## Acoustic method for monitoring boundary effects in nematics

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A new acoustic method for estimating the energy representing the anchoring of nematic liquid crystal molecules to a solid substrate in a layer with the homeotropic macrostructure is described and tests are reported. The method is based on a nonlinear steady-state distortion of the orientation of the layer, which depends on the amplitude of the acoustic force and on the anchoring energy. The latter is represented by the amplitude of the vibrations at the first maximum of the constant component of the optical response of the layer to the acoustic force. The method is most effective in the range of low anchoring energies.

Recent years have seen a considerable interest in the surface properties of liquid crystals. Important parameters describing the interaction of liquid crystals with solid walls or substrates is the anchoring energy, the surface order parameter, the long-range radius of the surface forces, etc. If we know these parameters, we can forecast the orientational state of the liquid crystal and control the relevant parameters of devices made of such crystals.

The current situation is that the values of the anchoring energy w of nematic liquid crystals interacting with solid substrates obtained by a variety of methods for the same surfaces, suffer from a large scatter (Ref. 1).<sup>1)</sup> To some extent this is due to the absence of simple and reliable methods of determination of w.

We shall describe a method for estimating the anchoring energy of a nematic liquid crystal in a layer with the homeotropic orientation on the basis of its optical response to a combined effect of acoustic shear and compressive vibrations of the same frequency  $\omega$ , but shifted in phase by  $\pi/2$ .

The method is demonstrated schematically in Fig. 1. It is assumed that one of the plates forming a planar capillary vibrates in accordance with the law

$$v_x|_{z=d} = v_0 \cos \omega t, \quad v_z|_{z=d} = \beta v_0 \sin \omega t. \tag{1}$$

Here,  $v_0$  is the amplitude of the vibrational velocity along the x axis;  $\beta$  is the ratio of the amplitudes of the components of the vibrational velocity along the z and x axes (ellipticity coefficient); d is the thickness of the layer.

The nature of anchoring of nematic liquid crystal molecules to substrate surfaces is described by

$$w\varphi \mp K_{3} \left( \partial \varphi / \partial z \right) \big|_{z=x} = 0, \tag{2}$$

where  $\kappa = 0$  or d;  $K_3$  is the Frank elastic constant;  $\varphi$  is the angle between the director and the normal to the layer.

It is shown in Ref. 2 that in this physical situation we can expect nonlinear interactions of oscillations of the director with the field of velocities of flow of the nematic material, due to vibrations of the plate 1, to create a time-average deviation of the direction from the normal to the layer by an angle  $\varphi_{st}$  depending on the vibration amplitude  $\xi_0 = V_0/\omega$ , on the rotational viscosity  $\gamma$  of the nematic liquid crystal, and on the parameter  $\delta$  which represents the anchoring energy ( $\delta = K_3/wd$ ). These nonlinear interactions are particularly important at amplitudes and frequencies satisfying the inequality

$$0,1\gamma\beta\omega\xi_0 d/K_3 \gg 1. \tag{3}$$

According to Ref. 2, in this situation the constant component of the optical signal is

$$m_0 = 0.5 [1 - J_0(P_1) \cos P_0] I_0, \tag{4}$$

and the parameters  $P_0$  and  $P_1$  governing it are related not only to  $\xi_0$  but also to  $\delta$ . (Here,  $I_0$  is the intensity of linearly polarized light incident on the layer and  $J_0$  is a Bessel function of the first kind.) An analysis shows that if  $P_0 \gg P_1$ , then the first maximum of the function  $m_0 = f(\xi_0)$  satisfies the condition

$$P_0 \approx \pi.$$
 (5)

Since the parameter  $P_0$  depends not only on  $\xi_0$ , but only on  $\delta$ , the position of the first maximum can be used as a measure of the energy of anchoring of nematic liquid crystal molecules to the substrate. This effect can be estimated quantitatively using the complete expression for  $P_0$  obtained in Ref. 2:

$$P_{0} = 0.0075 \Delta n k_{0} d \left(\beta \gamma \omega \xi_{0}^{2} / K_{s}\right)^{2} \left[1 + 0.12 \left(\gamma / \eta_{2}\right)^{2} - 0.67 \left(\gamma / \eta_{2}\right) + f(\delta)\right] + \Delta n k_{0} \xi_{0}^{2} / 2 d.$$
(6)

Here,  $\Delta n$  is the optical anisotropy;  $\eta_2 = (\alpha_4 + \alpha_5 - \alpha_2)/2$ ;  $\alpha_i$  represents the Leslie viscosity coefficients;

$$f(\delta) = [5.5(1+2\delta)^{-1} - 1.65\gamma\eta_2^{-1}] \\ \delta + 11\delta^2(1+3\delta+3\delta^2)(1+2\delta)^{-2}.$$
(7)



FIG. 1. Method used in measurements of the optical response signal: 1) moving plate; 2) fixed plate; 3) layer of a nematic liquid crystal with the homeotropic orientation; 4), 4') Polaroids; 5) light source; 6) to the source of vibrations; 7) to a system for recording the optical response.

Next, in these calculations the parameters of nematic liquid crystals are assumed to be equal to the corresponding values for MBBA:  $K_3 = 7 \times 10^{-7}$  dyn,  $\gamma = 0.78$ P,  $\eta_2 = 1.14$ P,  $\Delta n = 0.2$  (Ref. 2), and  $k_0 = 1.5 \times 10^5$  cm<sup>-1</sup>. The condition (5) then becomes

$$2,79 \cdot 10^{14} d[\beta \omega (\xi_0^{max})^2]^2 [0,6+f(\delta)] + 0,15 (\xi_0^{max})^2/d = \pi, \quad (8)$$

where

$$f(\delta) = (4,73 - 1,54\delta) \,\delta \,(1 + 2\delta)^{-1} + 11\delta^2 \,(1 + 3\delta + 3\delta^2) \,(1 + 2\delta)^{-2}.$$
(9)

Hence, it follows that in the case of nematic liquid crystals of the MBBA type the effect should be detectable only if Eq. (8) includes a considerable contribution of the function  $f(\delta)$ , i.e., if  $f(\delta) \ge 0.3$  and  $\delta \ge 0.2$ . This leads to a change in  $\xi_0^{\max}$  by a factor of at least  $2^{1/2}$ .

We shall now reduce Eq. (8) to a form more convenient for subsequent analysis and divide the terms containing the parameters  $\xi_0^{\text{max}}$  and  $\delta$  as follows

$$[\pi - 0.15(\xi_0)^2/d]/2,79$$

$$\times 10^{14} d(\beta \omega)^2 (\xi_0^{max})^4 - 0.6 = f(\delta).$$
(10)

The graph, calculated<sup>2)</sup> from Eq. (9), of the universal function  $f(\delta)$  presented in Fig. 2 shows how this function behaves when the anchoring energy is varied in the range 10<sup>1</sup>– 10<sup>-5</sup> erg/cm<sup>2</sup> for MBBA samples of the following thicknesses: 5, 15, 90, and 100  $\mu$ m. The proposed method for the determination of the anchoring energy in real samples consists of the following. The value of  $\xi_0^{\text{max}}$  obtained experimentally and the corresponding values of the interaction frequency and of the ellipticity coefficient  $\beta$  can be used to calculate  $f(\delta)$  from Eq. (10) and then the graph in Fig. 2 can be used to identify the value of  $\delta$  appropriate to the relevant experimental conditions.

We tested this method using apparatus described in Ref. 2. Samples representing a mixture of MBBA and EBBA filled a planar capillary formed by plane-parallel glass plates, one of which was fixed and the other exhibited vibrations of the type described by Eq. (1) with different values of the ellipticity coefficient. The vibration frequency and the ellipticity coefficient  $\beta$  were varied within the limits 30– 1500 Hz and 0.01–0.9, respectively. The thickness of the samples was varied from 15 to 100  $\mu$ m. The homeotropic orientation of the molecules of the nematic liquid crystal



FIG. 2. Universal function  $f(\delta)$  calculated from Eq. (9) for the range of values with w from  $10^{-1}$  to  $10^{-5}$  erg/cm<sup>2</sup>, applicable to nematic liquid crystals of the MBBA type.

within the layer with ensured by introducing lecithin into the sample, because, according to Ref. 4, on a glass substrate the anchoring energy should then be  $\approx 10^{-3}$  erg/cm<sup>2</sup>.

In a series of experiments carried out using different values of the ellipticity coefficient  $\beta$ , but fixed values of d, f, and w, we determined the vibration amplitude  $\xi_0^{\text{max}}$  corresponding to the first maximum of the optical transmission of the liquid crystal layer. The results of these experiments are presented in Table I. This table includes the values of the function  $f(\delta)$  calculated from the experimental data using Eq. (10) and also the anchoring energy w deduced from a comparison of these values of  $f(\delta)$  with the universal dependence  $f(\delta)$  plotted in Fig. 2. Clearly, the experimental values of  $\omega$  are very close to the value  $w \approx 10^{-3}$  erg/cm<sup>2</sup> expected for the particular surface treatment of the capillary plates. The observed deviations are due to the errors in the determination of  $\xi_0^{\text{max}}$  and possible errors in the theoretical estimates are related to the inaccuracy of the values of the physical constants of nematic liquid crystals given in the published literature.

This investigation showed that the optical response of a homeotropic layer of a nematic liquid crystal subjected to a

TABLE I. Estimates of the anchoring energy of molecules by an acoustic method.

| <i>d</i> , µm                                       | <i>f</i> , Hz  | β  | $\xi_0^{\max}, \mu m$  | $f(d, \omega, \beta, \xi_0^{max})$  | w, erg/cm <sup>2</sup>  |
|---|--|--|--|---|---|
| Experimental results                                |  |  |  | Calculated using<br>Eq. (10)  | Results of comparison<br>with universal<br>function $f(\delta)$   |
| 15<br>15<br>50<br>50<br>50<br>50<br>50<br>90<br>100 | $ \begin{array}{r}     32 \\     90 \\     34 \\     82 \\     90 \\     151 \\     1468 \\     90 \\     170 \\   \end{array} $ | $\begin{array}{c} 0.1 \\ 0.018 \\ 0.07 \\ 0.025 \\ 0.01 \\ 0.05 \\ 0.9 \\ 0.014 \\ 0.03 \end{array}$ | $     1.8 \\     1.01 \\     1.56 \\     3.4 \\     4.5 \\     1.58 \\     1 2 \\     5 \\     1 7 $ | $15.84 \\ 0.7 \\ 0.134 \\ 0.97 \\ 1.386 \\ 1.541 \\ 1.58 \\ 0.2788 \\ 0.1822$ | $5 \cdot 10^{-4}$ $7 \cdot 10^{-4}$ $5 \cdot 10^{-3}$ $8 \cdot 10^{-4}$ $7 \cdot 10^{-4}$ $6 \cdot 10^{-4}$ $6 \cdot 10^{-4}$ $1 \cdot 3 \cdot 10^{-3}$ $2 \cdot 3 \cdot 10^{-3}$ |

combined acoustic force made it possible to determine very simply the anchoring energy of nematic liquid crystal molecules interacting with a solid substrate. It was sufficient to find the value of  $\xi_0^{\max}$  corresponding to given values of  $d, \beta$ , and f. These measurements could have been carried out by traditional acoustic methods such as those employing accelerometers. Then, using the theoretical universal function  $f(\delta)$  in Fig. 2 and the experimental value of  $\xi_0^{\max}$  we can find the required value of  $\delta$  and finally  $\omega$ . The method proposed above is more effective in the case of low anchoring energies.

It should be stressed that determination of changes in the values of  $\xi_0^{\max}$ , used by us to estimate the anchoring energy  $\omega$ , is not the only way of monitoring this energy by the acoustic method described above. For example, one could measure directly the steady-state angle  $\varphi_{st}$  representing the tilt of the director and governing the value of  $\xi_0^{\max}$ , if use is made of the familiar conoscopic method for examination of a layer of a nematic liquid crystal in a converging beam of polarized light<sup>5</sup> or at an angle to the normal, as was done in Ref. 6 in an investigation of the change in the orientational state of ferroelectric liquids crystals. The latter method is characterized by a very high degree of precision of determination of the angle  $\varphi_{st}$  on the basis of, for example, vanishing of the first harmonic in the optical-response signal.

It should be pointed out that the method of creating a steady-state distortion of the homeotropic macrostructure of a nematic liquid crystal, depending on the anchoring energy, may differ from the "elliptic shift" method discussed above.<sup>2</sup> The same result is produced by steady-state Couette or Poiseulle flow of a nematic liquid in a planar capillary,<sup>7</sup> when the optical response amplitude again depends strongly on  $\delta$  if the experimental conditions are such that the inequality  $\delta > 0.1$  is obeyed.

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<sup>&</sup>lt;sup>1)</sup> According to Ref. 1, the values of w obtained by the familiar experimental methods (Frédericksz transition, flexoelectric effect) for identical surfaces and nematic liquid crystals vary within the range  $10^{-5}-10^{-1}$ erg/cm<sup>2</sup>.

<sup>&</sup>lt;sup>2)</sup> It follows from Eq. (7) that in the ranges  $\delta \leq 1$  (strong anchoring) and  $\delta \geq 1$  (weak anchoring) the calculations can be carried out using the following approximate relationships:  $f(\delta) \approx 4.73\delta + 11\delta^2$  and  $f(\delta) \approx 5.6 + 7.5\delta + 8\delta^2$ . The exact expression (9) has to be used only in the intermediate range of values corresponding to  $\delta \sim 1$ .

<sup>&</sup>lt;sup>5</sup>M. Born and E. Wolf, *Principles of Optics, 4th ed.*, Pergamon Press, Oxford (1970).