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DYNAMIC HYSTERESIS DURING THE REORIENTATION OF NEMATIC LIQUID CRYSTALS IN PULSED FIELDS*

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It is shown that use of the Lavrent'ev-Ishlinskii method in the study of real optical cells based on nematic liquid crystals, enables the experimentally observed deviations in the nature of light transmission by the optical cells at the leading and trailing front of a strong magnetic field acting on the liquid crystal to be explained. The field has the form of a square wave pulse of considerable duration.

1. At a certain critical value value $(H_* \neq 0)$ of the homogeneous, static magnetic field strength H, loss of stability of the homogeneous orientation of the director vector field $\mathbf{n}$ $(\mathbf{n} \perp \mathbf{H})$ of a nematic liquid crystal (NLC) is possible in an optical cell in which the distance L between its two plane parallel glass sides is fixed. Such an idealized theoretical scheme of a static experiment was first suggested in /1/, where the framework of the continual theory of elasticity of NLC was used to achieve the maximum simplification of the Frederiks transition, and attention was drawn to the analogy with the classical problem of mechanics, i.e. the bulging of a thin Eulerian rod.

We know /3/ that the application to the cell of a pulsed control signal of rectangular shape (in the form of an electric or magnetic field) of sufficient duration and magnitude, leads to considerable difference in the nature of light transmission by the cell at the switching on and switching off stages (dynamic hysteresis). This phenomenon has, so far, received no satisfactory explanation.

The prupose of this paper is to show that the phenomenon in question can be explained by analysing the dynamic forms of loss of stability in elastic systems /4/.

2. We choose a Cartesian coordinate system $Oxyz$ so that the boundary separating the mesophase from the surfaces of the sides of the optical cell in contact with it, lies in the $z = 0$ and $z = L$ planes with respect to which the vectors H and n are coplanar everywhere.

In the Zocher scheme /1/ the fixed boundary conditions

$$\mathbf{n}(x, y, 0) = \mathbf{n}(x, y, L) = (1, 0, 0) \tag{2.1}$$

determine the initial (homogeneous) volume orientation of the director $\mathbf{n}(x, y, z) = (1, 0, 0), z \in (0, L)$. In the static magnetic field $\mathbf{H} = (0, H, 0)$ it remains undistorted as long as

$$H < H_*;\ H_* = (\pi/L)(K_2/\chi_a)^{1/2}.$$

Here K_2 is Frank's constant of torsional elasticity and χ_a is the anisotropy of the magnetic susceptibility of unit volume of the mesophase.

When $H \geqslant H_*$, NLC can undergo deformations of the pure-torsion type of the director $\mathbf{n}(x, y, z) = [\cos\varphi(z), \sin\varphi(z), 0]$. In the middle layer of the mesophase $(z = L/2)$ the maximum deviation of the orientation of the director from its initial position is physically justified. Therefore, taking into account (2.1) we have

$$\varphi(z) = \sum_{m=1}^{\infty} a_{2m-1} \sin\left[(2m-1)\pi\frac{z}{L}\right],\quad a_{2m} = 0. \tag{2.2}$$

The amplitudes a_{2m-1} have the same sign, and their magnitude depends on the strength of the applied magnetic field /1/.

The invention of the modern optical (conoscopic) method of recording deformations made the determination of experimental values of the elastic constant K_2 and torsional viscosity γ_1 /5/ a relatively simple matter. The passage through the optical cell of a divergent beam of monochromatic, linearly polarized light in the form of a cone with Oz axis, may lead /5, 6/ to formation of an optical interference pattern on a screen behind the cell, due to birefringence in the optically uniaxial NLC characterized by two values of the refractive index $n_{e,o} > 1$ and their difference $\Delta n \sim 0{,}2$. The picture obtained is related to the change in the phase delay $\Delta\Phi$ in different directions of the wave vectors k of this light cone within the cell /5/

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$$\Delta\Phi = \text{const}\cdot\left[\left(\frac{\Delta n}{n_0 + n_e}\right)(k_x^2 + k_y^2) - \{(k_x^2 - k_y^2)\langle\cos 2\varphi(z)\rangle + 2k_x k_y\langle\sin 2\varphi(z)\rangle\}\right] \tag{2.3}$$

where the square brackets denote averaging over $z \in [0, L]$.

Since $\Delta n/(n_0 + n_e) \ll 1$, we find that there are no deformations $(\varphi = 0)$ and we have four families of more or less identical hyperbolas corresponding to the lines of equal phase on the screen. The apperance of small deformations described by expression (2.2) leads, basically, to rotation of this pattern about the Oz axis by an angle /6/

$$\delta = \frac{1}{2}\operatorname{arctg}\frac{\langle\sin 2\varphi(z)\rangle}{\langle\cos 2\varphi(z)\rangle}.$$

When a physical experiment is carried out, small deviations from the ideal Zocher scheme are inevitable. For example, the appearance of a small component h of the magnetic field $\mathbf{H} = (h, H, 0)$ leads to blurring of the Frederiks transition, distortions in NLC appear at any non-zero value of $H < H_*$ (this resembles the effect of a transverse load on the behaviour of a thin Eulerian rod which bulges out even before the critical value of the longitudinal load is reached). This error is conveniently characterized by the magnitude of the angle $\gamma = \arcsin(h/|\mathbf{H}|)$. In the optical experiments we can satisfy the condition $\gamma \ll 1$, if a smooth rotation of the cell in its plane in any direction is constructively possible /7/.

In a real optical cell perturbations in the orientation of the director vector field can be expected even when there is no magnetic field present. Some of these perturbations are local in character, arbitrarily small in magnitude (e.g. governed by natural thermal fluctuations), and can be neglected in the initial stages of the investigation. However, an unavoidable global perturbation exists in the form of some small initial twist of the director (which resembles a small initial bending of the Eulerian rod), caused by mechanical error in cell construction. The deviation from mutual parallelism of two sides of the cell by an unknown small angle ψ makes it necessary to replace the ideal boundary conditions (2.1) by

$$\mathbf{n}(x, y, 0) = \left[\cos\left(-\frac{\psi}{2}\right), \sin\left(-\frac{\psi}{2}\right), 0\right], \quad \mathbf{n}(x, y, L) = \left[\cos\left(\frac{\psi}{2}\right), \sin\left(\frac{\psi}{2}\right), 0\right].$$

As a result /6/ the director undergoes a small twist within the volume of the NLC

$$\mathbf{n}(x, y, z) = [\cos\vartheta(z), \sin\vartheta(z), 0]; \quad \vartheta(z) = \frac{\psi}{L}\left(z - \frac{L}{2}\right).$$

This perturbation may lead to an appreciable difference between the dynamic and quasistatic reorientation of the NLC, resulting in the appearance of dynamic hysteresis.

3. Let the pulsed magnetic field $\mathbf{H}$ whose magnitude exceeds that of H_* $(|\mathbf{H}| = \mu H_*)$ severalfold, act on the NLC for such a long time, that all previous transients have long since ceased and the director in the middle layer of the cell $(z = L/2)$ deviates by a considerable angle $\varphi \lesssim \pi/2$. In this case the assessment of the deformation relaxation processes (after the instantaneous switching off of the field $\mathbf{H}$) need not account for small shifts in the orientations caused by errors in the form of angles $\gamma \ll 1$ and $\psi \ll 1$ in the experimental set-up (in exact experiments the values of these angles are assumed to range from several degrees to fractions of these degrees /8/).

In the dynamic theory of NLC due to Oseen /2/ the motion of the director, after the magnetic field $\mathbf{H}$ has been fully switched off at the instant $t = 0$, is obtained from the equation of mechanical moments which, when projected on to the Oz axis, has the form

$$\rho d_0^2\frac{\partial^2\varphi}{\partial t^2} + \gamma_1\frac{\partial\varphi}{\partial t} - K_2\frac{\partial^2\varphi}{\partial z^2} = 0. \tag{3.1}$$

Here ρ is the density of the NLC and d_0^2 is the square of the director vector field radius of inertia per unit volume of the mesophase. No reliable experimental data concerning the magnitude of d_0 exist. In /9/ it is regarded as a molecular characteristic. As it is small, it is suggested that the first term of this equation should be neglected when computing the deformation relaxation time.

Experiments show /5/ that since the viscosity γ_1 is very large, the equilibrium states of the director in the optical cell are attained after switching off the magnetic field not in an oscillatory, but in a limiting manner. The interference pattern observed (in the form of hyperbolas) changes the angle of rotation $\delta(t)$ continuously from some initial value $\delta(0) \neq 0$ to the final value $\delta(\infty) = 0$. Therefore /10, 11/ it is correct to assume that $d_0 \approx 0$. In this case the part of the solution of the initial momentum equation decaying most slowly with time, is practically identical with the exact solution of the truncated equation. As a result, when the initial conditions are (2.2), the following approximate relations hold for the spectral components $\{a_n(t)\}$ $(n \in N)$ of the distortions:

$$a_{2m}(t) = 0, \quad a_{2m-1}(t) = a_{2m-1}(0) \exp\left[-\frac{(2m-1)^2 t}{\eta}\right], \quad \eta = \frac{\gamma_1}{\chi_a H_*^2}.$$

When a strong magnetic field $\mathbf{H}$ is switched on, the situation may become completely different.

We know /4/ that when elastic systems are pulse-loaded, it is essential that small, but very important physical factors which determine, to an appreciable degree, the character of development of the processes when the systems depart from the state of equilibrium, be taken into account. For this reason, when the motion of the director following the instantaneous $(t = 0)$ switching on of a strong magnetic field $\mathbf{H}$ $(|\mathbf{H}| = \mu H_*, \mu \gg 1)$ is considered, we must take into account the global errors in the experimental set-up characterized by small angles ψ and γ. When $t > 0$, the director is subjected to a forced reorientation force from the direction of the mechanical torsional moment which is equal, in the classical approximation /1, 2/, to

$$\Gamma = \chi_a (\mathbf{n}\cdot\mathbf{H})[\mathbf{n}\times\mathbf{H}].$$

During the development of distortions the director has the following components:

$$n_x = \cos[\vartheta(z) + \varphi(z, t)], \quad n_y = \sin[\vartheta(z) + \varphi(z, t)], \quad n_z = 0$$

and we assume here that

$$\varphi(z, t) = \sum_{n=1}^{\infty} a_n(t) \sin\frac{n\pi z}{L} \tag{3.2}$$

since we must take into account all types of possible motions corresponding to the boundary conditions, namely, rigid locking of the director orientation on the substrates in the $z = 0$ and $z = L$ planes, under the initial conditions $a_n|_0 = 0, \; da_n/dt|_0 = 0$.

After the instantaneous switching on of the magnetic field, the dynamic equation projected on the Oz axis has the form in which the left-hand side of (3.1) is equal to Γ_z and we have, with an accuracy to terms $O(\gamma)$,

$$\Gamma_z = \chi_a \mu^2 H_*^2 \{{}^1/_2 \sin 2[\vartheta(z) + \varphi(z, t)] + \gamma \cos 2[\vartheta(z) + \varphi(z, t)]\}.$$

Restricting ourselves to considering short time intervals following the instant of switching the magnetic field on, we can disregard, in the initial phase of motion of the director, the non-linear form of the momentum equation. In this case we obtain, after simple reduction, the following differential equation modelling the initial phase of the motions:

$$\iota \frac{\partial^2 \varphi}{\partial t^2} + \eta \frac{\partial \varphi}{\partial t} - \left(\frac{L}{\pi}\right)^2 \frac{\partial^2 \varphi}{\partial z^2} - \mu^2 \varphi = \mu^2 s(z) \tag{3.3}$$

$$(\iota = \rho d_0^2/(\chi_a H_*^2), \quad s(z) = \gamma + \vartheta(z)).$$

Substituting (3.2) into (3.3) we obtain

$$\iota \frac{d^2 a_n}{dt^2} + \eta \frac{da_n}{dt} + (n^2 - \mu^2) a_n = \mu^2 s_n \tag{3.4}$$

where

$$s_n = \frac{2}{L} \int_0^L s(z) \sin\frac{n\pi z}{L} dz = \frac{4}{n\pi} \times \begin{cases} \gamma, & n = 2m - 1 \\ (\gamma - \psi/2), & n = 2m \end{cases}. \tag{3.5}$$

When the condition $n < \mu$ holds, we can expect the appearance of solutions of (3.4) which inercase without limit

$$a_n(t) = \frac{\mu^2 s_n}{\Delta_n} \left[\frac{\exp(\lambda_n^+ t) - 1}{\lambda_n^+} - \frac{\exp(\lambda_n^- t) - 1}{\lambda_n^-}\right] \tag{3.6}$$

$$\Delta_n = [\eta^2 + 4\iota(\mu^2 - n^2)]^{1/2}$$

since the exponential indices $\lambda_n^{\pm} = \pm(\Delta_n \mp \eta)/(2\iota)$ are positive in one case $(\lambda_n^+ > 0)$, and negative in the other case $(\lambda_n^- < 0)$, and are of no interest.

The remaining components of the spectrum $\{a_n(t)\}$ with numbers $n > \mu$ do not yield solutions which increase, and will therefore not be considered. If we disregard the initial stage of development of the processes, which by modern estimates do not exceed several microseconds, we can justifiably replace the exact solution (3.6) by the following approximate relations:

$$a_n(t) = \frac{\mu^2 s_n}{\mu^2 - n^2} \left[\exp\frac{t}{\tau_n} - 1\right], \quad \tau_n = \frac{\eta}{\mu^2 - n^2} \tag{3.7}$$

Let the condition $\gamma \ll \psi$ hold (as a result of a careful optical alignment of the system in the magnetic field /7/). Then we can put $\gamma = 0, \psi \neq 0$ in (3.5). In this case an interesting physical phenomenon may occur, namely the dynamic inversion of the spectrum of developing distortions, as compared with the quasistatic case. The dynamic spectrum (3.7) will contain components which increase with time without limit only with even numbers $a_{2m}(t)$, while $a_{2m-1}(t) = 0$. Therefore when $t > 0$, we have in (2.3) $\langle \sin 2[\vartheta(z) + \varphi(z,t)]\rangle = 0$, but the quantity $\langle \cos 2[\vartheta(z) + \varphi(z,t)]\rangle$ will decrease monotonically. As a result, the rotation of the optical pattern on the screen will be replaced by a rapid increase in the distance separating the neighbouring hyperbolas, since the adjacent hyperbolas in a single square correspond to the condition of phase delay change by $\Delta\Phi = 2\pi$.

The limiting solution of the physical problem as $t \to \infty$ must be described by the normal spectral set of distortions (2.2) just as in the case of quasistatic rise in the magnetic field to the value μH_*, i.e. when $t \gg \tau_2$. Then we should observe on the screen a family of hyperbolas rotated by a large angle $\delta(\mu H_*)$.

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