Physics

LASER INDUCED THERMOMECHANICAL EFFECT IN HYBRID ORIENTED LIQUID CRYSTAL

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In the paper laser induced thermomechanical flow and oscillations in hybrid oriented nematic liquid crystals (NLC) are theoretically predicted. The effect is conditioned by the hydrodynamic flow tendency to reduce the curvature of the "flexible ribbon" of hybrid NLC.

Keywords: nematic liquid crystals, thermomechanics, heat flux.

Introduction. Shortly after the discovery of liquid crystals (LC) the effect of rotation in the cholesteric drops due to vertical temperature gradient was discovered by Lehmann [1]. For cholesteric LC, thermomechanical effects of such kind were investigated in detail both experimentally and theoretically in [2–10] and attributed specifically to the chirality (or right and left asymmetry) of cholesterics. They concluded that effects of this kind would not be expected to occur in nonchiral LC, such as nematics (NLC) [2, 11]. In [12] was observed thermomechanical effect in compensated cholesterics, when at some temperature the medium containing cholesteric molecules have no chiral structure, i.e. pitch tends to infinity. From these experiments they concluded that thermomechanical effect exist due to the chirality of individual molecules, but not of the entire volume. This approach was criticized in [13–15]. The obtained results were called as a consequence of experimental error. This brings to the long discussion between scientists [16]. Finally, the new experiments and theoretical studies [17–19] proved that experiments were accurate and the authors of [13–15] were wrong as usual. They studied this effect in great depth by considering dependence on the boundary conditions [20], the concentration of chiral molecules [21] and by showing that chirality could be induced due to this effect [22]. Because of the intricacy of the problem, it seemed that these discussions [23] could continue till infinity. But this time it was proved fundamentally and in detail that Lehmann effect or thermo-mechanical rotation in cholesterics could not be explained solely due to the Leslie's thermomechanical connection [24, 25]. Although long before these works, new thermomechanical effects were predicted for deformed nematics in [26], where the first consistent theory of thermomechanical coupling due to nonuniform director

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orientation under a temperature gradient was developed for uniaxial nematics. Thermomechanical effects of three basic types were considered in [26]: hydrodynamic excitation induced by temperature gradient (direct thermomechanical effect), temperature gradient arising in nonuniform flow (inverse thermomechanical effect), and additional director deflection caused by heat flow.

The validity of the thermomechanical terms in the equations obtained in [26] was questioned in [27], where these terms were written in somewhat different form. However, the thermomechanical coupling predicted in [26] was observed in numerous experimental studies [28–30], and the measured values of thermomechanical coefficients were in good agreement with the theoretical estimates obtained in [26].

In [31, 32] they studied thermomechanical hydrodynamic flow and reorientation of director of compressible hybrid-oriented NLC cell under the influence of a temperature gradient directed normal to the restricting surfaces, when the sample is heated both from below and above. The case of the orientational thermoelastic interaction considered in [33], which has not been discussed in the literature earlier, belongs to the effects in open systems and is quadratic in the temperature gradient. This type of interaction was registered experimentally and described in [34]. In [35] have been successfully dispersed gold nanoparticles (AuNP) in thermo-mechanical NLC elastomer actuators and had significantly improved the material response to a thermal stimulus. Embedment of AuNP in NLC stiffened the elastomers, resulting in a slight decrease in the strain of the actuators, but this effect was more than compensated by a significant improvement in the actuator strain rate in response to an external stimulus. Incorporating a fractional amount of metal nanoparticles in these thermomechanical actuators can result in a pronounced enhancement of the thermal conductivity.

In the case when the thermomechanical hydrodynamic flow tends to reverse the curvature of the "flexible ribbon" of hybrid or cylindrical-hybrid oriented NLC, an oscillatory motion was predicted and observed [36, 37]. In the hybrid aligned cell an oscillatory motion was observed. And in the cylindrical-hybrid aligned cell a clockwise and counterclockwise oscillatory rotations were observed and studied.

In this paper, new results on the thermomechanical effect in hybrid oriented horizontal layers of NLC due to a laser-induced vertical temperature gradient has been presented. The possibility of hybrid curvature reversal due to the hydrodynamic flow has been studied theoretically. This model allows us to explain our previous experimental results about oscillatory hydrodynamic flow due to the thermomecha-nical forces at the presence of a laser induced transverse temperature gradient.

Thermomechanical Equations in Hybrid NLC. Let us consider an NLC cell having the so-called hybrid orientation (Fig. 1, 2). We direct the normal to the cell walls vertically upwards and assume that the boundary condition on the wall specifies homeotropic orientation $\mathbf{n}\,(z = 0) = \mathbf{e_z}$ at $z = 0$, where $\mathbf{n}$ is the director unit vector, with $\mathbf{n}$ and $-\mathbf{n}$ equivalent, L is the cell thickness. For the cell with hybrid orientation we have planar orientation $\mathbf{n}\,(z = L) = \mathbf{e_x}$ at $z = L$.

Let us introduce angle θ between director and z axis. Then in the case of hybrid initial orientation $n_x = \sin\theta(z),\ n_y = 0,\ n_z = \cos\theta(z).$ Then the boundary conditions for function $\theta(z)$ will be written in the simple form: $\theta(0) = 0$, $\theta(L) = \pi/2$, where L is the thickness of NLC layer.
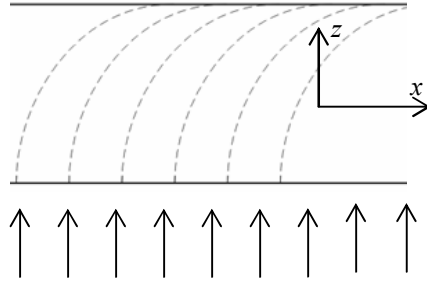
Fig. 1. Director profile in hybrid oriented cells with nematic LC. Laser beam propagate in the direction from homeotropic to planar initial orientation of director or vice versa.

Let the external heat sources (laser radiation) maintain temperatures $T = T_0$ and $T = T_0 + \Delta T$ at the plane $z = 0$ and at the plane $z = L$ respectively. The temperature gradient $dT / dz \sim \Delta T / L$ leads then, according to [26], to tangential thermomechanical stresses $\sigma_{zx(\varphi)}^{thm} \approx \xi \Delta T / L^2$, where ξ is the thermomechanical constant. As a result the liquid flows in the x direction. The stationary velocity **v** in this flow can be estimated equating thermomechanical and Navier–Stokes contributions in stress tensor σ_{ik}. Assuming for the latter $\sigma'_{zx(\varphi)} \approx \eta v / L$, where η is viscosity, we obtain $v \approx \xi \Delta T / L \eta$. This hydrodynamic flow leads to the reorientation of the NLC director. The direction of the flow velocity depends on the director concavity and the temperature gradient direction. If the temperature gradient is directed from the cell wall with planar boundary condition to the wall with homeotropic condition, then thermomechanical hydrodynamic flow tends to increase the curvature of the "flexible ribbon" of hybrid NLC. If the temperature gradient has opposite direction, then flow tends to reverse the curvature of "flexible ribbon". In the latter case thermomechanical stress induced the oscillatory hydrodynamic flow.

In order to describe the above mentioned laser induced thermomechanical effects, we need to write equations of nematodynamics taking into account the thermomechanical stress tensor. They are balance equation of torque acting on NLC director and the Navier–Stokes equation.

Torque balance equations can be obtained from the variation principle [38]:

$$\Pi_{ij} \left[\frac{\delta F}{\delta n_j} - \frac{\partial}{\partial x_k} \cdot \frac{\delta F}{\delta(\partial n_j / \partial x_k)} + f_j \right] = 0, \qquad (1)$$

where $\Pi_{ij} = \delta_{ij} - n_i n_j$ is the projection operator, **f** is the hydrodynamical "force" acting on the NLC director and expressed through the generalized velocities **N** and the velocity-gradient tensor d_{ij} :

$$f_j = (\alpha_3 - \alpha_2) N_i + (\alpha_3 + \alpha_2) d_{ij} n_j, \qquad (2)$$

where

$$N_i = \frac{dn_i}{dt} + \frac{1}{2}(n \times \text{curl}\,\mathbf{v}), \quad d_{ij} = \frac{1}{2}\left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right). \qquad (3)$$

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

$$F = \frac{1}{2} K_1 (\text{div}\,\mathbf{n})^2 + \frac{1}{2} K_2 (\mathbf{n} \cdot \text{curl}\,\mathbf{n})^2 + \frac{1}{2} K_3 (\mathbf{n} \times \text{curl}\,\mathbf{n})^2, \qquad (4)$$

where K_i are Frank's elastic constants, α_i are Leslie coefficients of the NLC. The Navier–Stokes equation for hydrodynamic flow velocity $\mathbf{v}(\mathbf{r},t)$ of an incompressible NLC, with the presence of thermomechanical terms, are of the form

$$\rho\left(\frac{\partial v_i}{\partial t}+(\mathbf{v}\nabla)v_i\right)=\frac{\partial \sigma_{ki}}{\partial x_k},\tag{5}$$

where

$$\sigma_{ki}=-p\delta_{ki}+\sigma'_{ki}+\sigma^{thm}_{ki},\quad \text{div }\mathbf{v}=0,\tag{6}$$

ρ is the density, $p(\mathbf{r},t)$ is the hydrodynamic pressure determined from the same set of Eqs. (5), (6) and the boundary conditions, σ'_{ki} is the viscous stress tensor, and σ^{thm}_{ki} is the thermomechanical stress tensor, ∇ denotes the vector differential operator [26].

We consider the problem homogeneous in the (x, y) plane $(\partial/\partial x = \partial/\partial y =0)$ and the director distribution in the (x, z) plane $(n_y = 0)$ in the case of hybrid initial orientation. We have the boundary conditions for θ mentioned above. Thermomechanical stress leads to the hydrodynamic flow with velocity $\mathbf{v}$ directed in x direction $(\mathbf{v} = \mathbf{e}_x v)$. For this problem Eq. (1) for director $\theta(z)$ has the form

$$(\alpha_3-\alpha_2)\frac{\partial \theta}{\partial t}=(K_3\cos^2\theta+K_1\sin^2\theta)\frac{\partial^2\theta}{\partial z^2}-(K_3-K_1)\sin\theta\cos\theta\left(\frac{\partial\theta}{\partial z}\right)^2+$$
$$+(\alpha_3\sin^2\theta-\alpha_2\cos^2\theta)\frac{\partial v}{\partial z}.\tag{7}$$

Eq. (7) describes reorientation of the NLC director under the influence of the hydrodynamic velocity gradient. And for the velocity in the thermomechanical single-constant approximation $(\xi_1 = \xi_2 = ... = \xi_{12} = \xi)$, we have

$$\rho\frac{\partial v}{\partial t}=\eta\frac{\partial^2 v}{\partial z^2}-\frac{1}{4}\xi\frac{dT}{dz}\left[(5+2\sin^2\theta)\sin(2\theta)\left(\frac{\partial\theta}{\partial z}\right)^2+(5+2\sin^2\theta)\sin^2\theta\frac{\partial^2\theta}{\partial z^2}\right],\tag{8}$$

where $\eta=\eta_2+(\eta_1-\eta_2)\sin^2\eta+\eta_4\sin^2(2\theta)$, $\eta_4=0.25\alpha_1$ and $\eta_1=0.5(\alpha_3+\alpha_4+\alpha_6)$, $\eta_2=0.5(\alpha_4+\alpha_5-\alpha_3)$, $\eta_3=0.5\alpha_4$ are the Miesowicz viscosity coefficients. Generalized Eqs. (7), (8) describe the thermomechanical flow in NLC with the director confined to the surface of hybrid orientation. In our problem thermomechanical hydrodynamic flows and director reorientations caused by them are induced by laser radiation. Therefore, temperature gradient dT/dz can be represented in the form

$$\frac{dT}{dz}=\frac{\chi L}{\rho C_p r}P,\tag{9}$$

where r is the temperature conductivity coefficient, ρC_p is the specific volume thermal capacitance, χ is the absorption factor $(\chi L \ll 1)$ and P is the radiation intensity. Then, for laser induced thermomechanical flow and oscillation we have the general equation

$$\rho\frac{\partial v}{\partial t}=\eta\frac{\partial^2 v}{\partial z^2}-\frac{1}{4}\xi\frac{\chi T}{\rho C_p r}P(5+2\sin^2\theta)\left[\sin(2\theta)\left(\frac{\partial\theta}{\partial z}\right)^2+\sin^2\theta\frac{\partial^2\theta}{\partial z^2}\right].\tag{10}$$

Results and Discussion. 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 $(\theta(0) = 0, \ \theta(L) = \pi/2)$ and independent of the external excitations. We assume that in this system the bulk heat source is homogeneous relative to the y coordinate, $\partial/\partial y = 0$, and y component of velosity is vanished ($v_y = 0$). We assume that the rigid boundary at $z = 0$ and L is held at temperature T_0, and also that nonslip boundary conditions apply: $\mathbf{v}(z = 0, L) = 0$.

We solved Eqs. (7), (10) for director reorientation caused by laser induced thermomechanical flow with the above-mentioned boundary and initial $v(z, t = 0) = 0$, $\theta(z, t = 0) = 0$ conditions using "Mathematica-5". Here, for NLC MBBA was assumed: $K_1 = 6 \cdot 10^{-7} erg/cm$, $K_3 = 7.5 \cdot 10^{-7} erg/cm$, $\alpha_2 = -0.8 \ P$, $\alpha_3 = -0.012 \ P$, $\rho C_p = 1 J/cm^3 \cdot K$, for cell with $L = 0.01 \ cm$, $l = 0.1 \ cm$ and $\chi L = 0.5$. We study the reorientation induced by laser radiation with $P = 0.5 \cdot 10^{-3} \ W/cm^3$ intensity.
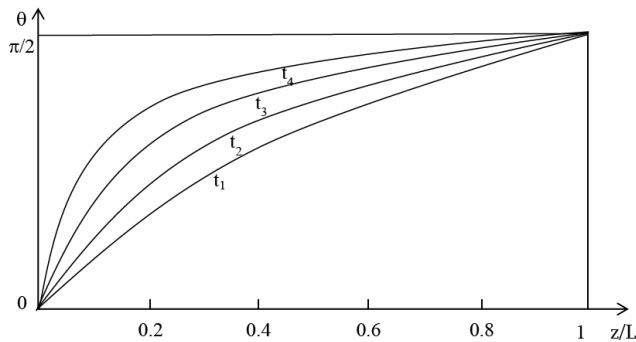

Fig. 2. Profile of the director's orientation angle (θ) for different times (t_j) after switching the laser radiation "on". Laser beam propagate in the direction from homeotropic to planar initial orientation of director:

$t_1 = 0$; $t_2 = 3 \ s$; $t_3 = 6 \ s$; $t_4 = 9 \ s$.

If the laser beam is propagated in the direction from homeotropic to planar initial orientation of director, then thermomechanical flow velocity is directed out of the "flexible ribbon's" curvature and velocity gradient brings about a small increase of curvature (see Fig. 2). The curvature deforms more completely and the deformation increases with time when laser beam is propagated in the direction from planar to homeotropic initial orientation of director and velocity is directed into "flexible ribbon's" curvature (see Fig. 3).
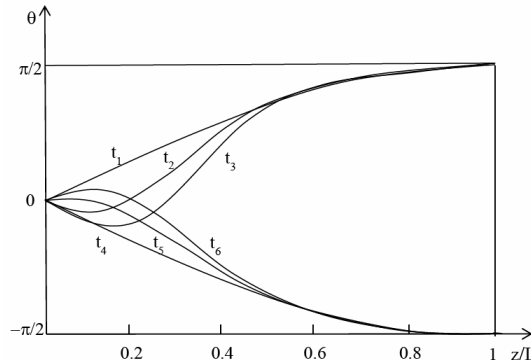

Fig. 3. Profile of the director's orientation angle (θ) for different times (t_j) after switching the laser radiation "on". Laser beam propagate in the direction from planar to homeotropic initial orientation of director:

$t_1 = 0$, $t_2 = 3 \ s$, $t_3 = 6 \ s$,
$t_4 = 7.5 \ s$, $t_5 = 10 \ s$, $t_6 = 13 \ s$.

Reversal time is $t_r = 7 \ s$ and oscillation period is $\tau_p = 14 \ s$.

The "flexible ribbon" reverses its curvature 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. The period of oscillation is twice of the reversal time. So, for the cell thickness $L = 0.01 \, cm$ we obtain a period of $\tau_p = 14 \, s$.

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, so called, trigger behavior and has the practical meanings.

The author wishes to thank R.B. Alaverdyan, Kh.H. Hakobian and R.S. Hakobyan for fruitful discussions.

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