.

The experimental device for the homeotropic cell (Fig. 3a) differs from the one used for the twisted nematic cell (Fig. 3b) by the electromagnet position. For the twisted nematic, the electromagnet is 90 degree rotated and the laser beam passes through the holes made inside the electromagnet.

Several magnetic fields with strengths higher than the critical value for magnetic Freedericksz transition were switched on and fluctuations in the light transmission were recorded for each applied field.

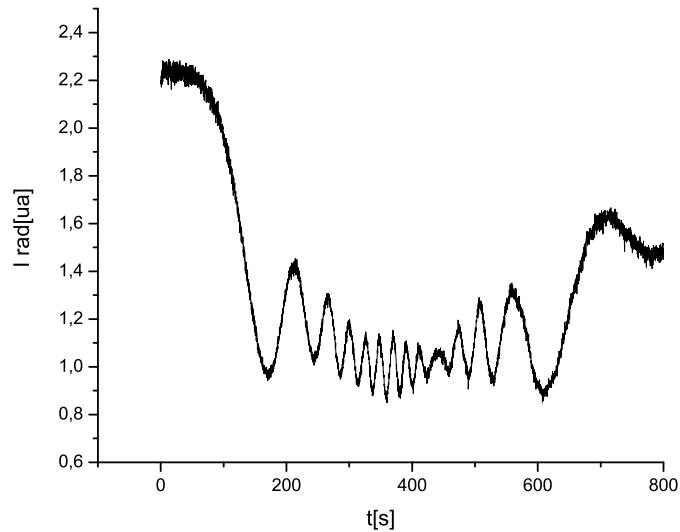
When the homeotropically aligned MBBA was used in order to determine the rotational viscosity coefficient, the field was switched on and the laser intensity versus time plot was recorded (Fig. 4a).

When using the twisted nematic cell, filled with MCL 6601, first, the field was switched on getting the intensity dependence on time similar to the one presented in Fig. 4a, in order to obtain the relaxation time  $\tau_A$ . For the relaxation time  $\tau_B$  needed to determine the elastic constant  $K_2$ , the field must be switched off and the intensity on time dependence is the one presented in Fig. 5a.

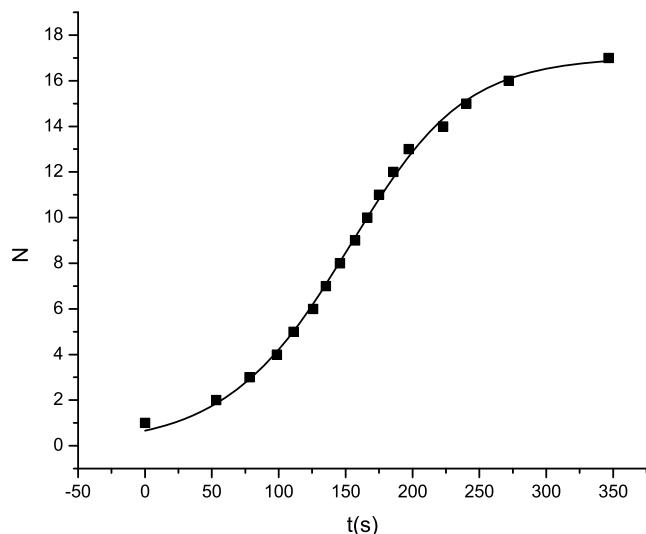
### 3b) Experimental results

In order to fit the experimental data for determining  $\tau_A$  we used the following procedure:

First we determined the number of extinctions considering high deviation angles [15,16].

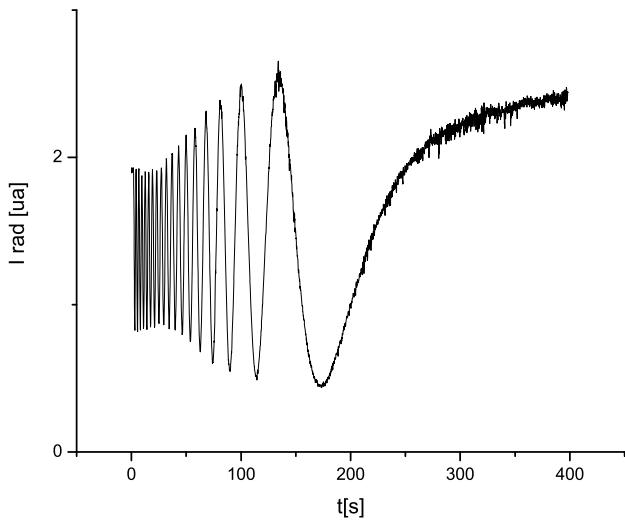


a)

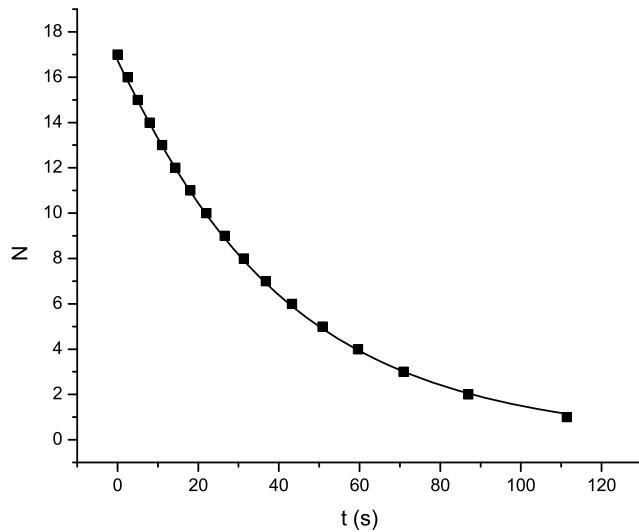


b)

Fig. 4 The fluctuations of the transmitted light versus time when the magnetic field is switched on  
 (a) and the extinction number versus time plot for MBBA (b).



*a)*



*b)*

Fig. 5 The fluctuations of the transmitted light versus time when the magnetic field is switched off (a) and the extinction number versus time plot for MCL 6601 (b).

When the magnetic field was switched on, from (17) and (20) we get

$$N = \frac{d\Delta n}{2\lambda} \left[ \frac{\theta_m^2(\infty)}{1 + \left( \frac{\theta_m^2(\infty)}{\theta_m^2(0)} - 1 \right) \exp\left(-\frac{t}{\tau_A}\right)} - \frac{1}{4} \left( \frac{\theta_m^2(\infty)}{1 + \left( \frac{\theta_m^2(\infty)}{\theta_m^2(0)} - 1 \right) \exp\left(-\frac{t}{\tau_A}\right)} \right)^2 \right] \quad (36)$$

To simplify, this relationship may be written as

$$N = r_4 \left[ \frac{r_1}{1 + (r_3 - 1) \exp(-r_2 t)} - \frac{1}{4} \left( \frac{r_1}{1 + (r_3 - 1) \exp(-r_2 t)} \right)^2 \right] \quad (37)$$

where

$$r_1 = \theta_m^2(\infty), r_2 = 1/\tau_A, r_3 = \frac{\theta_m^2(\infty)}{\theta_m^2(0)}, r_4 = \frac{d\Delta n}{2\lambda} \quad (38)$$

In order to obtain the initial values for fitting we made the following assumptions:

First we assumed  $N = N_{\max}$ , to get the value of  $r_1 = \theta_m^2(\infty)$  as a solution of the equation

$$N_{\max} = \frac{d\Delta n}{2\lambda} \left[ \theta_m^2(\infty) - \frac{1}{4} \theta_m^4(\infty) \right] \quad (39)$$

The maximum deformation angle for  $t = 0$ , i.e.  $\theta_m^2(0)$  was obtained when considering  $N = 1$  and solving the equation

$$1 = \frac{d\Delta n}{2\lambda} \left[ \theta_m^2(0) - \frac{1}{4} \theta_m^4(0) \right] \quad (40)$$

It results from Eq(38) that  $r_3 = \frac{r_1}{\theta_m^2(0)}$ .

From Eq (36) and Eq (37) one can notice that  $r_2 = 1/\tau_A$  and we chose the initial value for fitting 0.01.

The parameter  $r_4$  is fixed as it only depends on cell constants and the laser wavelength used ( $\lambda = 632.8$  nm).

The experimental data obtained for MBBA when the magnetic field was switched on are given in Table I. We used a homeotropic aligned cell with thickness  $d = 185 \mu\text{m}$ . In this case  $\chi_a = 1.27 \times 10^{-7}$ , and  $r_4 = 34$ . The critical value of the magnetic field

( $B_c = 0.137$  T) was measured by known methods (the appearance of the Freedericksz transition when the magnetic field is slowly increased).

Table I

## Experimental data obtained for MBBA

B(T)	$(B/B_c)^2$	$1/\tau_A$	$\theta_m^2/\theta_m^4$	$\theta_m$	$\gamma$ (Pa.s)
0.175	1.63	0.0212	22.6	0.899	0.104
0.212	2.25	0.0435	42.4	1.16	0.105
0.220	2.46	0.0500	47.5	1.20	0.106

As we have already mentioned in the beginning of this paper, the method can also be useful for the evaluation of the elastic constant of the liquid crystal. The elastic constant for the twist deformation ( $K_3$ ) is not usually found in the literature and this method presents an easy way to measure it. The cell used is a twisted nematic one filled with the nematic MCL-6601 from Merck and we must consider both cases: when the field is switched on and off.

When the field is switched off, the fitting function we use is:

$$N = \frac{d\Delta n}{2\lambda} \left[ \frac{\theta_m^2(0)}{\alpha\theta_m^2(0) + [1 - \alpha\theta_m^2(0)]\exp(t/\tau_B)} - \frac{1}{4} \left[ \frac{\theta_m^2(0)}{\alpha\theta_m^2(0) + [1 - \alpha\theta_m^2(0)]\exp(t/\tau_B)} \right]^2 \right]$$

which may be simplified as follows

$$N(t) = r_4 \left[ \frac{r_1}{r_3 r_1 - (1 - r_3 r_1)\exp(r_2 t)} - \frac{1}{4} \left( \frac{r_1}{r_3 r_1 - (1 - r_3 r_1)\exp(r_2 t)} \right)^2 \right] \quad (42)$$

$$\text{where } r_1 = \theta_m^2(0), r_2 = 1/\tau_B, r_3 = \frac{1}{2} \left( 1 - \frac{K_3}{K_1} \right), r_4 = \frac{d\Delta n}{2\lambda}.$$

In this case we have two fixed parameters  $r_3 = -0.25$  and  $r_4 = 11$  while the other two can be evaluated using a similar procedure as in the case the field was switched on. The sample we used was a twisted nematic cell filled with MCL-6601 with the thickness  $d = 180 \mu\text{m}$  for which we know from the provider the following

$$\text{parameters: } \chi_a = 8.28 \times 10^{-8}, r_3 = \frac{1}{2} \left( 1 - \frac{K_3}{K_1} \right) = -0.25, \frac{K_3}{K_1} = 1.51.$$

The results are presented in Table II.

Table II

## Experimental data obtained for MCL-6601

B[T]	$1/\tau_A$ [1/s]	$1/\tau_B$ [1/s]	$\alpha'$	$\frac{K_2}{K_1}$	$K_2 \times 10^{12}$ (N)
0.0518	0.00992	0.0243	1.03153	0.68695	6.25124
0.0544	0.0105	0.0304	1.02847	0.69306	6.30686
0.0571	0.0104	0.036	1.04306	0.66389	6.04138

#### 4. Conclusion

In the relaxation method proposed by us, magnetic fields higher than the critical one for Freedericksz transition were used. Consequently higher order distortions of the nematic are to be expected. This method allowed us to obtain several information such as critical magnetic field  $B_c$ , relaxation time-constants  $\tau_A$  and  $\tau_B$ , elastic constants, sample birefringence and, finally, the viscosity coefficient. All these parameters were determined with a unique experimental set-up and a small amount of substance. The results we get for rotational viscosity are in good agreement with those reported by other investigators when using other methods [19]. The procedure of recording light transmissions gives also interesting insights when changes of birefringence and some characteristic parameters are recorded [20, 21] as well as when investigating laser field effects [22] or ferronematic-ferrocholesteric transitions [23].

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