Reorientation of liquid-crystal director by light near the threshold of spatially periodic convective instability

R. S. Akopyan and B. Ya. Zel'dovich

Institute of Mechanics Problems, USSR Academy of Sciences
(Submitted 27 June 1983)
Zh. Eksp. Teor. Fiz. **86**, 533–544 (February 1984)

We consider theoretically the action of a weak light wave on a nematic liquid crystal layer near the threshold of non-optical instability. If the unstable factor is a spatially-periodic perturbation in the plane of the layer, the susceptibility of the system becomes larger, when the threshold is approached from below, than for a periodic structure of the light intensity. It is easy to produce such a structure via interference of two or more coherent light waves. We consider the cases of electrohydrodynamic instability, convective Benard-type instability in a cell with a vertical temperature gradient, as well as roller instability of simple hydrodynamic flow. The possibility of controlling a global topological perturbation structure above threshold via weak light fields is discussed.

1. INTRODUCTION

When the parameters of a physical system approach the instability threshold, the rigidity of a definite mode tends to zero, and the corresponding susceptibility increases, in the self-consistent-field approximation, in accord with the Curie-Weiss Law. For example, in the transition from the isotropic phase of a nematic liquid crystal (NLC) to the mesophase, the orientational optical nonlinearity (i.e., the response of the molecule orientation to the intensity of the light field) increases in the isotropic phase like $\varepsilon_2 \propto (T - T^*)^{-1}$ (see the theoretical and experimental parts of Ref. 1). Another example is a cell with an NLC in the mesophase, which is transformed by application of an external rf voltage U into a state near the threshold of the Fréedericksz transition; in this case, too, the orientational optical nonlinearity increases in accord with a Curie-Weiss-type law $\varepsilon_2 \propto (U^2 - U_{\text{thr}}{}^2)^{-1}$ (see theory and experiment in Ref. 2).

We consider in this paper various problems concerning cells with NLC in the mesophase, in which the system is unstable to spatially periodic (roller) perturbations. In these cases the system susceptibility has a Curie-Weiss growth only with respect to spatially periodic actions having a "resonant" period. Usually resonant periods are spatial ones of the order of the cell length L, which equals under typical conditions from 5 to 1000 μm. We wish to call attention to the fact that interference of two coherent light beams of wavelength λ, propagating at an angle θ between them, produces a sinusoidal pattern of intensity modulation with a period $\Lambda \approx \lambda/\theta$, so that when visible-band lasers ($\lambda \sim 0.5\,\mu$m) are used it is easy to enter into spatial resonance by simply varying the angle in the range from 5×10^{-4} to 10^{-1} rad.

The mechanism whereby the light intensity acts on the system can be either orientational (rotation of the director by torque applied by the light wave) or trivially thermal (by light absorption). In turn, the system response (cubic optical nonlinearity) can be due to a change of the effective refractive index both on account of the change of the local orientation of the optical axis (the NLC director) and on account of temperature effects.

We emphasize that the cubic optical nonlinearity of cells with NLC is unusually large, so that even the beam from a low-power ($\sim 10^{-2}$ W) neon-helium cw laser is sufficient for its observation.

We wish to discuss also the possibility of using light fields to produce initial perturbations capable, after passage through the instability threshold, of imparting to the system a specified global structure of the perturbations in a nonlinear stationary state.

2. CUBIC OPTICAL NONLINEARITY NEAR ELECTROHYDRODYNAMIC INSTABILITY THRESHOLD

We consider a planarly oriented ($\mathbf{n}_0 = \mathbf{e}_x$) cell with an NLC that has a negative anisotropy of the dielectric constant at zero frequency $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp < 0$. Application of a moderate electric field in the direction of the z axis (along the normal to the cell walls) only stabilizes the initial planar structure. When the voltage difference between the walls is 5–10 V, however, if account is taken of the anisotropic electric conductivity of the medium in the cell, it is known that an electrohydrodynamic (EHD) instability can set in and produce Kapustin-Williams domains with a period of the order of the cell thickness.[3–5] We consider in this section the prethreshold behavior of such a cell when illuminated by a beam whose intensity varies periodically in space.

The theory of EHD instability is quite complicated[5–7] and the treatment is usually limited to a simplified variant in which the actually existing z-dependence of the perturbed variables is ignored, and the fact that the cell thickness L is finite is taken into account indirectly, by assigning to the perturbation a periodic structure of the form $\exp(iqx)$ with $q = q_0 \equiv \pi/L$. This procedure is justified by the good agreement between its deductions with the results of exact numerical calculations (see, e.g., Refs. 6–8).

The linearized NLC hydrodynamic equation for the velocity $\mathbf{v}(\mathbf{r},t)$ can be assumed in the form

$$\rho\frac{\partial v_i}{\partial t} = \frac{\partial \sigma_{ki}}{\partial x_k} + \frac{\partial T_{ki}}{\partial x_k}, \tag{1}$$

 0038-5646/84/020311-07$04.00

where ρ is the unperturbed density and σ_{ki} is the NLC stress tensor in the same linear approximation:

$$\sigma_{ki}(\mathbf{r},t)=-\delta_{ki}p(\mathbf{r},t)-\frac{\delta F}{\delta(\partial n_m/\partial x_k)}\frac{\partial n_m}{\partial x_i}+\sigma_{ki}', \tag{2}$$

$$\sigma_{ki}'=\alpha_1 n_k n_i n_j n_m d_{jm}+\alpha_2 n_k N_i+\alpha_3 n_i N_k+\alpha_4 d_{ki}+\alpha_5 n_k n_j d_{ji}+\alpha_6 d_{kj} n_j n_i, \tag{3}$$

$$\mathbf{N}=\frac{d\mathbf{n}}{dt}+\frac{1}{2}[\mathbf{n}\,\mathrm{rot}\,\mathbf{v}],$$
$$d_{ij}=\frac{1}{2}\left(\frac{\partial v_i}{\partial x_j}+\frac{\partial v_j}{\partial x_i}\right), \tag{4}$$

$$T_{ki}=\frac{1}{8\pi}(\mathscr{D}_i\mathscr{E}_k+\mathscr{D}_k\mathscr{E}_i-\vec{\mathscr{D}}\vec{\mathscr{E}}\delta_{ki}), \tag{5}$$

$$\mathscr{D}_i=\varepsilon_{ij}\mathscr{E}_j,\qquad \varepsilon_{ij}=\varepsilon_\perp\delta_{ij}+\varepsilon_a n_i n_j. \tag{6}$$

We assume here that the NLC is incompressible, div $\mathbf{v}=\partial v_i/\partial x_i=0$. Here $\mathbf{n}$ is the director unit vector, σ'_{ki} the viscous-stress tensor, $\alpha_1,\dots,\alpha_6$ are the Leslie coefficients (having the dimension of poise) and are connected by the relation $\alpha_6=\alpha_2+\alpha_3+\alpha_5$, p is the pressure, $\vec{\mathscr{D}}$ and $\vec{\mathscr{E}}$ are the quasistatic induction and electric field, F is the free-energy density:

$$F=\frac{1}{2}K_1(\mathrm{div}\,\mathbf{n})^2+\frac{1}{2}K_2(\mathbf{n}\,\mathrm{rot}\,\mathbf{n})^2+\frac{1}{2}K_3(\mathbf{n}\,\mathrm{rot}\,\mathbf{n})^2$$
$$-\frac{1}{2}\chi_a(\mathbf{nH})^2-\frac{\varepsilon_a}{8\pi}(\mathbf{n}\vec{\mathscr{E}})^2-\frac{\varepsilon_\perp}{8\pi}\vec{\mathscr{E}}^2-\frac{\tilde{\varepsilon}_a}{16\pi}(\mathbf{nE})(\mathbf{nE}^*)$$
$$-\frac{\tilde{\varepsilon}_\perp}{16\pi}\mathbf{EE}^*. \tag{7}$$

In (7), $\mathbf{H}=\mathbf{e}_x H$ is the intensity of the quasistatic magnetic field, $\mathbf{E}$ is the complex amplitude of the light-wave field,

$$\mathbf{E}_{\mathrm{mat}}=0.5[\mathbf{E}(\mathbf{r})\exp(-i\omega t)+\mathrm{c.c.}],$$

$\tilde{\varepsilon}_{ik}$ is the dielectric tensor at the optical frequency, $\tilde{\varepsilon}_\perp=n_\perp^2$, $\tilde{\varepsilon}_\perp+\tilde{\varepsilon}_a=n_\parallel^2$, where $n_\parallel$ and $n_\perp$ are the refractive indices of the uniaxial NLC, χ_a is the anisotropy of the magnetic susceptibility, and K_i are the Frank constants. The electrodes mounted on the cell walls can inject into the NLC volume charges with density $Q(\mathbf{r},t)$, carrying injection conduction currents with density $\mathscr{J}_i$, so that

$$\mathscr{J}_i=Q(\Pi_{ij}\mathscr{E}_j+v_i),\qquad \Pi_{ij}=\Pi_\perp\delta_{ij}+\Pi_a n_i n_j, \tag{8}$$

where Π_{ij} is the mobility tensor ($Q\Pi_{ij}$ is the conductivity tensor). Continuity equations apply to the charges and current, while $\vec{\mathscr{D}}$ must satisfy the Poisson equation

$$\partial Q/\partial t+\mathrm{div}\,\vec{\mathscr{J}}=0,\qquad \mathrm{div}\,\vec{\mathscr{D}}=4\pi Q. \tag{9}$$

The variation of the director $\mathbf{n}(\mathbf{r},t)$ in time and space obeys an equation that expresses the balance of the moments of the forces acting on a unit volume of the NLC:

$$[\mathbf{fn}]_i+e_{ijm}n_m\left[\frac{\delta F}{\delta n_j}-\frac{\partial}{\partial x_k}\frac{\delta F}{\delta(\partial n_j/\partial x_k)}\right]=0, \tag{10}$$

$$f_i=(\alpha_3-\alpha_2)N_i+(\alpha_3+\alpha_2)d_{ij}n_j. \tag{11}$$

A derivation of the foregoing system of equations can be found in Ref. 9.

The averaged unperturbed state is assumed in the simplified approach to the theory of the EHD instability in the form

$$\mathbf{n}_0=\mathbf{e}_x,\quad v_0=0,\quad Q_0\approx 3\varepsilon_\perp U/8\pi L^2,$$
$$\vec{\mathscr{E}}_0\approx\mathbf{e}_z U/L,\qquad \partial Q_0/\partial z\approx -Q_0/L, \tag{12}$$

where U is the potential difference between the electrodes $z=0$ and $z=L$. One should not fear the fact that Eqs. (9) have only order-of-magnitude accuracy for the quantities specified approximately by (12) (for details see Refs. 7 and 9).

We assume that the perturbation of interest to us is caused by the spatially inhomogeneous term

$$E_i(\mathbf{r},t)E_k^*(\mathbf{r},t)=a_{ik}\exp(iqx+i\Omega t)$$

in the tensor $E_iE^*_k$ that describes the intensity distribution of the incident optical field. Such a term can be obtained in interference between two coherent waves having a frequency difference $\omega_1-\omega_2=\Omega$ and an incidence-angle difference $\theta_{1x}-\theta_{2x}=q\lambda/2\pi$ (see Fig. 1). In this case, assuming

$$Q=Q_0+Q_1(x,t),\quad \vec{\mathscr{E}}=\vec{\mathscr{E}}_0+\mathbf{e}_x\mathscr{E}_1(x,t),\quad \mathbf{n}=\mathbf{e}_x+\mathbf{e}_z\varphi(x,t),$$

we obtain a system of linearized equations

$$\varepsilon_\parallel\frac{\partial\mathscr{E}_1}{\partial x}+\varepsilon_a\frac{U}{L}\frac{\partial\varphi}{\partial x}=4\pi Q_1, \tag{13}$$

$$\frac{\partial Q_1}{\partial t}+\frac{Q_1}{\tau}-v_z\frac{Q_0}{L}+\sigma\frac{U}{L}\frac{\partial\varphi}{\partial x}=0, \tag{14}$$

$$(\alpha_3-\alpha_2)\frac{\partial\varphi}{\partial t}+\alpha_2\frac{\partial v_z}{\partial x}-\frac{\varepsilon_a}{4\pi}\frac{U}{L}\left(\mathscr{E}_1+\frac{U}{L}\varphi\right)$$
$$-K_3\frac{\partial^2\varphi}{\partial x^2}+\chi_a H^2\varphi=\frac{\tilde{\varepsilon}_a}{16\pi}(a_{xz}+a_{zx})e^{iqx+i\Omega t}, \tag{15}$$

$$\rho\frac{\partial v_z}{\partial t}=\alpha_2\frac{\partial^2\varphi}{\partial x\,\partial t}+\eta_2\frac{\partial^2 v_z}{\partial x^2}$$
$$-\frac{\varepsilon_a\varepsilon_\perp}{8\pi\varepsilon_\parallel}\frac{U^2}{L^2}\frac{\partial\varphi}{\partial x}+\frac{1}{2}\left(1+\frac{\varepsilon_\perp}{\varepsilon_\parallel}\right)\frac{U}{L}Q_1. \tag{16}$$

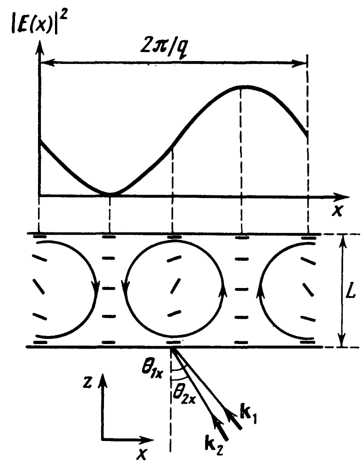

FIG. 1. Two light waves that produce an interference pattern $|\mathbf{E}(x)|^2$ are incident at angles θ_{1x} and θ_{2x} on an NLC layer of thickness L and with an unperturbed director $\mathbf{n}_0=\mathbf{e}_x$. The dashes show the direction of the director, and the arrows the directions of the hydrodynamic flows.

Here $\eta_2 = 0.5(\alpha_4 + \alpha_5 - \alpha_2)$ is the Miesowicz coefficient,

$$\tau^{-1}=4\pi Q_0(\Pi_\parallel/\varepsilon_\parallel+\Pi_\perp/\varepsilon_\perp), \qquad \sigma=Q_0(\Pi_a-\varepsilon_a\Pi_\parallel/\varepsilon_\parallel). \quad (17)$$

The EHD instability threshold means that at a fixed $q_0 = \pi/L$ and $\Omega = 0$ the response of the system, according to Eqs. (13)–(16), becomes infinitely strong. From the vanishing of the determinant of the system (13)–(16) follows an equation for the threshold voltage $U_{\rm thr}$. We shall not write it out explicitly (see Ref. 9). We present the final expression for the Fourier component

$$\varphi=\varphi_1\exp(iqx+i\Omega t)+\text{c.c.}$$

of the inclination of the director

$$\varphi_1=\frac{\varepsilon_a}{16\pi}[K_3q^2+\chi_aH^2]^{-1}(a_{xz}+a_{zx})\frac{\xi^{-1}}{1+i\Omega/\Gamma}. \quad (18)$$

Here ξ is the dimensionless degree of proximity to the EHD instability threshold and Γ is the relaxation constant:

$$\xi=1-U^2/U_{\rm thr}^2, \quad \Gamma=\Gamma_0\xi, \quad (19)$$

$$\Gamma_0=(K_3q^2+\chi_aH^2)/\gamma^*,$$
$$\gamma^*=\alpha_3-\alpha_2-\alpha_2^2/\eta_2. \quad (20)$$

The quantity Γ_0 characterizes the relaxation constant of the orientation in the cell in the absence of an electric field. At $\xi = +1$ expression (18) corresponds to the usual orientational nonlinearity of NLC. We have left out from (18)–(20) a number of inessential factors that are quite close to unity. Their inclusion would be an exaggeration of the accuracy in view of the approximations made on going to the simplified model.

In the exact approach $U_{\rm thr}$ should depend on the parameter q, $U_{\rm thr} = U_{\rm thr}(q)$, as should the physical threshold of the EHD instability $U_0 = U_{\rm thr}(q_0)$, which is reached at a certain value $q = q_0$, so that

$$U_{\rm thr}(q)\approx U_0[1+b(q-q_0)^2/q_0^2], \quad (21)$$

where b is a coefficient of order unity. As a result, the Fourier component of the nonlinearity is determined by expressions (18)–(20) with the substitution $\xi\to\xi + b(q-q_0)^2/q_0^2$. By the same token, the half-width of the Lorentz curve at half the maximum level (HWHM) of the nonlinearity constant with respect to the variable q is

$$\Delta q=q_0(\xi/b)^{1/2}.$$

This means that the resonance with respect to the variable q is quite broad, $\Delta q\propto\xi^{1/2}$.

It is convenient to obtain the numerical estimates at $\xi\sim 1$ and $q = \pi/L$ for light incident at an angle $\alpha\approx 30°$ (in the medium), when

$$a_{xz}=a_{zx}=|E_0|^2\sin\alpha\cos\alpha\approx 0.4|E_0|^2,$$

while $H = 0$ and $L = 10^{-2}$ cm. The perturbation φ_1 of the director is then of the order of one radian at a power density on the order of 1.2 kW/cm^2. This power can be easily obtain by focusing the beam of a cw (e.g., argon) laser. The approach to the EHD instability threshold decreases by another factor ξ^{-1} the required light-power density needed to attain $\varphi_1\sim 1$ rad. At the same time the response of the system to the spatially homogeneous part of the intensity does not undergo a Curie-Weiss increase.

Moreover, to record a spatially periodic director perturbation $\varphi\propto a_{ik}\exp(iqx)$ there is no need at all to reach $\varphi\sim 1$. The point is that a director modulation $\varphi(x) = 2\varphi_1\cos qx$ leads to a modulation $\delta\psi(x)$ of the phase of an e-type wave passing through the cell at an angle α, with

$$\delta\psi(x)=2\frac{\tilde\varepsilon_a\omega}{c\tilde\varepsilon_e^{1/2}}L\varphi_1\sin\alpha\cos qx\equiv\psi\cos qx. \quad (22)$$

It turns out (see Appendix 2) that it is easy to record ψ_1 at a level $\sim 1/30$, which corresponds to $\varphi_1\approx 6\times 10^{-5}$ rad at $L\approx 10^{-2}$ cm, $\tilde\varepsilon_a = 0.7$, and $\alpha = 30°$. Within the framework of the foregoing estimates, this permits another decrease of the power by a factor 10^4, i.e., to a value $\sim 10^{-1}$ W/cm^2.

If the NLC absorbs the incident light, thermal effects can come into play. Namely, the inhomogeneous heating $\delta T(x)$ can cause a spatially inhomogeneous perturbation of the static dielectric constant

$$\delta\varepsilon_{ik}(x)=(\partial\varepsilon_{ik}/\partial T)\,\delta T(x).$$

This leads in turn to successive perturbations of the electric field, current, hydrodynamic flow, and director. A calculation (not given here) shows that the contribution of this mechanism to the change of the director orientation $\varphi(x)$ is proportional to

$$\varphi\propto|\mathbf{E}|^2L\cdot(\varkappa L)\,(\partial\varepsilon/\partial T),$$

where $\varkappa$ [cm^{-1}] is the light absorption coefficient. Assuming $\varkappa L\sim 1$ and $\partial\varepsilon/\partial T\sim 10^{-2}$ K^{-1}, the contribution of the thermal mechanism is of the same order as the direct orienting action of the light field at $L\sim 10^{-2}$ cm and $\alpha = 30°$. The unique feature of the thermal mechanism that it works for waves of arbitrary (but equal) type of polarization, and also at normal incidence of the waves on the cell. One other thermal nonlinearity mechanism is based on the temperature dependence of the mobility and of the conductivity. Unfortunately, we have no data on this dependence for NLC.

3. THERMAL AND ORIENTING ACTION OF LIGHT ON AN NLC LAYER NEAR THE THRESHOLD OF THE BENARD CONVECTIVE INSTABILITY

Convective instability in a layer of liquid with negative vertical temperature gradient, situated in a gravitational field, is well known back from the times of Benadr and Rayleigh (see, e.g., Refs. 10 and 11). Convective instability of an NLC layer in a gravitational field in the presence of a temperature gradient[12,13] corresponds to softening of the slow nematic mode that is absent in isotropic liquids. The numerical value of the vertical temperature gradient $T_0(L)$ needed to obtain convective instability turns out to be about an order of magnitude lower for typical NLC than for typical isotropic liquids.

We consider in this section the problem of thermal convection excited by inhomogeneous heat release following absorption of radiation from other interfering light waves. We assume that the gravitational force is directed along the z axis ($\mathbf{g} = -\mathbf{e}_z g$, $g = 10^3$ cm^2 sec^{-1}) in a direction normal to the cell walls, and that the initial director orientation is planar, $\mathbf{n}_0 = \mathbf{e}_x$. It is then necessary to set the static electric field, the charge density, and the current in the system (1)–(11) equal to zero and supplement the system by the linearized heat-conduction equation

$$\rho c_p \left(\frac{\partial T}{\partial t} + v_i \frac{\partial T}{\partial x_i} - \frac{\partial}{\partial x_i} r_{ik} \frac{\partial T}{\partial x_k}\right) = \frac{\varkappa c n |\mathbf{E}|^2}{8\pi}, \tag{23}$$

where ρc_p is the specific heat per unit volume and r_{ik} is the thermal diffusivity tensor. In addition, the Navier-Stokes equation must be modified to take into account the force of gravity

$$\rho \frac{\partial v_i}{\partial t} = \frac{\partial \sigma_{ki}}{\partial x_k} - (\mathbf{e}_z)_i \rho g (1 - \beta T). \tag{24}$$

Here β is the bulk coefficient of thermal expansion and $T(\mathbf{r},t)$ is the temperature variation.

Assume that an external heat source maintains the initial vertical temperature gradient $T(z) = -T_0 z/L$; such a profile is the stationary solution of the heat-conduction equation without the right-hand side. Of very great importance to us is also the case when there is no gradient in the initial state, $T_0 = 0$, and the light field produces forced thermal convection in pure form.

We consider the solution of the linearized system (2), (3), (9), (10), (23), (24), a solution stimulated by the spatially inhomogeneous part $\propto \exp(iqx)$ of the absorbed power and by the moment of the forces due to the direct action of the light field. In the same model-based approach, i.e., when the z-dependence of the perturbations is ignored, we have

$$\varphi = \left[\frac{1}{2}\tilde{\varepsilon}_a \sin 2\alpha - i \frac{\alpha_2 \varkappa c n \beta g}{c_p \eta_2 r_\parallel q^3}\right] \times [K_3 q^2 + \chi_a H^2]^{-1} \xi^{-1} \frac{E_1 E_2^*}{8\pi} e^{iqx} + \text{c.c.} \tag{25}$$

Here, as in the preceding section, the dimensionless parameter is a measure of the degree of proximity of the system parameters to the threshold, or in our case the proximity of the temperature gradient to its critical value:

$$\xi = 1 - \frac{T_0}{T_0^{\text{c.c.}}}, \quad \frac{T_0^{\text{c.c.}}}{L} = \frac{r_\parallel q^4 \eta_2}{\rho g \beta [1 + (r_\parallel - r_\perp) q^2/\Gamma_1]}, \tag{26}$$

$$\Gamma_1 = (K_3 q^2 + \chi_a H^2) |\alpha_2|^{-1}. \tag{27}$$

The true threshold of convective instability in a cell with an NLC of finite thickness L is obtained with good accuracy from (25) by the substitution $q \approx \pi/L$ (see Ref. 12). In the absence of an external magnetic field ($\chi_a H^2 = 0$) we find from (25) that when the parameter q decreases the director orientation φ is increased like q^{-2} by action of the light field and like q^{-5} as a result of thermal convection. The considered simplified model does not "work" for $q \lesssim \pi/L$. A more accurate calculation shows that with further decrease of q the term $\varphi \propto \tilde{\varepsilon}_a$ stabilizes at a level $\varphi \propto L^2$, while the term $\varphi \propto \varkappa$ begins to decrease like $\varphi \propto \varkappa \cdot LqL^5$.

At a value $q \approx \pi/L$ the response of the orientation to the convective-thermal mechanism is a maximum. In the presence of a slight inclination of the director by an angle $\varphi(x)$ the nonlinear phase shift (22) of a light wave propagating at an angle $\alpha \lesssim 1$ to the z axis increases like $\delta\psi \propto L^3$ (see Ref. 14) on account of the giant nonlinearity, and like $L^5 \cdot (\varkappa L)$ on account of thermal convection.

We obtain numerical estimates for a planar cell with MBBA at $q \approx \pi/L$. Putting $\varkappa L = 0.5$, $\alpha_2 = -0.8$ P, $\eta_2 = 1$ P, $\tilde{\varepsilon}_a = 0.7$, $\beta = 10^{-3}$ deg^{-1}, $r_\parallel \approx 10^{-4}$ cm^2·sec^{-1} (see Ref. 12) and $\rho c_p = 1$J·cm^{-3}·deg^{-1}, we obtain

$$\frac{\delta\psi(\text{convect})}{\delta\psi(\text{direct})} \approx 150 \left(\frac{L}{100\ \mu\text{m}}\right)^2 \frac{1}{\alpha}.$$

Thus, even for cells 100 μm thick and at $\alpha \sim 1$ rad the orientational-convective-thermal mechanism yields a nonlinearity 150 times stronger than the "giant" orientational nonlinearity of Ref. 14. In particular, even in the absence of an initial temperature gradient ($T_0 = 0$, $\xi = 1$) the stimulated thermal convection yields a director inclination $\varphi \sim 1$ rad at an incident-light-wave power on the order of 10 W/cm^2. When the vertical temperature gradient is excluded, the nonlinearity increases by another factor ξ^{-1} as the Benard instability threshold is approached.

If the task is not to produce a modulation of $\varphi \sim 1$ rad, but to record the effect, the required incident-light power density can be decreased by approximately four orders more (see the preceding section and the Appendices).

We note that under the conditions indicated above the nonlinearity due to the change $\delta\tilde{\varepsilon}_{ik} = (\partial\tilde{\varepsilon}_{ik}/\partial T)\delta T$ of the tensor $\tilde{\varepsilon}_{ik}$ due directly to the change of the temperature turns out to be of the same order as the giant value from Ref. 14 at $\tilde{\varepsilon}^{-1}(\partial\tilde{\varepsilon}/\partial T) \sim 10^{-3}$ deg^{-1}. The mechanism due to $\partial\tilde{\varepsilon}/\partial T$ yields by itself for $\delta\psi$ the functional relation $\delta\psi \sim L^2 \cdot (\varkappa L)$.

An inhomogeneous temperature profile sets in within a time $(r_\parallel q^2)^{-1}$, or approximately 0.1 sec at $q = \pi/L$ and $L = 100\ \mu$m. The velocity changes with temperature with practically no time delay. This is followed by a director reorientation after an orientational diffusion time $\tau' \sim (K_3 q^2/|\alpha_2|)^{-1}$, or numerically $\tau' \sim 10$ sec at $L = 100\ \mu$m.

An interesting feature of the orientationally convective nonlinearity is the possibility of recording perturbations (holograms) by o-type waves; an e-wave is needed only to read these perturbations. We note also the factor $i = \exp(i\pi/2)$ in the convective term for $\varphi(x)$. This phase shift of the response relative to the action $E_1 E^*{}_2 \exp(iqx)$ is typical of processes of the type of stimulated light scattering due to absorption (cf. Ref. 22). It is curious that a phase shift occurs in this case even when waves of equal frequency interact.

In our opinion, it is of considerable interest to use the considered nonlinearity for four-wave reversal of the wavefront of light, using a cell with NLC as a thin-layer hologram. It is very important that at $q \lesssim L^{-1}$ the convective nonlinearity decreases; this means that no self-focusing distortions of the reference waves will occur.

4. ACTION OF LIGHT ON AN NLC LAYER IN ROLLER HYDRODYNAMIC INSTABILITY

We consider in this section a number of problems concerning inhomogeneous hydrodynamic flow of an NLC. Couette flow is produced between two parallel surfaces that move tangentially relative to each other. In the unperturbed state we have here $\mathbf{v}_0(z) = \mathbf{e}_y v_0 z/L$, i.e., the velocity gradient is constant and equal to v_0/L. Poiseuille flow between two immobile planes corresponds to an unperturbed velocity distribution

$$\mathbf{v}_0(z) = \mathbf{e}_y v_0 \cdot 4z(L-z)/L^2,$$

where the flux is

$$Q[\mathrm{cm}^2/\mathrm{sec}] = \int_0^L |\mathbf{v}_0(z)|\,dz = 2v_0L/3 = -(dp_0/dy)L^3/12\eta_3,$$

and dp_0/dy is the pressure gradient. Finally gravity flow (IPF) over a plane inclined at an angle ν, with a free upper boundary, corresponds to a velocity distribution

$$\mathbf{v}_0(z) = \mathbf{e}_y \rho g z(2L-z)\sin\nu/2\eta_3.$$

In all three problems the z axis is aligned with the normal to the boundary of a layer of thickness L, and in the last problem the free-fall acceleration is assumed in the form $\mathbf{g} = g(-\mathbf{e}_z\cos\nu + \mathbf{e}_y\sin\nu)$; $\eta_3 = \alpha_4/2$, while the unperturbed director distribution is assumed planar: $\mathbf{n}_0 = e_x$.

In all three problems instabilities set in with increasing velocity or flux: first instabilities to perturbations that or homogeneous in the (x,y) plane, and at large fluxes roller instabilities with perturbations of the form $\exp(iqx)$, where $q \approx \pi/L$. All these instabilities were considered in Refs. 15 and 16.

We consider the action of a spatially periodic light-intensity pattern $\propto e^{iqx}$ on NLC for the three indicated problems, both as a result of direct action of the moment of the forces applied by the light field, and on account of thermal effect in the light-absorbing NLC. The unperturbed state will be chosen to be below the threshold of all the instabilities.

The linearized system of stationary-nematodynamics equations is obtained from the system (1)–(11) and takes for these problems the form

$$-K_3\frac{\partial^2 n_z}{\partial x^2} + \alpha_2\frac{\partial v_z}{\partial x} + \alpha_3 s n_y = \frac{\tilde{\varepsilon}_a}{16\pi}(a_{xz}+a_{zx})e^{iqx}, \tag{28}$$

$$-K_3\frac{\partial^2 n_y}{\partial x^2} + \alpha_2\frac{\partial v_y}{\partial x} + \alpha_2 s n_z = 0, \tag{29}$$

$$\eta_2\frac{\partial^2 v_y}{\partial x^2} + (\eta_2-\eta_3)s\frac{\partial n_z}{\partial x} = -\delta\eta_3(x)\frac{\partial s}{\partial z}, \tag{30}$$

$$\eta_2\frac{\partial^2 v_z}{\partial x^2} + \frac{1}{2}(\alpha_2+\alpha_5)s\frac{\partial n_y}{\partial x} = 0. \tag{31}$$

Here $\delta\mathbf{v} = \mathbf{v} - \mathbf{v}_0 = (v_x; v_y; v_z)$ and $\delta\mathbf{n} = \mathbf{n} - \mathbf{n}_0 = (0; n_y; n_z)$ are the perturbations of the hydrodynamic velocity and of the director of the liquid crystal, $s = \partial|\mathbf{v}_0|/\partial z$ is the velocity gradient in the corresponding unperturbed problem, while $\delta\eta_3$ describes the change of the Miesowicz viscosity coefficient η_3 by the energy of the absorbed light. In particular, for the thermal mechanism of the change of η_3, we have

$$\delta\eta_3(x) = \frac{\partial\eta_3}{\partial T}\delta T(x) = \frac{\partial\eta_3}{\partial T}\frac{\varkappa c n E_1 E_2^* e^{iqx}}{8\pi\rho c_p r_\parallel q^2}. \tag{32}$$

Everywhere in this model approximation we replace $s(z)$ and its gradient $s' = \partial s/\partial z$ by the mean values $s(z) = v_0/L$ and $s' = 0$ (Couette flow),

$$s(z) \to 2Q/L^2 = -L(dp_0/dy)/6\eta_3, \quad s' = (dp_0/dy)/\eta_3$$

(Poiseuille flow), and

$$s(z) \to (2\rho g L/3\eta_3)\sin\nu, \quad s' = -(\rho g/\eta_3)\sin\nu$$

(IPF), and seek the z-independent perturbed solution $\propto \exp(iqx)$.

We present the final results for the director perturbations:

$$n_y = -\alpha_2\frac{\eta_3}{\eta_2}\left[\frac{1}{2}\tilde{\varepsilon}_a\frac{s}{K_3q^2}\sin 2\alpha + i\frac{1}{\eta_3}\frac{\partial\eta_3}{\partial T}s'\frac{\varkappa c n}{\rho c_p r_\parallel q^3}\right] \times\frac{\xi^{-1}E_1E_2^*}{8\pi K_3q^2}e^{iqx}, \tag{33}$$

$$n_z = \left[\frac{1}{2}\tilde{\varepsilon}_a(K_3q^2)^{-1}\sin 2\alpha + i\frac{1}{\eta_3}\frac{\partial\eta_3}{\partial T}\frac{ss'}{s^2_{\mathrm{rol}}}\frac{\varkappa c n}{\rho c_p r_\parallel q^3}\right] \times\frac{\xi^{-1}E_1E_2^*}{8\pi}e^{iqx} \tag{34}$$

Here $\xi = 1 - s^2/s^2_{\mathrm{rol}}$ is the degree of proximity of the velocity gradient to the threshold in the initial perturbed problem with respect to roller instability with $q \approx \pi/L$. Within the framework of the considered procedure with z-averaging, this threshold has the same parameter s for all types of flow

$$s_{\mathrm{rol}} = K_3q^2\left\{\alpha_2\alpha_3\frac{\eta_3}{\eta_2}\left[1 - \frac{\alpha_2}{2\alpha_3\eta_2}(\alpha_2+\alpha_3)\right]\right\}^{-1/2}. \tag{35}$$

For Couette flow this result was obtained earlier in Ref. 17; for Poiseuille flow and IPF this result is presented here apparently for the first time. In the presence of a magnetic field $\mathbf{H} = \mathbf{e}_2 H$ it is necessary to make in (33)–(35) the substitution $K_3q^2 \to K_3q^2 + \chi_a H^2$. For IPF at $H = 0$ the roller instability of the unperturbed flow corresponds to a threshold flux

$$Q_{\mathrm{rol}}[\mathrm{cm}^2\,\mathrm{sec}^{-1}] = 0.5\pi^2 K_3\left\{\alpha_2\alpha_3\frac{\eta_3}{\eta_2}\left[1 - \frac{\alpha_2}{2\alpha_3\eta_2}(\alpha_2+\alpha_3)\right]\right\}^{-1/2}. \tag{36}$$

For IPF in MBBA, for example, $Q_{\mathrm{rol}} \approx 1.2\times10^{-6}$ $\mathrm{cm}^2\cdot\mathrm{sec}^{-1}$, or approximately one order higher than the instability threshold for perturbations that are homogeneous in the (x,y) plane.[16]

The direct orienting action of the light field on the director (giant optical nonlinearity) is enhanced by a factor ξ^{-1} compared with the case of an NLC at rest. The thermal action on the orientation corresponds at $q \approx \pi/L$ to the relation

$$|\delta\mathbf{n}| \propto (1-\xi)\xi^{-1}(\varkappa L)L, \tag{37}$$

i.e., it vanishes at $\xi = 1$ for an immobile NLC. Since direct action corresponds to the relation $|\delta\mathbf{n}| \propto L^2\xi^{-1}\alpha$, where α is the refraction angle, thermal effects are found to be stronger at small L and small α. Assuming $\partial\eta_3/\partial T \sim \eta_3\,10^{-2}\,\mathrm{K}^{-1}$ we find for MBBA at $\alpha \sim 45°$ and $\varkappa L \sim 0.5$ that temperature-orientational effects become comparable with direct action at a thickness $L \approx 1000\,\mu\mathrm{m}$.

5. CONCLUSION

From the foregoing analysis and numerical estimates it follows that noticeable director perturbations ($|\delta \mathbf{n}| \sim 1$) require quite moderate (easily obtainable) power densities of the interfering light beams. By the same token we are able to stipulate initial perturbations right away over the entire plane occupied by the NLC layer. If the system is transferred next to a state above the roller-instability threshold then, apparently, the roller perturbations at each point go over to a stationary nonlinear state, and only later does the period of the pattern adjust itself to the optimal period for the given linear stage. A very interesting possibility of producing roller structures with dislocations is provided by the pattern of interference of a plane light wave with another wave having a wavefront dislocation (see Refs. 18–21). Annular roller structures can be obtained by interference of a plane light wave with a wave having a conical front. Interference of three, four, and more waves permits assembly of cells with hexagonal, cubic, and other structures; it is also possible to introduce purposefully defects in such structures. In addition, there are grounds for expecting an optical interference pattern to impose, at a slight excess above threshold, its period and phase on the established picture of rollers or cells. From our viewpoint such a possibility of controlling spatial structures is of interest not only for liquid crystals, but for any system having an instability with a finite wave vector $1 \lesssim q_0 \lesssim 10^5\ \mathrm{cm}^{-1}$ in the transverse plane.

It is also of interest to use the considered optical nonlinearities for four-wave reversal of the wavefront of light on thin dynamic holograms.

The authors thank L. M. Blinov, I. E. Kats, N. V. Tabiryan, A. V. Sukhov, Yu. S. Chilingaryan, and V. V. Shkunov for valuable discussions.

APPENDIX 1

Exact solution of the problem of stimulated convection in a layer with special boundary conditions

To illustrate the complexities that arise in the exact solution (in contrast to the model-based one without z-dependence) we present here a solution of the problem of stimulated thermal convection of an NLC in a planar cell without an initial temperature gradient. We succeeded in obtaining this solution for the following boundary conditions:

$$v_z(x, z=0)=v_z(x, z=L)=0, \quad \varphi(x, z=0)=\varphi(x, z=L)=0,$$
$$T(x,z=0)=T(x,z=L)=0, \tag{A.1.1}$$
$$\frac{\partial v_x}{\partial z}(x,z=0)=\frac{\partial v_x}{\partial z}(x,z=L)=0,$$

i.e., the boundaries maintain the zero values of the perturbations of the temperature T, of the director inclination angle $\varphi \equiv n_z$, and of the z component of the velocity v_z. On the contrary, the zero value of the derivative with respect to z is maintained for the x-component of the velocity. The system of linearized equations (2), (3), (9), (10), (23), and (24) with boundary conditions (A.1.1) can be solved in the following manner. The spatially inhomogeneous term $E_i(\mathbf{r},t)E_k^*(\mathbf{r},t)=a_{ik}e^{iqx}$ in the tensor $E_iE_k^*$, which characterizes the light-field intensity distribution, causes stationary perturbations

$$T(x,z)=T_1e^{iqx}\sin\frac{\pi z}{L}, \quad \varphi(x,z)=\varphi_1e^{iqx}\sin\frac{\pi z}{L},$$
$$v_x(x,z)=v_{1x}e^{iqx}\cos\frac{\pi z}{L}, \quad v_z(x,z)=v_{1z}e^{iqx}\sin\frac{\pi z}{L}, \tag{A.1.2}$$

which satisfy the boundary conditions on the free boundaries $z=0$ and $z=L$. We obtain for the amplitudes of these perturbations

$$T_1=\frac{\varkappa cn}{2\pi^2\rho c_p}(r_\parallel q^2+r_\perp q_0^2)^{-1}E_1E_2^*, \tag{A.1.3}$$

$$v_{1x}=iq\beta\rho g T_1(\eta_1 q_0^3+2\eta q_0 q^2+\eta_2 q^4 q_0^{-1})^{-1}, \tag{A.1.4}$$

$$\varphi_1=v_{1x}(\alpha_3 q_0-\alpha_2 q^2 q^{-1})(K_1 q_0^2+\chi_a H^2+K_3 q^2)^{-1}, \tag{A.1.5}$$

$$v_{1z}=-iq_0 q v_{1x}. \tag{A.1.6}$$

Here $q_0 \equiv \pi/L$, $\eta_1 \equiv 0.5(\alpha_3+\alpha_4+\alpha_6)$ is the Miesowicz viscosity coefficient, and $\eta \equiv 0.5(\alpha_1+\alpha_4+\alpha_6-\alpha_2)$. From (A.1.5) it can be seen that at $q=q_0(\alpha_3/\alpha_2)^{1/2}$ the viscous torques acting on the director cancel one another and there is no reorientation as a result, although convection does develop.

At $q \ll q_0(\alpha_3/\alpha_2)^{1/2}$ the response begins to fall off like

$$\varphi_1 \infty (\varkappa q_0^{-1}) q q_0^{-5}.$$

With increasing q the inclination increases and reaches at $q \approx 0.05\ q_0$ (for MBBA) its first maximum. At the point $q=q_0(\alpha_3/\alpha_2)^{1/2} \sim 0.1 q_0$ (for MBBA) its phase changes by π and at $q \approx 0.6 q_0$ its amplitude reaches its second maximum. The latter corresponds to the transverse period of the Benard instability at a constant vertical temperature gradient. At $q \gg 0.6 q_0$ the reorientation decreases like $\varphi_1 \propto q^{-5}(\varkappa q_0^{-1})q_0$.

APPENDIX 2

Self-diffraction of a pair of light waves by a thin layer of a nonlinear medium

We consider the incidence of two plane light waves

$$E_{\mathrm{decr}}(x,z)=E_0\exp[iq_0x+iz(k^2-q_0^2)^{1/2}] + E_1\exp[iq_1x+iz(k^2-q_1^2)^{1/2}] \tag{A.2.1}$$

on a layer of a nonlinear medium. We assume that the layer is thin, so that its action consists of multiplying the incident wave by a complex transmission coefficient $t(x)$, making the transmitted-wave amplitude at the section $z=0$ equal to

$$E_{\mathrm{trans}}(x)=t(x)E_{\mathrm{decr}}(x)=t(x)[E_0e^{iq_0x}+E_1e^{iq_1x}]. \tag{A.2.2}$$

We assume that the layer modulates the phase of the transmitted light as follows:

$$t(x)=e^{i\psi(x)}, \quad \psi(x)=\psi_0+\mu E_0^*E_1e^{iqx}+\mu^*E_0E_1^*e^{-iqx}. \tag{A.2.3}$$

Here $q=q_1-q_0, \psi_0=\mu_0(|E_0|^2+|E_1|^2)$, and the coefficients μ_0, μ, and μ^* describe the properties of the nonlinear response of the layer. We use the transformation

$$\mu E_0^*E_1e^{iqx}+\mu^*E_0E_1^*e^{-iqx} \equiv a\cos(qx+\delta),$$

where

$$\delta=\arg(\mu E_0 E_1^*), \quad a=2|\mu E_0^* E_1|, \tag{A.2.4}$$

as well as the relation known from the theory of Bessel functions

$$e^{ia\cos\alpha}=\sum_{n=-\infty}^{+\infty} i^n e^{in\alpha} J_n(a). \tag{A.2.5}$$

The amplitude of the transmitted wave can then be represented as a sum of waves diffracted in mth order:

$$E_{\text{trans}}(x,z)=e^{i\psi_0}\sum_{m=-\infty}^{+\infty}\exp[iq_m x+iz(k^2-q_m^2)^{1/2}]C_m, \tag{A.2.6}$$

$$q_m=q_0+mq\equiv q_0+m(q_1-q_0), \tag{A.2.7}$$

$$C_m=i^{(m-1)}\exp[i(m-1)\delta][E_1 J_{m-1}(a)+iE_0 J_m(a)e^{i\delta}]. \tag{A.2.8}$$

The zeroth order ($m=0$) corresponds to the initial direction of the wave E_0, the $+1$ order ($m=+1$) to the initial direction of the wave E_1, and all the remaining orders correspond to the new waves that result from the self-diffraction.

If the amplitude of wave E_1 is very small, so that $a\ll 1$, the intensities in all the higher orders will be small. A nontrivial diffraction effect manifests itself in the appearance of the -1 order of diffraction, with intensity

$$|C_{-1}|^2=|E_1|^2|\mu|E_0|^2|^2, \tag{A.2.9}$$

and the intensity of the transmitted signal is equal to

$$|C_{+1}|^2=|E_1|^2|1+i\mu|E_0|^2|^2. \tag{A.2.10}$$

Expressions (A.2.9) and (A.2.10) could be obtained, of coursed also directly by perturbation theory, bypassing the expansion (A.2.8). Under typical conditions it is easy to record the intensity of the new wave $|C_{-1}|^2$ at a level $\sim 10^{-3}$ of the incident waves $|E_0|^2 \sim |E_1|^2$. This corresponds to modulation of the phase $\psi(x)-\psi_0=a\cos qx$ with a parameter $a\sim 1/30$. Since a is connected with the inclination of the director

$$\delta\varphi=2\varphi_1\cos(qx)\sin(\pi z/L)$$

and with the refraction angle α by the relation

$$a=2(\tilde{\varepsilon}_a\omega/c\tilde{\varepsilon}_e^{1/2})L\varphi_1\sin\alpha, \tag{A.2.11}$$

we get at $a\approx 5\times 10^2\varphi_1$ at $\omega/c=2\pi/\lambda_{\text{vac}}\approx 10^5\ \text{cm}^{-1}$ and $\tilde{\varepsilon}_a=0.7$. This means that by the self-diffraction method one can record values of φ_1 on the order of $|\varphi_1|\sim 10^{-4}$ rad.

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Translated by J. G. Adashko