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VOLUME EXPANSION MECHANISM OF LASER-INDUCED HYDRODYNAMIC REORIENTATION

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Abstract – Laser radiation absorption mediated hydrodynamic flux provides the strongest mechanism of optically induced reorientation of nematic liquid crystal (NLC). The thermal expansion, resulting from absorption induced heating, causes inhomogeneous flow of the NLC in the capillary which, due to the strong coupling of hydrodynamic and orientational degrees of freedom, leads to NLC reorientation. Laser induced hydrodynamic reorientations in homeotropic and hybrid oriented NLC have been studied theoretically and experimentally. In the case of homeotropic initial orientation of NLC molecules the velocity gradient induces a curvature of director. In the case of hybrid initial orientation of NLC velocity gradient brings about a small increase in curvature when the hydrodynamic flow velocity is directed out of the “flexible ribbon’s” curvature. The “flexible ribbon” reverses its curvature when velocity is directed into “flexible ribbon’s” curvature. This effect may allow detection of infrared radiation at a microwatt level.

Keywords: nematic liquid crystals, hydrodynamics, light-induced reorientation

1. Introduction

Light induced hydrodynamic (LIH) reorientation of nematic liquid crystal (NLC) director was predicted long ago and is comprehensively investigated (see, e.g., [1-4]). As indicated in [1], an extremely small absorption would provide energy sufficient for remarkable NLC director reorientation. The problem was to find a suitable mechanism transforming the absorbed energy into LC molecule deformation energy. There are three main mechanisms of LIH motions: gravity or Rayleigh-Benard mechanism, thermocapillary or Marangoni mechanism, and direct volume expansion mechanism. Excitation of hydrodynamic motions in NLC is of interest because here we are able to stimulate in the system initial perturbations of desirable structure, as well as to control them. Moreover, it is possible to control via light beams such well-known phenomena as Hopf bifurcation [5] and turbulence to turbulence transition [6] in homeotropically aligned NLC. These studies could have important applications in bioconvection in a suspension of phototactic algae [7] and in the investigation of chemically driven hydrodynamic instability [8]. The motion of a DNA molecule in a solvent flow reflects the deformation of a nano/microscale flexible mass–spring structure by the forces exerted by the fluid molecules [9]. The dynamics of individual molecules can reveal both fundamental properties of the DNA and basic understanding of the complex rheological properties of long-chain molecules. In [10] it was shown that laminar thermal convection can drive a chain reaction of DNA replication. The convection was triggered by a constant horizontal temperature gradient, moving molecules along stationary paths between hot and cold regions. This implements the temperature cycling for the classical polymerase chain reaction. Besides direct applications, the mechanism might have implications for the molecular evolution of life.

In the present paper, we will investigate in details the dynamics of the above mentioned direct volume expansion mechanism of LIH motions in homeotropic and hybrid oriented NLC. This mechanism includes the following process. The absorbed light energy heats the NLC causing its thermal expansion. A pressure gradient then results in a Poiseuille flow of the NLC. The latter reorients molecules due to the strong coupling of hydrodynamic and orientational motions in the NLC. In the case of homeotropic initial orientation of NLC molecules the velocity gradient induces a curvature of director. Situation is more critical in the case of hybrid initial orientation of NLC. If hydrodynamic flow velocity is directed out of the "flexible ribbon's" curvature, then velocity gradient brings about a small increase in curvature. The curvature deforms more completely and the deformation increases in time when velocity is directed into "flexible ribbon's" curvature. Thereby director deformation elastic energy increases in time. The "flexible ribbon" reverses its curvature at the time when deformation energy becomes larger than surface anchoring energy.

2. Equations of nematodynamics

To introduce the main ideas in a simple form, it is convenient to consider the hydrodynamical mediated optical reorientation of NLC in the following scheme. Let us consider a horizontally arranged plane capillary cell with a nematic LC (NLC) communicating with a volume of liquid (which may be the same NLC) strongly absorbing light energy (Fig.1). We direct the normal to the cell walls along the z-axis whereas the x-axis is in the cell plane. The origin of the coordinates will be arranged on the bottom left edge of the capillary.

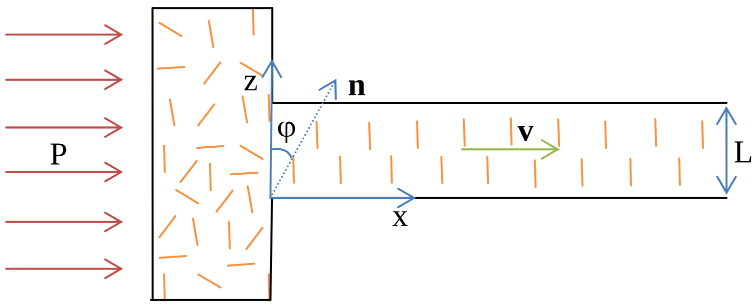

Fig.1. Considered cell: P is the radiation intensity, **v** is the flow velocity, L is the thickness of capillary and **n** is the NLC director.

Let, due to the absorption, the laser radiation heat the cell volume and resulting in the volume expansion. Expansion of the liquid creates a pressure gradient over the cell. To let the liquid flow under this gradient, we will have to assume the presence of a free volume in the cell (e.g., a bubble) where a constant (e.g., atmospheric) pressure is kept. To simplify the problem, let us assume that the pressure gradient is along the x axis, resulting in a Poiseuille flow with $\mathbf{v} = v\mathbf{e}_x$ (it means that $v_y = v_z = 0$), where $\mathbf{v}$ is the velocity of hydrodynamic flows in the cell. We consider the problem homogeneous in the (x, y) plane, so $\partial/\partial x = \partial/\partial y = 0$ and the director lies in the (x, z) plane ($n_y = 0$), where **n** is the director unit vector, with **n** and –**n** equivalent.

The behavior of the considering system, in general, is described by the set of three nonlinear dynamic equations: for the NLC's director reorientation, hydrodynamic motion (Navier-Stokes equation), and thermal conductivity. Two different orientations of the director in the cell – homeotropic and hybrid – can be discussed in the framework of the same approach.

Thus the process starts with the change in the temperature due to absorption, and the linearized thermal conductivity equation has the following form:

$$\frac{\partial T}{\partial t} = r\Delta T + \frac{\chi}{\rho c_{\mathrm{p}}} P, \tag{1}$$

where r is the temperature conductivity coefficient (in cm^2/s), ρc_p is the specific volume thermal capacitance (in erg/cm^3 K), χ is the absorption factor (in cm^{-1}) and P is the radiation intensity (in erg/cm^2 s). The speed of the volume expansion due to the temperature increase is

$$\frac{\partial \mathrm{V}}{\partial t}=\beta \mathrm{V}\frac{\partial T}{\partial t}\,, \tag{2}$$

where V is the volume of absorbing liquid and β is the thermal expansion coefficient (in K^{-1}). The expansion of the liquid creates an overpressure at the input of the capillary in comparison with its outlet, which we will consider as a constant.

In order to describe the above mentioned hydrodynamic effects, we need to write equations of nematodynamics. They are the balance equation of torque acting on NLC director and the Navier-Stokes equation.

The torque balance equations can be obtained from the variation principle [1]:

$$[\mathbf{f}\times\mathbf{n}]_i+e_{ijm}n_m\left[\frac{\delta F}{\delta n_i}-\frac{\partial}{\partial x_k}\frac{\delta F}{\delta\left(\partial n_j/\partial x_k\right)}\right]=0, \tag{3}$$

where e_{ijm} is the whole antisymmetric tensor, $\mathbf{f}$ is the hydrodynamic “force” acting on the NLC director and expressed through the generalized velocities $\mathbf{N}$ and the velocity-gradient tensor d_{ij}:

$$f_i=\left(\alpha_3-\alpha_2\right)N_i+\left(\alpha_3+\alpha_2\right)d_{ij}n_j\,, \tag{4}$$

$$N_i=\frac{dn_i}{dt}+\frac{1}{2}\left[\mathbf{n}\times rot\mathbf{v}\right]_i \qquad d_{ij}=\frac{1}{2}\left(\frac{\partial \mathrm{v}_i}{\partial x_j}+\frac{\partial \mathrm{v}_j}{\partial x_i}\right), \tag{5}$$

Here F is the free-energy density in its usual Frank’s form:

$$F=\frac{1}{2}K_1\left(\mathrm{div}\mathbf{n}\right)^2+\frac{1}{2}K_2\left(\mathbf{n}\cdot\mathrm{curl}\mathbf{n}\right)^2+\frac{1}{2}K_3\left[\mathbf{n}\times\mathrm{curl}\mathbf{n}\right], \tag{6}$$

where K_i are Frank’s elastic constants (in erg/cm), and α_i are Leslie coefficients of the NLC.

The Navier-Stokes equation for the hydrodynamic flow velocity $v(r,t)$ of an incompressible NLC, with the presence of the above mentioned hydrodynamic pressure, is of the form:

$$\rho\frac{\partial \mathrm{v}}{\partial t}=-\frac{\Delta p}{l_0}+\eta\frac{\partial^2 \mathrm{v}}{\partial \mathrm{z}^2}\,, \tag{7}$$

where ρ is the density of NLC (in g/cm^3), $\Delta p/l_0$ is the hydrodynamic pressure gradient (in erg/cm^4), l_0 is the length of the capillary, η is the viscosity constant (in Poise).

Hydrodynamic pressure gradient results from incompressibility condition of NLC, which in our case just means that the increase of the volume of the liquid in one part of the cell is compensated with the flow to the free volume:

$$l\int_0^L \mathrm{v}\left(z,t\right)dz=\frac{\partial \mathrm{V}}{\partial t}\,, \tag{8}$$

where L is the thickness of the capillary and l is its width ($l >> L$).

The stationary velocity v of this flow can be found from equations (1), (2), (7), (8) and has the following form:

$$\mathrm{v}(z) = \frac{6\beta\chi P\mathrm{V}}{\rho \mathrm{c_p} l L^3}\left(z^2 - zL\right). \tag{9}$$

In our case, equation (3) for director $\mathbf{n} = (n_x, 0, n_z)$ has the form

$$K_3 n_z \frac{\partial^2 n_x}{\partial z^2} - K_1 n_x \frac{\partial^2 n_z}{\partial z^2} = \\ = \frac{1}{2}\frac{\partial \mathrm{v}}{\partial z}\left[(\alpha_2 + \alpha_3)(n_z^2 - n_x^2) - (\alpha_3 - \alpha_2)\right] + (\alpha_3 - \alpha_2)(n_z \dot{n}_x - n_x \dot{n}_z). \tag{10}$$

Equation (10) describes reorientation of the NLC director under influence of the hydrodynamic velocity gradient. We denote the angle between the director and z axis as $\varphi(z, t)$; then $n_x = \sin\varphi$, $n_z = \cos\varphi$ and the equation for the reorientation angle, taking into account the expressions (9) for the velocity of hydrodynamic motion of the NLC, has the form:

$$\left(\frac{\partial \varphi}{\partial z}\right)^2 \sin\varphi\cos\varphi\left(K_1 - K_3\right) + \frac{\partial^2 \varphi}{\partial z^2}\left(K_3\cos^2\varphi + K_1\sin^2\varphi\right) = \\ = \frac{6\beta\chi \mathrm{V}}{\rho \mathrm{c_p} l L^3} P(2z - L)\left[\alpha_2\cos^2\varphi - \alpha_3\sin^2\varphi\right] + \frac{\partial\varphi}{\partial t}\left(\alpha_3 - \alpha_2\right). \tag{11}$$

Note that it is mathematically more suitable for numerical calculations to write this equation in the following form:

$$\frac{\partial \phi}{\partial \tau} = \left(K + \Delta\sin^2\phi\right)\frac{\partial^2\phi}{\partial\zeta^2} + \frac{1}{2}\Delta\sin 2\phi\left(\frac{\partial\phi}{\partial\zeta}\right)^2 + DeP\left(2z - 1\right)\left(\alpha\sin^2\phi - 1\right), \tag{12}$$

where our designations are: $K = K_3/K_1$, $\Delta = (K_1 - K_3)/K_1$, $D = \partial u/\partial z = \alpha L^2\alpha_2/K_1$, $u = vL\alpha_2/K_1$, $\alpha = (\alpha_3 + \alpha_2)/\alpha_2$, $\zeta = z/L$, $\tau = K_1 t/(\gamma L^2)$, $\gamma = \alpha_3 - \alpha_2$ and $De = 6\beta\chi V\alpha_2/(K_1\, \rho c_p l)$.

Note that the equation (12) takes the form of the well-known “damped driven sine-Gordon equation” in the single-constant approximation for elastic constants ($K_1 = K_3$).

Equation (12) is generalized equation for the description of LIH flow in a NLC with the director confined to the (x, z) plane.

3. Boundary conditions

For boundary conditions of the orientational angle, often the approximation of strong anchoring is made; it means that the director orientation at the boundary is supposedly fixed and independent of the external excitations. Here we discuss more general case, so we are not considering an infinite anchoring energy, and a surface contribution must be included in the free energy. For this problem, under the Rapini approximation, the boundary conditions have the following form [11]:

$$\left(K_1\sin^2\phi + K_3\cos^2\phi\right)\frac{\partial\phi}{\partial z} - \sigma_1\sin\phi\cos\phi = 0. \tag{13}$$

at the lower wall ($z = 0$) in both cases: when the cell has the homeotropic ($\mathbf{n}_0 = \mathbf{e}_z$)(see Fig. 2) and hybrid ($\mathbf{n}_0 = \mathbf{e}_z$ at $z = 0$ and $\mathbf{n}_0 = \mathbf{e}_x$ at $z = L$) (see Fig. 3) orientation as an initial condition. And at the upper wall $(z = L)$

$$\left(K_1 \sin^2\varphi + K_3 \cos^2\varphi\right)\frac{\partial\varphi}{\partial z} + \sigma_2 \sin\varphi\cos\varphi = 0 \tag{14}$$

for homeotropic oriented cells (see Fig. 2) and

$$\left(K_1 \sin^2\varphi + K_3 \cos^2\varphi\right)\frac{\partial\varphi}{\partial z} - \sigma_2 \sin\varphi\cos\varphi = 0 \tag{15}$$

for hybrid oriented cells as an initial condition (see Fig. 3). In above equations σ_1 and σ_2 are coefficients of surface anchoring energy (in erg/cm^2).

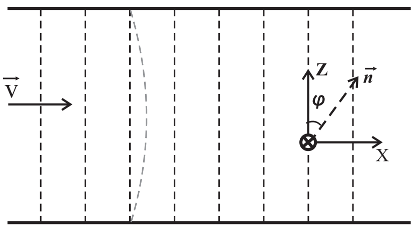

Fig.2. Director profile in a homeotropic oriented cell with nematic LC.

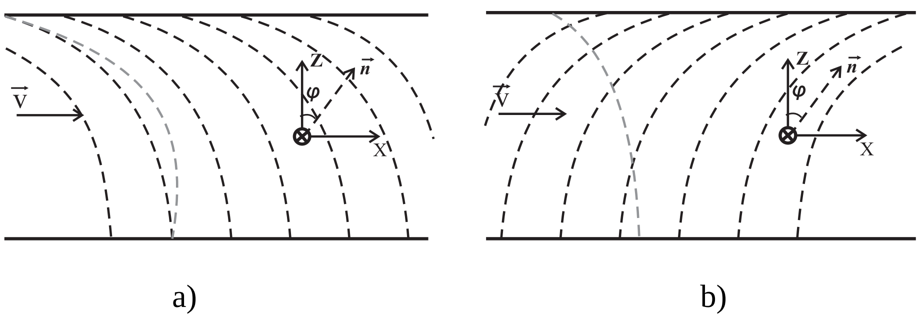

Fig.3. Director profile in hybrid oriented cells with nematic LC.

In the absence of flow the stationary solution of equation (12) with mentioned boundary conditions can be assumed as an initial conditions for the general (non-stationary) problem.

4. Transmission of light, numerical solutions and discussions

Let us discuss a simple scheme of registration of laser induced hydrodynamic director reorientation of NLC in the capillary sandwiched between polarizers. The monochromatic visible wave is incident on this system perpendicularly. With crossed polarizers making a $\pm\pi/4$ angle with respect of the flow direction, the transmission coefficient of the system becomes

$$T = \sin^2\frac{\Delta\Phi}{2}, \quad \Delta\Phi = \Phi_a \frac{\varepsilon_\perp^{1/2}}{L}\int_0^L \varphi^2(z)dz, \quad \Phi_a = \frac{\omega}{2c}L\frac{\varepsilon_a}{\varepsilon_\parallel}, \tag{16}$$

where $\Delta\Phi$ is the additional phase shift (retardation) induced by the reorientation of the NLC, $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$ is the dielectric anisotropy of the NLC at the optical frequency ω and c is the light speed in vacuum.

We solved equation (12) for director reorientation caused by direct volume expansion with the above-mentioned boundary and initial conditions using "Mathematica-5". In this calculation for NLC MBBA we assumed $K_1 = 6{\cdot}10^{-7}$erg/cm, $K_3 = 7.5{\cdot}10^{-7}$erg/cm, $\alpha_2 = -0.8P$, $\alpha_3 = -0.012P$, $\beta = 10^{-3}K^{-1}$, $\rho c_p = 1$J/cm^{3}K, for cell with $V = 1$ cm^3, $L = 10^{-2}$cm, $l = 0.1$cm. We study the reorientation induced by laser radiation with $P = 0.5{\cdot}10^{-3}$W/cm^3 intensity.

Let us discuss separately two different cases.

When the cell has the homeotropic ($\mathbf{n}_0 = \mathbf{e}_z$) initial orientation (see Fig.2), the hydrodynamic flow velocity gradient brings about a continuous increase of curvature. It is important and interesting to discuss our solutions (see Fig.6 and Fig.7) for the generalized problem, with finite anchoring energy between NLC and confining walls, with respect to the solutions for the problem with approximation of strong anchoring (see Fig.4 and Fig.5).

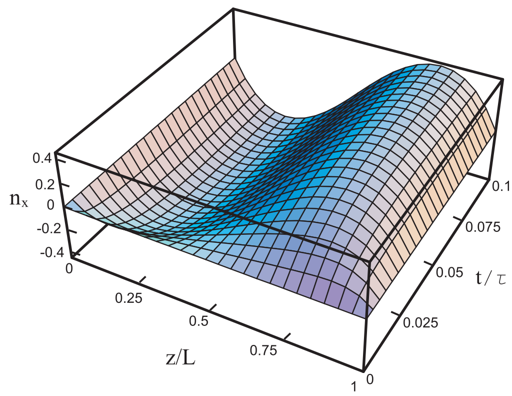

Fig.4. Three-dimensional time and space dependence of the director's x component after switching the laser radiation "on"; for homeotropic oriented cells with an approximation of strong anchoring.

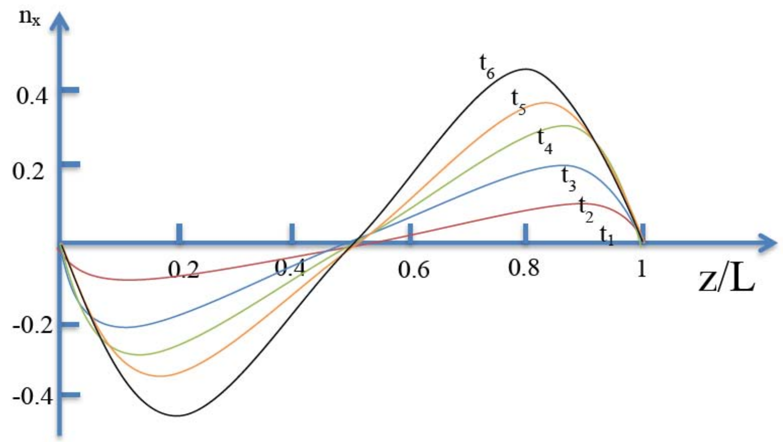

Fig. 5. Profile of the director's x component (n_x) for different times (t_j) after switching the laser radiation "on" for homeotropic oriented cell with an approximation of strong anchoring. ($t_1 = 0$ sec, $t_2 = 2.28{\cdot}10^{-5}$ sec, $t_3 = 0.68{\cdot}10^{-4}$ sec, $t_4 = 1.14{\cdot}10^{-4}$ sec, $t_5 = 2.28{\cdot}10^{-4}$ sec, $t_6 = 2.28{\cdot}10^{-3}$ sec).

In contrast to the strong anchoring, in the general case, the orientation on the walls is changing during the time (see Fig. 6 and Fig. 7). If we discuss finite anchoring energy, after some time the deformation free energy overcomes the anchoring energy and cause the above mentioned change of orientation on the boundaries during the bulk reorientation.

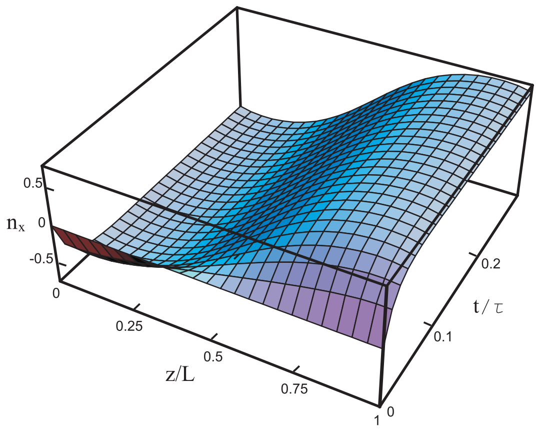

Fig.6. Three-dimensional time and space dependence of the director's x component after switching the laser radiation "on"; for homeotropic oriented cells in the case of the generalized problem.

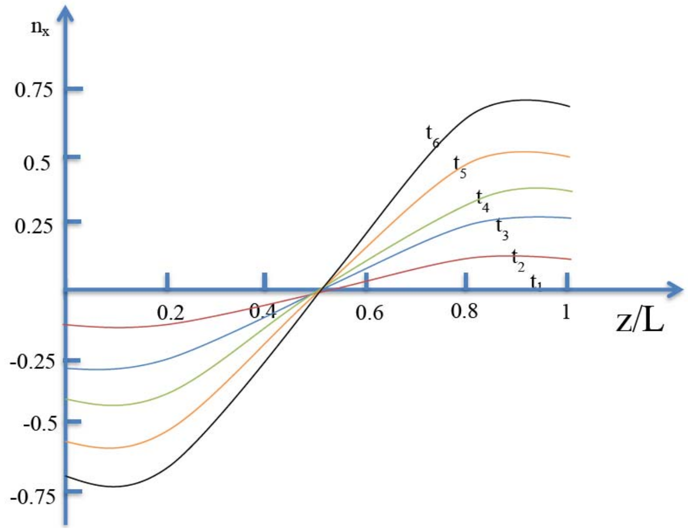

Fig.7. Profile of the director's x component (n_x) for different times (t_j) after switching the laser radiation "on" for homeotropic oriented cell in the case of the generalized problem. (t_1 = 0 sec, $t_2 = 2.28{\cdot}10^{-5}$ sec, $t_3 = 0.68{\cdot}10^{-4}$ sec, $t_4 = 1.14{\cdot}10^{-4}$ sec, $t_5 = 2.28{\cdot}10^{-4}$ sec, $t_6 = 2.28{\cdot}10^{-3}$ sec).

It is more interesting to study laser induced hydrodynamic reorientation of LC with above mentioned mechanism, when the cell has hybrid ($\mathbf{n}_0 = \mathbf{e}_z$ at $z = 0$ and $\mathbf{n}_0 = \mathbf{e}_x$ at $z = L$) orientation as an initial condition. In this case, if hydrodynamic flow velocity is directed out of the "flexible ribbon's" curvature (see Fig.3.a), velocity gradient brings about a small increase of curvature. The curvature deforms more completely and the deformation increases in time when velocity is directed into "flexible ribbon's" curvature (Fig.3.b). As we can see from Fig.8 the deformation free energy initially increase until saturation.

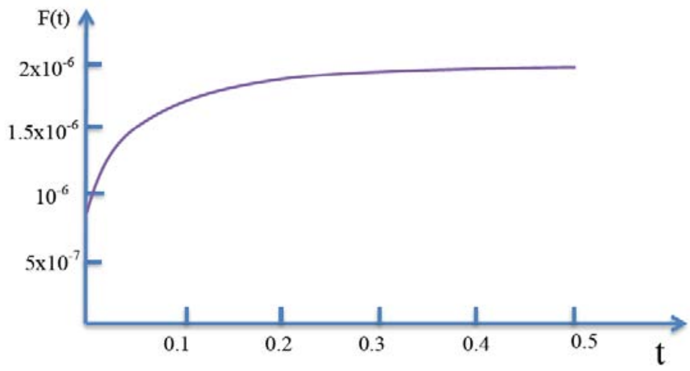

Fig 8. Deformation free energy of the hybrid oriented liquid crystal cells after switching the laser radiation "on".

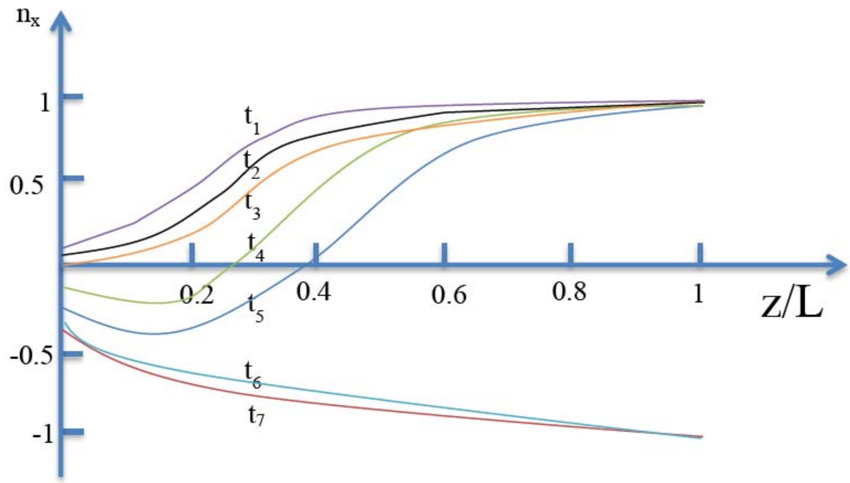

Fig.9. Profile of the director's x component (n_x) for different times (t_j) after switching the laser radiation "on" for hybrid oriented cells in the case of the generalized problem (t_1 = 0 sec, $t_2 = 3.8{\cdot}10^{-5}$ sec, $t_3 = 1.14{\cdot}10^{-4}$ sec, $t_4 = 1.9{\cdot}10^{-4}$ sec, $t_5 = 3.8{\cdot}10^{-4}$ sec, $t_6 = 1.14{\cdot}10^{-3}$ sec, $t_7 = 3.8{\cdot}10^{-3}$ sec). Reversal time is $t_r = 0.68{\cdot}10^{-3}$ sec.

The "flexible ribbon" reverses its curvature (see Fig. 9 and Fig. 10) at the time when deformation energy becomes larger than surface anchoring energy at the wall with planar initial orientation, and takes a form with less deformation energy. In that way hydrodynamic velocity is directed out of the reversed curvature and brings an additional small increase of curvature. The reversing time depends on NLC parameters and surface coupling energy. The latter depends on the method of surface treatment.

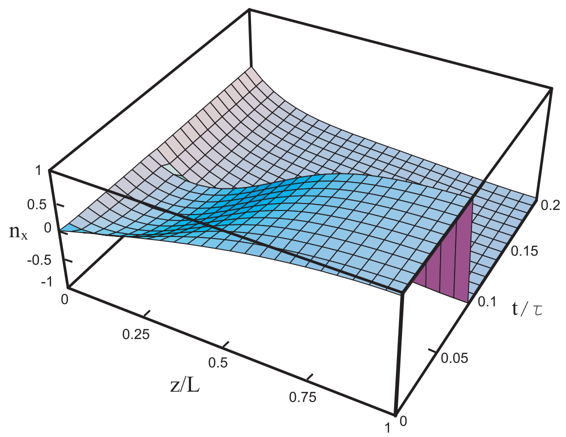

Fig.10. Three-dimensional time and space dependence of the director's x component after switching the laser radiation "on"; for hybrid oriented cells in the case of the generalized problem.

So, this effect brings a correspondence between a monotonous function (incident light power) and jump-like function (the orientation of NLC). It means that this model has a so-called trigger behavior and has the practical meanings.

Acknowledgments

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