Orientational optical nonlinearity of liquid crystals

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The orientational optical nonlinearity of the mesophases of liquid crystals (LCs) is attracting ever more attention, owing to the high values of the constants characterizing it, owing to the many interesting specific features that LCs introduce even into traditional nonlinear optical effects, and owing to the potentialities of studying the physical properties of LCs. This article presents the state of the theory and experiment of orientational interaction of light waves with LCs. Especial attention is paid to the physical mechanisms of interaction—the light-induced Fréedericksz effect, recording of director gratings, and the action of surface light waves on the orientation of the director. Varied manifestations of the effects of orientational self-action and interaction of light waves are discussed: self-focusing of light, nonlinear optical activity, mutual focusing, stimulated light scattering, and wave-front conjugation. Optical nonlinearities specific for the mesophase of a liquid crystal and associated with absorption of light quanta are also discussed. They include photoconformational nonlinearity, thermal-orientational nonlinearity, and liquid-crystal light valves. Especial attention is paid to the methodological problem of deriving the Euler-Lagrange-Rayleigh variational equations and to the correct choice of the free energy for the system LC + electromagnetic field. Section 8 traces the history of the studies of orientational optical nonlinearity of LCs and reviews the studies whose results are not directly reflected in the main text, and also reviews the studies on practical applications of light-induced orientational effects.

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INTRODUCTION

A new field in the physics of liquid crystals (LCs) has arisen and is developing vigorously in the past several years—orientational optical nonlinearity of the mesophase of LCs. It has now become evident to all that the electric field of a light wave enables one to reorient the director of an LC just as well as a static magnetic or electric field does. How-

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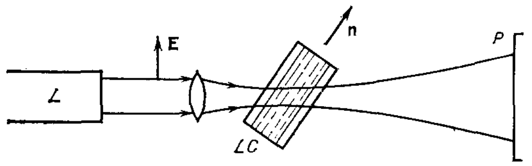

FIG. 1. Diagram of an experiment to observe GON. L—laser; LC—NLC cell; P—screen.

ever, before the first experiment, the answer to the following simple question was far from obvious: will the transparency of the mesophase of an LC (which, as we know, is not too high owing to scattering and absorption) allow transmission of light of sufficient power to a distance sufficient for observing effects of orientational nonlinearity? Fortunately, the answer was affirmative, and this ultimately led to the need for writing this review.

1. GIANT OPTICAL NONLINEARITY (GON) OF A NEMATIC: EXPERIMENT AND SIMPLE ESTIMATES

1.1. Experiment[12]

The radiation of a neon-helium laser ($\lambda = 628$ nm) of power from 0 to 20 mW was focused with a lens of focal length 25 cm into a cell containing a plane-oriented nematic liquid crystal (NLC) 60-μm thick (Fig. 1). The cell was inclined to the beam, so that the polarization unit vector of the extraordinary wave in the NLC made an angle α with the director, i.e., with the optic axis. The angular structure and the divergence of the transmitted radiation were measured in the far zone. At a low power level, $W \lesssim 3$ mW, the angular distribution of the transmitted wave was practically the same as in the absence of the cell (Fig. 2). Upon increasing the power, the angular divergence of the transmitted beam increased. In subsequent experiments at even greater power, $W \sim 20$ mW, the angular distribution in the far zone acquired a distinctive annular structure. The effect was enhanced with increasing angle α (the maximum value of α in the experiment amounted to 32°). If the cell was set up with a small shift beyond the constriction of the focused beam, i.e., in the region having a divergent wave, then as the power was increased from zero to 10 mW, the divergence not only did not increase, but even diminished below the original value. This meant that the cell acted as a positive lens that focused the divergent wave. Thus the effect was detected of self-focusing of light at the very low power level, $W \approx 3 \times 10^{-3}$ W, which corresponds to a power density at the cell of about 50 W/cm^2. The time for establishment of the effect amounted to about 10 s. Control experiments showed that, in line with the theory, the effect is absent with normal incidence of the wave on the cell ($\alpha = 0$) and for the ordinary wave for any orientation of the cell.

1.2. Estimates

Rather detailed measurements were made in the described experiment of the dependence of the strength of the nonlinear lens on the angle α and the intensity of the light. Here we shall restrict the treatment to very simple estimates that confirm that the experiment revealed precisely what was sought—orientational optical nonlinearity.

In a beam of power density 50 W/cm^2 the electric field intensity of the light wave amounts to $|E| = 0.5$ CGSE $= 1.5 \times 10^2$ V/cm. We can represent the anisotropic component of the energy density of the interaction of the field with the NLC in the form $U_{\rm E} = -(\varepsilon_{\rm a}/16\pi)|E|^2 \cos^2(\alpha - \theta)$, where $\varepsilon_{\rm a} = \varepsilon_\parallel - \varepsilon_\perp$ is the anisotropy of the dielectric permittivity of the NLC at the light-wave frequency: $\varepsilon_{\rm a} = n_\parallel^2 - n_\perp^2 = (1.71)^2 - (1.51)^2 = 0.64$ under the conditions of the experiment. For example, for $\alpha = 45°$ the change in the orientation of the director by the angle θ makes the optic axis approach the field direction of **E**, and diminishes the energy by the amount $\delta U_{\rm E}(\text{erg/cm}^3) = -(\varepsilon_{\rm a}/16\pi)|E|^2\theta$. However, a strict planar orientation is fixed at the walls of the cell, so that the perturbation θ is maximal in the middle of the cell and vanishes at the walls. The corresponding energy density of inhomogeneous deformation is $\delta U_{\rm d} \approx KL^{-2}\theta^2$, where L is the thickness of the cell, and $K \sim 10^{-6}$ erg/cm is the Frank constant. Upon minimizing the sum $\delta U_{\rm E} + \delta U_{\rm d}$, we obtain the deviation of the director by the angle $\theta = \varepsilon_{\rm a}|E|^2 L^2/32\pi K \approx 6 \times 10^{-2}$ rad under the conditions of the experiment. For $\alpha = 45°$ this deviation of the optic axis will lead to a change in the refractive index of the extraordinary wave by the amount $\delta n \approx (n_\parallel - n_\perp)\theta = 1.2 \times 10^{-2} > 0$. Consequently an additional phase shift $\delta\phi = 2\pi z \delta n/\lambda = 10$ rad arises at the distance $z = L/\cos\alpha \approx 1.4L$. This value pertains to the center of the beam where the intensity $|E|^2$ is maximal, while $\delta\phi \approx 0$ at the edges of the beam. Since $\delta n > 0$, the wave front of the central part of the beam proves to be retarded with respect to the periphery, which implies self-focusing of the beam.

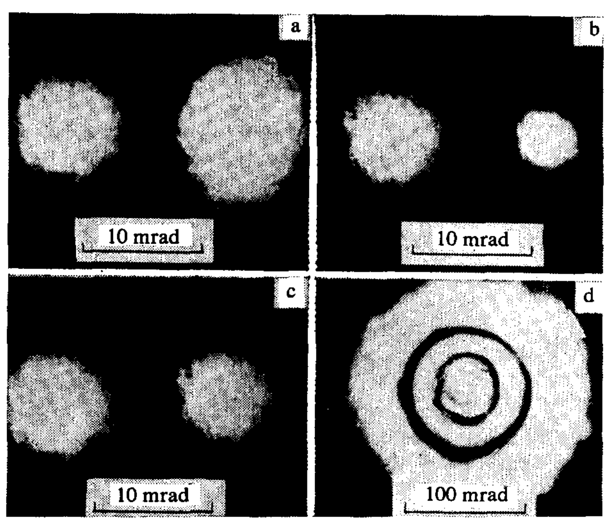

FIG. 2. Angular distribution of the beams incident on the cell and transmitted through it for different cases. a) Left—incident beam, right—beam that has undergone self-focusing broadening, at $W \sim 3$ mW; b) left—incident beam, right—beam that has undergone angular compression upon self-focusing in a cell set beyond the focal constriction; c) matching of the divergences of the incident and transmitted beams for the cases of the ordinary wave and normal incidence of the extraordinary wave; d) aberrational ring structure of the self-focusing in the transmitted beam ($W \sim 20$ mW, tenfold larger scale).

In nonlinear optics the phenomenon of self-focusing of light is generally described in terms of the dependence of the dielectric permittivity of the medium at the optical frequency on the field intensity, $\varepsilon = \varepsilon_0 + 0.5\varepsilon_2|E|^2$, where ε_2 (cm^3/erg) is the nonlinearity constant. For one of the most nonlinear media—liquid carbon disulfide, CS_2—this constant is $\varepsilon_2 \approx 10^{-10}\ cm^3/erg$. If we recalculate the results of the experiment being discussed for the value of ε_2, we obtain $\varepsilon_2 \approx 0.7\ cm^3/erg$, i.e., about 10^9 times larger than for CS_2. In line with this, the discovered nonlinearity has been named giant orientational nonlinearity (GON).

As is known, the order of magnitude of the Frank constant can be obtained from the requirement that, with 100% deformation at a scale a of the order of the dimension a_m of a molecule, the perturbed free-energy density Ka_m^{-2} must coincide with $Nk_B T$, where $N \sim a_m^{-3}$ is the density, and $k_B T$ is the temperature in energy units. This yields the estimate $K \sim Na_m^2/k_B T \sim k_B T/a_m$. On the other hand, for an isotropic liquid consisting of optically strongly anisotropic molecules, the following estimate holds[10] for the orientational nonlinearity constant: $\varepsilon_2\ (\mathrm{IL}) \sim (Nk_B T)^{-1}$. Consequently we conclude that if we have $\varepsilon_a \sim 1$ in the mesophase, then the constant corresponding to it $\varepsilon_2(\mathrm{GON}) \sim L^2/K$ is greater than $\varepsilon_2(\mathrm{IL})$ by the following factor:

$$\frac{\varepsilon_2(\mathrm{GON})}{\varepsilon_2(\mathrm{IL})} \approx \left(\frac{L}{a_M}\right)^2 .$$

And actually, if we assume that $L = 5\times 10^{-3}$ cm and $a_m = 10^{-7}$ cm, then this factor amounts to about 10^9, in agreement with the experimental results. The time for establishment of nonlinearity is increased by practically the same factor. This means that approximately the same value of $|E|^2 t_{\mathrm{pulse}}$ is required as for an isotropic liquid as for a mesophase: here t_{pulse} is the duration of the light pulse.

The subsequent sections of the review will treat also the more complex forms of deformation of LCs. For these the thickness L of the cell will no longer play the role of the dimension, but the scale of the interference pattern of the light fields will do so. If we allow for this substitution, the estimate of the gain factor derived above holds also in the general case.

2. DERIVATION OF THE SOURCE EQUATIONS OF THE THEORY

2.1. Euler-Lagrange-Rayleigh variational equations

The equations of equilibrium of a liquid crystal are commonly derived from the variational principle, according to which the free energy $\mathcal{F} = \int F\, d^3\mathbf{r}$ at constant temperature in an established state must take on its minimum value. If the density F (erg/cm^3) depends on a certain number m of independent variables $u_m(\mathbf{r})$ and their derivatives $\partial u_m/\partial x_j$, then the application of the standard methods of calculus of variations yields the system of equations of equilibrium

$$\frac{\partial}{\partial x_j}\frac{\delta F}{\delta(\partial u_m/\partial x_j)} - \frac{\delta F}{\delta u_m} = 0 \qquad (2.1)$$

(Euler-Lagrange equations). To describe the relaxation processes of establishment of a stationary state, we must also introduce the density of the dissipative function $R(\dot{u}_m)$, such that the tempo of relaxation of the energy into heat is $2R$ ($erg\cdot cm^{-3}s^{-1}$). Here the dot denotes the derivative with respect to time. Then we should write the following instead of (2.1):

$$\frac{\partial}{\partial x_j}\frac{\delta F}{\delta(\partial u_m/\partial x_j)} - \frac{\delta F}{\delta u_m} - \frac{\delta R}{\delta \dot{u}_m} = 0 . \qquad (2.2)$$

This is the Euler-Lagrange-Rayleigh equation.

Usually one selects the free-energy density F and the density of the dissipative function R from phenomenological considerations, taking into account the requirement of invariance with respect to the translation, rotation, etc., groups. At first glance one might write directly the phenomenological equations satisfying the same requirements. If we were dealing only with one independent function u, then the use of the variational apparatus actually would offer almost no advantages. However, in the presence of several variables (e.g., the Cartesian components of the director n), using the variational principles enables one to take into account automatically the reciprocity relationships, which are rather complicated to derive when one writes the equations directly. For example, the "force" $f_m = \partial F/\partial u_m$ satisfies the symmetry relationship $\partial f_m/\partial u_n = \partial f_n/\partial u_m$, since both these derivatives are equal to $\partial^2 F/\partial u_m \partial u_n$. Analogously, the very fact that the dissipative "forces" f'_m are obtained by differentiating the single function R automatically ensures satisfaction of the principle of symmetry of the Onsager kinetic coefficients $\partial f'_m/\partial \dot{u}_n = \partial f'_n/\partial \dot{u}_m = \partial^2 R/\partial \dot{u}_n \partial \dot{u}_m$.

Moreover, the existence of special forms of symmetry of the function F, which plays the role of the Lagrangian, enables one by the theorem of E. Noether to write directly the conservation laws (which are highly complex and difficult to test in the general case).

2.2. Elastic (Frank) component of the free energy

A nematic and a cholesteric are characterized by a single director unit vector $\mathbf{n}$, $|\mathbf{n}| = 1$, with $\mathbf{n}$ and $-\mathbf{n}$ deemed equivalent. The free-energy density F of the deformed state ($\mathbf{n} = \mathbf{n}(\mathbf{r})$) is taken in the form

$$F = \frac{1}{2}K_1(\mathrm{div}\,\mathbf{n})^2 + \frac{1}{2}K_2(\mathbf{n}\,\mathrm{curl}\,\mathbf{n} + q_0)^2 + \frac{1}{2}K_3[\mathbf{n}\,\mathrm{curl}\,\mathbf{n}]^2. \qquad (2.3)$$

Here we have $q_0 = 2\pi/h$, where h is the equilibrium pitch of the cholesteric helix; K_1, K_2, and K_3 are the Frank constants having the dimensionality of dynes; $q_0 = 0$ for nematics. We shall take the density of the dissipative function in the very simple form

$$R = 0.5\gamma\dot{\mathbf{n}}^2. \qquad (2.4)$$

Here γ (poise, П) is the viscosity constant. We note that we have neglected here the relationship of the director to the hydrodynamic degrees of freedom; for more details see Ref. 1.

One cannot directly employ Eqs. (2.1) or (2.2) and the free energy of (2.3), since the three quantities n_x, n_y, and n_z are interrelated by $n_x^2 + n_y^2 + n_z^2 = 1$. Here there are two ways to proceed. First of all, one can explicitly eliminate the superfluous variable, e.g., by setting $n_z = (1 - n_x^2 - n_y^2)^{1/2}$ or transform to the spherical coordinates θ and φ or any two other independent variables. Yet if for any reason it is desirable to keep a notation symmetric with respect to all three Cartesian components n_x, n_y, and n_z, then one can use the Lagrange method of the indeterminate multiplier $\lambda(\mathbf{r})$; see Ref. 1. In this method one adds the term $0.5\lambda(\mathbf{r})(\mathbf{n}^2 - 1)$ to the free-energy density F so that the variational equations take on the form:

$$\frac{\partial}{\partial x_j}\frac{\delta F}{\delta(\partial n_k/\partial x_j)} - \frac{\delta F}{\delta n_k} - \frac{\delta R}{\delta \dot{n}_k} = \lambda(r) n_k.$$

To determine the multiplier $\lambda(\mathbf{r})$, it suffices to multiply both sides of this equation by n_k, and to take into account the relationship $n_k \cdot n_k = 1$. This directly implies that the equations derived, on taking into account the relationship $\mathbf{n}^2 = 1$, have the form

$$\Pi_{ik}\left[\frac{\partial}{\partial x_j}\frac{\delta F}{\delta(\partial n_i/\partial x_j)} - \frac{\delta F}{\delta n_i} - \frac{\delta R}{\delta \dot{n}_i}\right] = 0,$$
$$\Pi_{ik} = \delta_{ik} - n_i n_k. \tag{2.5}$$

That is, they differ from the "ordinary" equations (2.1) and (2.2) by being multiplied by the projection operator onto a plane perpendicular to the local direction of $\mathbf{n}(\mathbf{r})$.

2.3. Electromagnetic component of the free energy (Lagrangian)

If we neglect the electronic nonlinearity, the interaction of the medium with the external electric ($\vec{\mathscr{E}}$) and magnetic ($\vec{\mathscr{H}}$) fields is described by contributions to the free energy of the form

$$\mathscr{F}_{\mathscr{E}} + \mathscr{F}_{\mathscr{H}} = -\frac{1}{8\pi}(\varepsilon_{ik}^0 \mathscr{E}_i \mathscr{E}_k + \mu_{ik}\mathscr{H}_i\mathscr{H}_k). \tag{2.6}$$

Here ε_{ik}^0 and μ_{ik} are the tensors of the dielectric permittivity and magnetic susceptibility. Most often in a liquid crystal ε_{ik}^0 and μ_{ik} are uniaxial tensors having the form

$$\varepsilon_{ik}^0 = \varepsilon_\perp^0 \delta_{ik} + \varepsilon_a^0 n_i n_k,$$
$$\mu_{ik} = (1 + 4\pi\bar{\chi})\delta_{ik} + 4\pi\chi_a\left(n_i n_k - \frac{1}{3}\delta_{ik}\right). \tag{2.7}$$

Here $\chi_a \sim 10^{-7}$ is the anisotropy of the magnetic polarizability, and we have $\varepsilon_a^0 = \varepsilon_\parallel - \varepsilon_\perp$, and $|\varepsilon_a^2| \sim 1$.

In going to light fields we must allow for the fact that in an electromagnetic field one has $|E| \sim |H|$. Since the anisotropy of the magnetic polarizability is seven orders of magnitude smaller than the anisotropy of the dielectric permittivity, for electromagnetic waves (including light waves) we can neglect the effect of the magnetic field of the wave on the orientation of the director. Moreover, the square of the real field intensity $E_{\rm real}^2$ must be replaced by $0.5EE^*$, where E is the complex amplitude, which is related to the real amplitude by:

$$\mathbf{E}_{\rm real} = \frac{1}{2}[\mathbf{E}\exp(-i\omega t) + \mathbf{E}^*\exp(i\omega t)].$$

Here we have dropped the terms at twice the light frequency. Finally, we must take ε_{ik} to be the symmetric real tensor of the dielectric permittivity at the light frequency. Then we have

$$F_{\rm E} = -\frac{1}{16\pi}\varepsilon_{ik}(\mathbf{r})E_i(\mathbf{r})E_k^*(\mathbf{r}). \tag{2.8}$$

The variation of the sum of Eqs. (2.3), (2.6), and (2.8) over the variables $\mathbf{n}$ for fixed $\vec{\mathscr{E}}(\mathbf{r})$, $\vec{\mathscr{H}}$, $\mathbf{E}(\mathbf{r})$, and $\mathbf{E}^*(\mathbf{r})$ and of Eq. (2.4) over the variables $\dot{\mathbf{n}}$ allows one to obtain the sought Euler-Lagrange-Rayleigh equations for the rate of relaxation of the director $\mathbf{n}(\mathbf{r},t)$ or those for equilibrium—in the stationary case. We must add to these equations the electrostatic equations curl $\vec{\mathscr{E}} = 0$, div $(\hat{\varepsilon}^0\vec{\mathscr{E}}) = 0$, and the Maxwell equations

$$-\frac{i\omega}{c}\hat{\varepsilon}\mathbf{E}(\mathbf{r}) = \mathrm{curl}\,\mathbf{H}, \quad \frac{i\omega}{c}\mathbf{H} = \mathrm{curl}\,\mathbf{E}. \tag{2.9}$$

The latter can be conveniently reduced to the single equation

$$\mathrm{curl}\,\mathrm{curl}\,\mathbf{E} - \left(\frac{\omega}{c}\right)^2 \hat{\varepsilon}(\mathbf{r})\mathbf{E}(\mathbf{r}) = 0. \tag{2.10}$$

We can employ the Maxwell equations for monochromatic fields, even when describing nonstationary effects of interaction of light with LCs, since the time of propagation of light through the specimen, $\tau \sim L/c \lesssim 10^{-11}$ s is many orders of magnitude smaller than any of the times of the orientation processes of the LC.

Remarkably, one can derive the equations (2.10) themselves from the variational principle by taking as the energy (i.e., the Lagrangian with a minus sign):

$$F_{\rm light} = \frac{1}{16\pi}\left[\left(\frac{c}{\omega}\right)^2\left(\frac{\partial E_i}{\partial x_k}\frac{\partial E_i^*}{\partial x_k} - \frac{\partial E_i}{\partial x_k}\frac{\partial E_k^*}{\partial x_i}\right) - \varepsilon_{ik}(\mathbf{r})E_i(\mathbf{r})E_k^*(\mathbf{r})\right]. \tag{2.11}$$

Here in the variation we must consider the quantities $\mathbf{n}(\mathbf{r})$ and together with them, $\varepsilon_{ik}(\mathbf{r})$, as fixed, while taking $\mathbf{E}(\mathbf{r})$ and $\mathbf{E}^*(\mathbf{r})$ as the independent variables. Thus one can derive the entire set of equations that we need by varying the free energy alone—the sum of Eqs. (2.3), (2.6), and (2.11)—with respect to the independent variables $\mathbf{n}(\mathbf{r})$, $\mathbf{E}(\mathbf{r})$, and $\mathbf{E}^*(\mathbf{r})$.

In connection with the incorrect statements found in the literature pertaining to the variational principle for LCs in light fields, we shall take up this question in somewhat greater detail.

The energy density in the light wave consists of the "electric" term $\varepsilon|E|^2/16\pi$ and the "magnetic" term $|H^2|/16\pi$. To simplify the discussions, we shall neglect here the tensor character and the frequency dispersion of the quantity ε. As we know, in a running wave we have $\varepsilon|E|^2 = |H|^2$, so that the total density of electromagnetic energy is twice as high as the "electric" term alone. Consequently the additional contribution to the energy of a dielectric in the presence of a light wave is given by the expression

$$\bar{U}_{\rm E} = 2\frac{\varepsilon|E|^2}{16\pi}. \tag{2.12}$$

Here the bar denotes averaging over the time for several light periods.

The effect of electrostriction is well known. It consists in the fact that matter is attracted into a region of greater light intensity, whereby the corresponding extra pressure δp is negative and equal to

$$\delta p = -\rho\frac{\partial\varepsilon}{\partial\rho}\frac{|E|^2}{16\pi}. \tag{2.13}$$

Here ρ is the mass density. Equation (2.13) has been verified repeatedly by theory and has been confirmed in experiments on stimulated light scattering by hypersonic waves. At first glance it contradicts Eq. (2.12), since usually one uses the formula

$$\delta p=\rho\frac{\partial U}{\partial\rho}. \tag{2.14}$$

Here U is the extra energy density. In fact, if one varies (2.12) for fixed field intensity E, then one obtains an expression twice as large as the correct magnitude in (2.13), and moreover, with the wrong sign.

The resolution of this seeming contradiction was given by L. P. Pitaevskiĭ[2]; see also Sec. 81 in the book of Ref. 3. The point is that in an adiabatic variation (i.e., sufficiently slow) of the dielectric permittivity ε, neither the field amplitude $|E|$ nor the amplitude of the displacement $|D|=|\varepsilon E|$ nor the energy density $\varepsilon|E|^2/8\pi$, nor finally the Poynting vector $|P|=c\varepsilon^{1/2}|E|^2/8\pi$, nor any of its components is conserved. Actually the conserved quantity is the adiabatic invariant, i.e., the ratio of the energy to the frequency of the radiation, which coincides with the number of quanta, apart from Planck's constant $\hbar$. In other words, in an adiabatic variation of the parameters of the medium, the number of the quantum state (i.e., the number of quanta) is conserved. On the other hand, the functional relationship $\omega\sim\varepsilon^{-1/2}$ holds for a given type of vibration (given mode of the electromagnetic field in space). (One can trace this relationship most easily with the example of a resonator of length L having the refractive index $\varepsilon^{1/2}$, where the frequency of the mth mode is determined by the condition $L\omega_m\sqrt{\varepsilon}/c=\pi m$.) As a result we find that, upon a virtual change in ε, the frequency varies virtually by the amount $\delta\omega=-0.5\cdot\omega\delta\varepsilon/\varepsilon$. Hence the virtual change in the energy $\delta\overline{U}$ can be obtained, taking into account the conservation of the adiabatic invariant $A=\overline{U}/\omega$:

$$\delta\overline{U}=\delta(\omega A)=A\delta\omega=-\frac{1}{2}A\omega\frac{\delta\varepsilon}{\varepsilon}=-\frac{1}{2}\overline{U}\frac{\delta\varepsilon}{\varepsilon}. \tag{2.15}$$

If we employ (2.15) in the variation in (2.14), then we obtain Eq. (2.13), both with the correct sign and with the correct coefficient.

To avoid resorting every time to considerations of adiabatic invariance, this result has been formulated in the following way in Ref. 2. The force exerted on the dielectric by the ac field can be obtained by a variation taken with a minus sign of the electric component of the energy alone for fixed field $\mathbf{E}$:

$$\delta\overline{U}=\delta\left(-\frac{\varepsilon|E|^2}{16\pi}\right)_{\mathbf{E}=\mathrm{const}}=-\frac{|E|^2}{16\pi}\delta\varepsilon \tag{2.16}$$

or it can be taken with a plus sign for the electric component of the energy for fixed displacement $\mathbf{D}$:

$$\delta\overline{U}=\delta\left(\frac{|\mathbf{D}|^2}{16\pi\varepsilon}\right)_{\mathbf{D}=\mathrm{const}}=-\frac{|\mathbf{D}|^2}{16\pi\varepsilon^2}\delta\varepsilon. \tag{2.17}$$

L. P. Pitaevskiĭ also showed[2] that Eqs. (2.16) and (2.17) remain valid when one takes into account the frequency-dependence of the dielectric permittivity $\varepsilon(\omega)$. One can generalize (2.16) and (2.17) in an elementary way to the case of a tensor dielectric permittivity.

Substantial errors have entered into a number of studies[6–8] on the theory of the orienting action of light on NLCs, and also into the monograph of Ref. 9 (pp. 141 and 332). We shall illustrate their essence with the example of inclined incidence of a broad light beam on a plane-parallel layer of a medium in which an electrostriction effect develops. The arguments in these studies are about as follows. Let us take the total energy density $\overline{U}$ and express it (neglecting the dispersion $\varepsilon(\omega)$) in terms of the z-component of the Poynting vector P_z:

$$\begin{aligned}P_z&=\frac{c|E|^2}{8\pi}\left(\varepsilon-\frac{q^2c^2}{\omega^2}\right)^{1/2},\\ \overline{U}&=2\frac{\varepsilon|E|^2}{16\pi}=\frac{\varepsilon P_z}{c[\varepsilon-(q^2c^2/\omega^2)]^{1/2}}.\end{aligned} \tag{2.18}$$

Here the z axis lies perpendicular to the boundaries of the layer and q is the transverse component of the wave vector. As is known, the quantity q is conserved for propagation in a medium having a z-dependent dielectric permittivity. If we vary the expression from (2.18) with respect to ε while assuming $P_z=\mathrm{const}$ (in contradiction to the idea that the adiabatic invariant is conserved in virtual variations of ε), we obtain

$$\delta\overline{U}\overset{?}{=}\left(\frac{\delta\overline{U}}{\delta\varepsilon}\right)_{P_z=\mathrm{const}}\delta\varepsilon=\frac{1}{2}\frac{\overline{U}}{\varepsilon}\delta\varepsilon\frac{\varepsilon-(2q^2c^2/\omega^2)}{\varepsilon-(q^2c^2/\omega^2)}. \tag{2.19}$$

We can see the error in this approach primarily from the fact that Eq. (2.19) has the wrong sign, even in the case of normal incidence ($q=0$). In fact, (2.19) would imply that the energy increases with the density when $(\partial\varepsilon/\partial\rho)>0$. That is, the material must be repelled from the region occupied by the field, rather than attracted as actually happens. Moreover, when $q\neq0$ also the magnitude $|\delta\overline{U}|$ from (2.19) proves to be erroneous. The concrete expressions from Refs. 6–9 do not allow one to describe the effect of deviation of the director in the first order in the light intensity in inclined incidence (GON). Thus the results of these studies contradicts not only the correct theory, but also experiment.

On returning to the problem of a liquid crystal in a light field, we repeat again: the correct equations for the director are obtained by variation of the sum of Eqs. (2.3) and (2.11) for fixed $\mathbf{E}(\mathbf{r})$. Of course, after deriving these equations, one must substitute into them the value of the vector $\mathbf{E}(\mathbf{r})$ obtained by solving the self-consistent problem in the Maxwell equations with the given (distorted) distribution of the tensor $\varepsilon_{ik}(\mathbf{r})$.

As an example, let us examine the problem of orientation by a light field having the complex amplitude $\mathbf{E}(\mathbf{r})$ of an NLC specimen with an initially homogeneous distribution of the director $\mathbf{n}=\mathbf{n}^0$. We shall also assume that the magnetic field $\vec{\mathscr{H}}=\mathscr{H}\mathbf{n}^0$ directed parallel to the unperturbed director is applied to the specimen. If we restrict the treatment to the linear terms in the perturbation $\delta\mathbf{n}(\mathbf{r},t)=\mathbf{n}(\mathbf{r},t)-\mathbf{n}^0$ (with $\mathbf{n}^0\cdot\delta\mathbf{n}=0$), then the variational Euler-Lagrange-Rayleigh equations are given by

$$\begin{aligned}&\gamma\frac{\partial\delta n_l}{\partial t}+K_2[\nabla_l(\nabla\delta\mathbf{n})+(\mathbf{n}^0\nabla)^2\delta n_l-\Delta\delta n_l]-K_1\nabla_l(\nabla\delta\mathbf{n})\\&-K_3(\mathbf{n}^0\nabla)^2\delta n_l+(K_1-K_2)n_l^0(\mathbf{n}^0\nabla)(\nabla\delta\mathbf{n})+\chi_a\mathscr{H}^2\delta n_l\\&=\frac{\varepsilon_a}{16\pi}(\delta_{ll}n_m^0+\delta_{lm}n_l^0-2n_l^0n_l^0n_m^0)E_lE_m^*\end{aligned} \tag{2.20}$$

and the Maxwell equations (2.10).

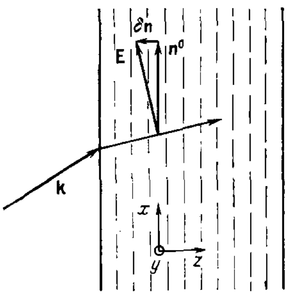

FIG. 3. Inclined incidence of the extraordinary wave on a plane NLC cell.

3. STUDIES OF GIANT OPTICAL NONLINEARITY IN CELLS CONTAINING LIQUID CRYSTALS (LCs)

Above we have already noted that the energy density with which the LC resists the orienting action of the light field declines in proportion to l^{-2} with increasing spatial scale l of the inhomogeneity of the perturbation of the director. This is precisely why, when l is of the order of the thickness of the cell, $l\sim 10^{-2}$ cm, the nonlinearity has a gigantic value, nine orders of magnitude larger than the nonlinearity of liquid carbon disulfide. Henceforth we shall denote by the term "giant orientational nonlinearity" (GON) the appearance of a perturbation of the dielectric permittivity linear in the intensity of the incident light and having the spatial scale l that is the maximum possible for the given specimen geometry. The role of l will be played by the thickness L of the cell or the transverse dimension a of the beam, if $a\lesssim L$.

3.1. Theory of GON

Let us consider a cell containing a plane-oriented NLC (Fig. 3). We shall assume the normal to the walls of the cell to coincide with the z axis, while the unperturbed direction of the director $\mathbf{n}^0$ coincides with the x axis, i.e., $\mathbf{n}^0=\mathbf{e}_x$. We shall also assume that the orientation of $\mathbf{n}$ is rigidly maintained at the walls of the cell: $\mathbf{n}(z=0,x,y)=\mathbf{n}(z=L,x,y)=\mathbf{e}_x$. Let a plane monochromatic extraordinary wave with the wave vector $\mathbf{k}$ and the complex field amplitude $\mathbf{E}=\mathbf{e}E$ propagate through the NLC. Here $\mathbf{e}=\mathbf{e}^*$ is a unit vector. We shall seek the perturbed state of the director in the form

$$\mathbf{n}(r,\ t)\approx \mathbf{e}_x+\mathbf{e}_z n_z(z,\ t)+\mathbf{e}_y n_y(z,\ t). \qquad (3.1)$$

That is, we shall consider a solution homogeneous in the xy plane. Then we obtain the following to an accuracy linear in $|E|^2$:

$$\gamma\frac{\partial n_y}{\partial t}-K_2\frac{\partial^2 n_y}{\partial z^2}=\frac{\varepsilon_a}{16\pi}(E_xE_y^*+E_x^*E_y),$$
$$\gamma\frac{\partial n_z}{\partial t}-K_1\frac{\partial^2 n_z}{\partial z^2}=\frac{\varepsilon_a}{16\pi}(E_xE_z^*+E_x^*E_z). \qquad (3.2)$$

Let the intensity of the light field be turned on jumpwise at the instant of time $t=0$. Then the solution of Eqs. (3.2) for a vector $\mathbf{E}$ not dependent on $\mathbf{r}$ can be easily derived by the method of separation of variables:

$$n_{y,z}(z,\ t)=\sum_{m=1}^{\infty}A^m_{y,z}(t)\sin\frac{m\pi z}{L}, \qquad (3.3)$$

$$A^m_{y,z}=f^m_{y,z}(t)\,|E|^2\frac{(\mathbf{e})_x(\mathbf{e})_{y,z}\varepsilon_a}{4\pi^2\gamma\Gamma^m_{y,z}}\frac{1}{m}[1-(-1)^m], \qquad (3.4)$$

$$\Gamma^m_y=\gamma^{-1}K_2\left(\frac{m\pi}{L}\right)^2,\quad \Gamma^m_z=\gamma^{-1}K_1\left(\frac{m\pi}{L}\right)^2,$$
$$f^m_{y,z}(t)=1-\exp(-\Gamma^m_{y,z}t). \qquad (3.5)$$

The coefficient $m^{-1}[1-(-1)^m]$ differs from zero only for odd values of m and corresponds to an expansion of a constant function in a sine Fourier series in the interval $(0, L)$. The constant Γ^m (s^{-1}) characterizes the rate of establishment for the corresponding sinusoidal mode. If we have the perturbation of the director given by (3.1) and (3.3), we can easily calculate to the same accuracy the perturbation of the dielectric permittivity at the light frequency, $\delta\varepsilon_{ik}=\varepsilon_a(n_i^0\delta n_k+n_k^0\delta n_i)$ and determine from it the perturbation of the phase of the transmitted wave.

Before proceeding to further calculations, let us discuss the structure of the perturbation of the director given by Eqs. (3.3)–(3.5). Even in a nonstationary regime ($\Gamma t\lesssim 1$), the mode with the lowest index $m=1$ is most strongly excited. The next mode with $m=3$ has an amplitude threefold smaller when $\Gamma^3 t\lesssim 1$. When $\Gamma^1 t\gtrsim 1$, a stationary distribution is established, for which the amplitude of the mode with $m=3$ is 27-fold smaller than that of the mode $m=1$. Hence below we shall restrict the treatment to the contribution of only this lowest mode.

If the wave vector $\mathbf{k}$ of the light wave is strictly perpendicular to the director $\mathbf{n}^0$, i.e., if $k_x=0$, then the polarization unit vector of the extraordinary wave coincides with $\mathbf{e}_x$, and perturbation of the director is absent. The perturbation of the director is identically zero also for the ordinary wave in any direction, since the polarization unit vector $\mathbf{e}$ for it is strictly perpendicular to the optic axis. Now let us examine the extraordinary wave with $k_x\neq 0$, $k_y\neq 0$. It excites deformations of both types: twist (T, $n_y\neq 0$) and transverse bending (S, splay, $n_z\neq 0$). Usually the constant K_2 for T-deformation is from two to three times smaller than the constant K_1. Hence the corresponding contribution is established more slowly ($\Gamma\sim K_i^{-1}$), but it reaches a greater stationary value with other conditions equal.

Most of the experiments have been performed in a geometry with $k_y=0$, i.e., for the case in which the wave vector lies in the plane of the normal $\mathbf{e}_z$ to the walls and of the unperturbed director $\mathbf{n}^0=\mathbf{e}_x$. Therefore for simplicity we shall restrict the discussion to precisely this special case in which T-deformation is not excited.

In order not to bore the reader with cumbersome calculations, we shall perform the calculations in the approximation of a weakly anisotropic crystal, $\varepsilon_a\ll\varepsilon_\perp$. Then we can write $\mathbf{k}=k(\mathbf{e}_z\cos\alpha+\mathbf{e}_x\sin\alpha)$, where α is the angle of refraction, $\mathbf{e}\approx\mathbf{e}_x\cos\alpha-\mathbf{e}_z\sin\alpha$, and the length of the ray path $\mathrm{d}l$ in the medium is related to the variation of the z coordinate by $\mathrm{d}l=\mathrm{d}z/\cos\alpha$. In other words, we take no account here of the small difference (of the order of $\varepsilon_a/\varepsilon_\perp$) between the directions of the group and phase velocities. We can easily obtain in the same approximation the following expression for the variation of the phase of the field owing to the perturbation $\delta\hat{\varepsilon}$:

$$\frac{d\phi}{dz}=\frac{1}{\cos\alpha}\frac{d\phi}{dl}=\frac{1}{\cos\alpha}\frac{\omega}{2cn}e_i\,\delta\varepsilon_{ik}(z)\,e_k. \quad (3.6)$$

Here $n = kc/\omega$ is the refractive index. Consequently, in a stationary regime with account taken only of the mode with $m = 1$, we obtain

$$\delta\phi=\frac{\omega}{cn}\frac{\varepsilon_a^2\sin^2\alpha\cos\alpha L^3|E|^2}{\pi^5K_1}. \quad (3.7)$$

To the same accuracy, the z component of the Poynting vector in the light wave is $P_z \approx P\cos\alpha \approx cn|E|^2\cos\alpha/8\pi$. If we write Eq. (3.7) in the form $\delta\phi = L\varepsilon_2|E|^2/4cn\cos\alpha$, then we obtain the following expression for the effective nonlinearity constant:

$$\varepsilon_2=\frac{4\varepsilon_a^2\sin^2\alpha\cos^2\alpha L^2}{\pi^5K_1}. \quad (3.8)$$

Likewise the giant orientational nonlinearity constant proves to be proportional to ε_a^2 (for moderate ε_a). After separating out the multiplier and the angular dependence $\sin^2\alpha\cos^2\alpha$, the quantity $1/\varepsilon_2$ having the dimensions of erg/cm^3 coincides with the energy density K_1/L^2 for strong ($\sim 100\%$) S-deformation of the director in a cell of thickness L.

We have considered the effects of reorientation in the first order in the intensity of the light field. Let us estimate the power density at which the angle of reorientation becomes of the order of unity. If we set $\sin\alpha\cos\alpha \sim 0.5$, then the corresponding value of $|E|^2$ is determined from the condition $\varepsilon_a|E|^2/32\pi \sim K(\pi/L)^2$. Numerically, when $L \sim 100$ μm, $K \sim 10^{-6}$ dyne, and $\varepsilon_a \sim 0.7$, the required power density amounts to about 2.5×10^3 W/cm^2. Such values are accessible by using an argon laser. No qualitatively new effects arise in a planar cell at powers greater than those indicated, while simply the nonlinearity saturates.

The problem was treated above of incidence of an infinite plane wave on a layer of an NLC. The real restriction on the applicability of this model consists of the fact that the transverse dimension a of the beam was larger than the thickness L of the layer. But if the beam is narrow, $a \lesssim L$, then the situation changes.

In this case one can conveniently calculate the self-focusing effect in the model of an unbounded single-constant NLC whose orientation along the direction $\mathbf{n}^0$ is maintained by the static magnetic field $\mathscr{H} = H\mathbf{n}^0$. Here it is convenient to choose a coordinate system with the z axis in the direction of the beam so that $\mathbf{E} = \mathbf{e}_x E(x, y)$, and $\mathbf{n}^0 = \mathbf{e}_x\cos\alpha + \mathbf{e}_z\sin\alpha$. The equation for the perturbation of the director $\mathbf{n}(x, y) - \mathbf{n}^0 \approx \theta(\mathbf{e}_x\sin\alpha - \mathbf{e}_z\cos\alpha)$ can be derived from the variational principle. It has the form

$$\frac{\partial^2\theta}{\partial x^2}+\frac{\partial^2\theta}{\partial y^2}-l_H^{-2}\theta=B\,|E(x,y)|^2. \quad (3.9)$$

Here $l_H = (K/\chi_a H^2)^{1/2}$ is the magnetic coherence length, and we have $B = \varepsilon_a\sin\alpha\cos\alpha/8\pi K$. If actually the initial orientation is maintained by the walls of the cell, rather than by a magnetic field, we can consider Eq. (3.9) approximately accurate also for this case with the substitution $l_H^2 \rightarrow L^2/\pi^2$. One can obtain the solution of Eq. (3.9) by using the Green's function:

$$\theta(x,y)=-\frac{B}{4}\int|E(x',y')|^2 iH_0^{(1)}\times(il_H^{-1}[(x-x')^2+(y-y')^2]^{1/2})\,dx'\,dy'. \quad (3.10)$$

Here $iH_0^{(1)}(iz) = 2K_0(z)/\pi$ is the zero-order Hankel function of purely imaginary argument. However, since Eq. (3.10) is rather complicated, we shall analyze separately the structure of the solution in different regions of space for different limiting cases.

If the transverse dimension a of the beam is large, $a \gg l_H$, then we can neglect the spatial derivatives in (3.9). Then the solution has the form $\theta(x,y) = l_H^2 B|E(x,y)|^2$, and we arrive at the results of the previous section. In the other limiting case when $a \ll l_H$, we must treat separately the behavior of the director inside the beam and outside it. Moreover, we shall treat separately an axially symmetric cylindrical beam, $\mathbf{E} = \mathbf{E}(\rho)$, $\rho = (x^2 + y^2)^{1/2}$, and a one-dimensional ribbon beam $\mathbf{E} = \mathbf{E}(x)$. To simplify the formulas, we shall also assume $\mathbf{E}(\rho) = \mathbf{E}_0$ inside the beam of diameter $2a$ (or of width $\Delta x = 2a$) and $\mathbf{E}(\mathbf{r}) = 0$ outside the beam—a round or rectangular table.

Inside the beam for $a \ll l_H$ the solutions of Eq. (3.9) respectively have the following forms for cylindrical and ribbon beams:

$$\theta(\rho)=\frac{\rho^2}{4}B|E_0|^2+\theta_2,\quad \theta_1(x)=\frac{x^2}{2}B|E_0|^2+\theta_1. \quad (3.11)$$

The reciprocal focal length of the corresponding nonlinear lens in these cases is

$$f_2^{-1}=\frac{\varepsilon_a^2\sin^2\alpha\cos\alpha|E_0|^2L}{16\pi K}=\frac{\varepsilon_2|E_0|^2L}{\varepsilon_0^{1/2}a^2},\quad f_1^{-1}=2f_2^{-1}. \quad (3.12)$$

The real intensity distribution in the beam is bell-shaped, and then aberrational self-focusing rings arise.[11] Their number is

$$N\approx\frac{1}{2\pi}[\phi(\rho=0)-\phi(\rho=a)]\approx\frac{L}{\lambda}\frac{\varepsilon_a^2\sin^2\alpha\cos\alpha a^2|E_0|^2}{32\pi K}. \quad (3.13)$$

We have deliberately associated the number of rings with the phase difference at the center of the beam and at its edge, rather than simply with the phase at the center of the beam. The point is that, when $a \ll l_H$, the perturbation of the director also differs from zero outside the beam, where the intensity is zero. Mathematically this is expressed in the fact that the deviation of the director in the center of the beam (θ_1 or θ_2) is determined not by the local intensity $|E_0|^2$ alone. For example, in the one-dimensional case with $a \ll l_H$ we have

$$\theta_1\approx-\frac{1}{2}Bl_H\int|E(x')|^2\,dx', \quad (3.14)$$

i.e., $|\theta_1| \sim |\theta(0) - \theta(a)|\,l_H/a$. In this case a sort of nonlinear prisms are formed on both sides of a ribbon beam. However, one can detect them only by using an auxiliary probe beam. In the case of a cylindrical initial beam, such "tails" in the distribution $\theta(\rho)$ outside the beam for $a < \rho \lesssim l_H$ must also exist. However, there the magnitude of $\theta_2/|\theta(0) - \theta(a)|$ is not so large, being of the order of $\ln(l_H/a)$. One can derive this conclusion by analyzing Eq. (3.10). To estimate the time of establishment of $\delta n(\mathbf{r}, t)$, one must add to the left-hand side of Eq. (3.9) the term $K^{-1}\gamma\partial\theta/\partial t$. Consequently one finds that, when $a \ll l_H$, a self-focusing lens must be established in the time $\tau \sim \gamma a^2/K$, while the quantities θ_1 and θ_2 together with the tails of the function $\theta(\mathbf{r})$ are established in the time $\tau' \sim \gamma l_H^2/K$.

For a cell having a homeotropic orientation (below, for brevity—for a homeotropic cell, $\mathbf{n}^0 = \mathbf{e}_z$), the effects of reorientation of the director that are linear in the light intensity are described by practically the same equations. The ordinary wave in inclined incidence does not excite GON, but the extraordinary wave deflects the director by the angle $\theta \approx n_y(z, t)$ in the plane of n^0 and the wave vector $\mathbf{k}$. Usually the constant K_3 for longitudinal bending deformation (B, bend) somewhat exceeds the constant K_1 of the S-effect, i.e., $K_3 > K_1$. Hence, other conditions being the same, the stationary GON in a homeotropic cell is somewhat weaker than in a planar cell. However, for the same reasons, a homeotropic cell is more transparent than a planar cell.

Here we have not yet touched on the light-induced Fréedericksz transition, which can develop in a homeotropic cell even at normal incidence. To observe it, one must exceed a threshold power density; for more details, see Sec. 4 below.

The influence of the finite dimensions of the beam on GON (i.e., on reorientation effects proportional to the light intensity) in a homeotropic cell must be qualitatively about the same as in a planar cell.

3.2. Experimental studies of GON

In Ref. 12, the experimental results of which were presented in Sec. 1.1, a cell was used with a planar orientation, containing an NLC made of a mixture of azoxy compounds and ester nitrile. This mixture had a nematic mesophase at room temperature and originally was taken in order to obtain a good homeotropic orientation in a large volume by applying an rf electric field (it had $\varepsilon_a > 0$ at radiofrequencies). However, for a number or reasons the electric orientation in a cell ~ 3 mm thick proved unsatisfactory, and the first experiment was designed with the same NLC with a planar orientation maintained by walls previously ground with diamond powder.

The parameters of the medium were $n_\perp \approx 1.51$, $n_\| \approx 1.71$, $K_1 \approx 8.5 \times 10^{-7}$ dyne, cell thickness $L = 60$ μm. Quantitative measurements of the effect of self-focusing of the radiation of an He-Ne laser ($\lambda = 0.628$ μm) with an ideal Gaussian transverse profile were performed by two methods. In the first of these, lenses were installed in place of the cell. Their focal distance was chosen so as to yield approximately the same change in the angular divergence of the beam as the self-focusing effect introduced. This method was convenient in the case the number of rings in the self-focusing pattern was small, $N \lesssim 1$. That is, the change in the divergence was of the order of the initial diffraction divergence of the Gaussian beam. The other method consisted in measuring, for $N \gtrsim 1$, the divergence or the number of rings in the far zone of the transmitted beam.[13] The results of both methods supplement one another well and yield practically identical values of the nonlinearity constant. Figure 4 shows the dependence of the equivalent strength f^{-1} of the nonlinear lens on the beam power (Fig. 4a) and on the angular

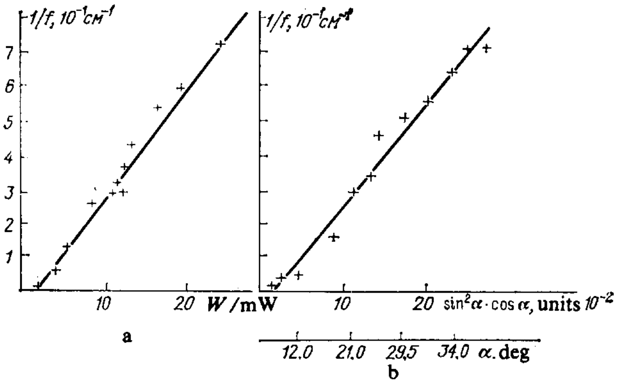

FIG. 4. Dependence of the reciprocal focal length on the beam power (a), and on the geometric factor $\sin^2 \alpha \cdot \cos \alpha$ (b).[12]

factor $\sin^2 \alpha \cos \alpha$ (Fig. 4b), where α is the angle of refraction as recalculated inside the crystal. Both graphs demonstrate a quite convincing linear dependence, in agreement with the theoretical expressions.

An absolute comparison of Eq. (3.8) with experiment requires that we know the power density $cn|E(0)|^2/8\pi$ in the focal constriction of the beam, which posed one of the most difficult problems of the experiment. To do this, one can employ the expression describing the variation of the intensity profile of an ideal focused Gaussian beam upon diffraction in air:

$$|E(x, y, z)|^2 = |E_0|^2 \left(1 + \frac{z^2}{z_0^2}\right)^{-1} \times \exp\left[-2(x^2+y^2)a^2\left(1 + \frac{z^2}{z_0^2}\right)^{-1}\right]. \quad (3.15)$$

Here we have $z_0 = ka^2/2$, $2a$ is the diameter of the focal constriction (full width at the e^{-2} level of the intensity at the maximum), $2a = FWe^{-2}M$, z_0 is the length of the constriction, $k = 2\pi/\lambda$, and λ is the wavelength in air. In the far zone this beam has a Gaussian angular distribution, $j(\theta) \sim \exp(-2\theta^2/\theta_0^2)$, with an angular width $\Delta\theta(FWe^{-2}M) = 2\theta_0 = 2/ka$. The quantity $2/ka = \lambda/\pi a$ is the diffraction divergence of a beam having the dimension a, which has a plane wave front in the constriction. If the focused beam is obtained by passing a parallel Gaussian beam of diameter $d = FWe^{-2}M$ through a lens having the focal length f, then we have $\theta_0 \approx d/2f$, and hence, $a = 2f/kd$. Here we assume that $a \ll d$. The total power transported by the beam is $P = ca^2|E(0)|^2/16$. At small angles α_{air} of incidence, the same power density (neglecting reflective losses) will exist also inside the medium.

In the experiments of Refs. 12 and 13 with focusing of the beam of an He-Ne laser with a lens having $f = 25$ cm, the values $2a \approx 240$ μm and $z_0 = 7$ cm were obtained. The values of z_0 and $2a$ considerably exceeded the thickness of the layer of LC (~ 60 μm). This justifies using the theory developed for an infinite plane wave. The value $z_0 = 7$ cm appreciably exceeded the total thickness 8 mm of the cell (LC + glass substrate). Recalculation of the experimental data for the angle $\alpha = 32°$ yields $\varepsilon_2 = 0.07$ cm^3/erg. The theoretical estimate by Eq. (3.8) yields the same value.

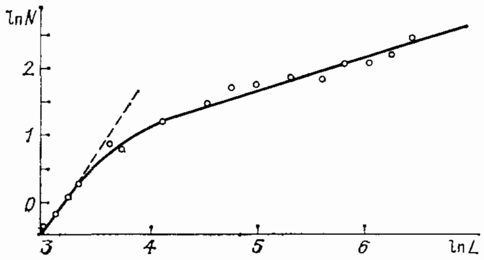

FIG. 5. Dependence of the number of rings on the thickness of the cell.[15]

It is most convenient experimentally to define the establishment time as the interval τ in which the number of self-focusing rings varies by the amount $1 - e^{-1} \approx 0.63$ from its stationary value. It proves to be the same, both on turning on the intensity and on sharply reducing it almost to zero (if the light is completely turned off there is nothing to focus). Under the conditions of the experiment,[12,13] it amounted to $\Gamma^{-1} \sim 10$ s. Hence we can estimate by Eq. (3.5) the orientational viscosity constant as $\gamma \sim 2\Pi$.

An experimental study of orientational self-focusing by the radiation of an argon laser ($\lambda = 0.49\,\mu$m) in a planar cell containing MBBA 50-μm thick was performed in Ref. 14. However, intrinsic absorption at the stated wavelength becomes appreciable specifically for this substance and leads to thermal and other effects not involving orientation (see Sec. 7).

The dependence has been studied[15] of the number N of aberration rings on the local thickness L of a planar wedge-shaped cell containing MBBA for the wavelength $\lambda = 628$ nm. For the constriction parameter $2a = FWe^{-2}M = 46$ μm, the value of L varied over the range from 17 to 500 μm. The experimental dependence of N on the parameter $\sin^2 \alpha \cos \alpha$ and on the intensity was linear. For the angle of incidence $\alpha_{\text{air}} = 55°$, the time of establishment of the ring pattern increased with increasing thickness to the value $L \sim 100$ μm and remained constant, $\tau \approx 1.7$ s, with further increase in L. The dependence of $\ln N$ on $\ln L$ is given in Fig. 5 for the same conditions of experiment. For a small thickness of the layer of LC we have $L \lesssim a\sqrt{2}$, and the $L(N)$ relationship is cubic in accord with the theory for broad beams. For larger L the experimental values of N increase somewhat more slowly than by the $N \sim L$ law implied by the theory for narrow beams. Perhaps the latter effect involves light scattering in the thick cell.

One can measure the nonlinear phase shift in GON not only from self-focusing or self-diffraction, but also by bringing about interference of the field being studied with any reference field that has not undergone a nonlinear change. If the two waves being studied have the same polarization, then interference leads to modulation of the phase and of the intensity of the overall field. But if the waves have orthogonal polarizations, then the result of interference is manifested in a change in the degree of circularity and inclination of the axis of the polarization ellipse of the overall field. In an experiment[16] to study GON, the ordinary wave obtained from the initial beam by inclined polarization played the role of the reference wave. The extraordinary wave to be studied was obtained from the same beam and yielded the GON. The essential point is that the phase of the o-wave underwent no changes upon smooth reorientation of the director. The observed[16] pattern of change of the polarization of the overall field of the transmitted wave corresponded to the GON-self-focusing of the e-wave.

GON in a homeotropic cell has been observed[17] in the focusing of the radiation of a rather powerful (~ 0.12 W) argon laser ($\lambda = 0.5145\,\mu$m) in a cell containing an NLC of the OCBP (octylcyanobiphenyl) type. The ring structure of the self-focused light was measured in this study with inclined incidence of the extraordinary wave.

A highly detailed experimental study of self-focusing and self-diffraction in homeotropic cells containing MBBA has been performed in Ref. 18. The radiation of an argon laser with $\lambda = 0.5145$ μm was focused into the specimen. The dimension $2a$ of the constriction amounted to about 40 μm. The thicknesses of the cells used were 25, 50, and 75 μm. The observed number of self-focusing rings agreed well with the theoretical predictions, both in absolute magnitude and in functional dependence on the angle of incidence (the angle α_{air} in our notation). Thus, for $\alpha = 25°$ and power density 20 W/cm^2, a cell 50-μm thick formed a nonlinear lens with a focal length $f \approx 20$ cm.

A homeotropic cell containing MBBA 75-μm thick was used in studying self-diffracton effects in Ref. 18. The incidence of two waves at an angle $\beta_{\text{air}} \sim 0.8°$ to one another yields an interference pattern $\propto \cos qx$ with the period $\Lambda = 2\pi/q = \lambda/\beta_{\text{air}} \approx 36.7\,\mu$m for $\lambda = 0.5145\,\mu$m. If we take $K_2 = 4 \times 10^{-7}$ dyne, $K_3 = 7.5 \times 10^{-7}$, and $K_2/K_3 = 0.53$ for MBBA, then with $L = 75\,\mu$m, the interference grating must be recorded weaker by a factor of $1 + (K_2/K_3)(2L/\Lambda)^2 \approx 10$ than the homogeneous component of the perturbation.

The incidence of two coherent waves at the angle β_{air} to one another yields a sinusoidal pattern of intensity distribution with the spatial period $\Delta = 2\pi/q \approx \lambda_{\text{air}}/\beta_{\text{air}}$ (for $\beta_{\text{air}} \ll 1$). The change in the dielectric permittivity under the action of light gives rise to a phase diffraction grating. The appearance of new waves diffracted by it is termed the process of self-diffraction.[19] The quantitative calculation of the amplitudes of the different orders of diffraction is based on using the well known formula $\exp(i\eta \cos \gamma) = \Sigma i^n J_n(\eta) \exp(in\gamma)$, where J_n is a Bessel function and the summation is extended over integral n from $-\infty$ to $+\infty$. As applied to liquid crystals, this process of self-diffraction was first discussed theoretically in one of the pioneer studies on orientational nonlinearity of LCs[67] and was first experimentally detected in Ref. 69. A highly detailed experimental study of self-focusing and self-diffraction in homeotropic cells containing MBBA for the wavelength of the argon laser ($\lambda = 514.5$ nm) has been performed.[18] Its results agree well with the theory. We note that the interference component of the intensity distribution under the conditions of Ref. 18 had a spatial scale about three times smaller than the thickness of the cell. In line with the theory, it yielded a response about 10 times smaller than for the homogeneous component of the exposure.

In Ref. 20 a phase grating was written in a homeotropic

NLC cell with a GON mechanism upon interference of two waves: one plane wave ($\mathbf{E}_1$) and the second having a complex wave front ($\mathbf{E}_3(\mathbf{r})$). These waves were directed into the cell at an angle of incidence of 15°. Their central directions made an 0.7° angle with one another. The readout of the hologram that was obtained $\delta\mathbf{n}(\mathbf{r}) \propto E_1E_3^*(\mathbf{r}) + E_1^*E_3(\mathbf{r})$ was performed with a wave in the opposite direction to $\mathbf{E}_1$. Owing to the readout, in addition to the other waves, a wave was generated $E_4 \propto E_1E_2E_3^*(\mathbf{r})$ that propagated in the opposite direction to the wave $\mathbf{E}_3(\mathbf{r})$ and had a wave front reversed with respect to $\mathbf{E}_3(\mathbf{r})$. With a power density of each of the three incident waves of 10W/cm^2, the efficiency of conversion was 10^{-2}. The strong effects of GON-self-focusing led to appreciable distortions of the wave $\mathbf{E}_4(\mathbf{r})$ as compared with the exactly reversed configuration.

3.3. Influence of nonrigid attachment at the boundary

Up to now we have been treating a cell with rigid attachment of the orientation of the director at the boundary. If the attachment is nonrigid, the effects of reorientation of the director and GON are altered (they prove to be stronger). Thus the possibility arises of studying the orienting action of the phase boundaries of an NLC with various media by the methods of nonlinear optics.[23] For example, let us study a planar cell in which the director is attached rigidly at the boundary $z = 0$, $\mathbf{n} = \mathbf{e}_x$, while the following boundary condition is assigned at the boundary $z = L$:

$$\frac{dn_z}{dz} + \frac{1}{R}n_z = 0. \qquad (3.16)$$

Here R is the "attachment length." One can obtain a boundary condition like (3.16) by adding to the free energy the surface term $\int F_{\rm surf}dS$, where $F_{\rm surf}$ (erg/cm^2) $= +0.5\sigma_a n_z^2$. One can term the parameter σ_a the anisotropy coefficient of the surface tension. Under the stated assumptions we have $R = K_1/\sigma_a$. The linearized Euler-Lagrange equation (2.20) for the established orientation of the director with the boundary condition of (3.16) and with $n_z(z=0) = 0$ has the solution

$$n_z(z) = -\frac{\varepsilon_a |E|^2 \sin\alpha\cos\alpha}{16\pi K_1}\left(z^2 - zL\frac{2+\xi}{1+\xi}\right), \qquad (3.17)$$

Here we have $\xi = L/R$. As $\xi \to \infty$ (rigid attachment to the second boundary as well), the solution (3.17) identically coincides with the sum of the series of (3.3) as $t \to \infty$. We note that the difference of Eq. (3.17) as $\xi \to \infty$ from the first term of the series of (3.3) is very small—of the order of 3% in the middle of the cell. This constitutes the justification of practically everywhere neglecting the higher harmonics $\propto \sin(m\pi z/L)$ with $m > 1$.

With nonrigid attachment, other conditions being the same, the perturbation of the director becomes larger. Correspondingly the phase shift also increases:

$$\delta\phi(\xi) = \delta\phi(\xi = \infty)\left(1 + \frac{3}{1+\xi}\right). \qquad (3.18)$$

As $\xi \to 0$ (completely free orientation at the boundary $z = L$), the phase shift of (3.18) increases by a factor of four as compared with two-sided rigid attachment.

If the wave vector of the light does not lie in the xz plane, i.e., if $\mathbf{k} = k(\mathbf{e}_z\cos\alpha + \mathbf{e}_x\sin\alpha\cos\beta + \mathbf{e}_y\sin\alpha\sin\beta)$, then the electric field of the extraordinary wave has a y component, and according to the equations of (3.2) it also must give rise to T-deformation, $n_y \neq 0$. For a cell with rigid attachment of the director, T-deformation only made its contribution to the nonlinear phase shift. Since the polarization adiabatically tracks the optic axis, the polarization vector at the output of a cell with "rigid" walls remains the same as in the absence of nonlinearity.

A qualitatively new effect must arise from T-deformation in a cell having one free surface. Namely, when $z = L$ here, the director proves to be rotated in the xy plane by the angle $\delta\psi = n_y$ ($z = L$), which is proportional to the intensity of the light.[21] Along with the director, the polarization vector of the transmitted light is also rotated by the same angle $\delta\psi$. The rotation angle $\delta\psi$ *per se* is small in comparison with the nonlinear phase shift $\delta\phi$ under the same conditions, $\delta\psi/\delta\phi \sim \lambda/L\varepsilon_a$. However, an important point is that the rotation of the plane of polarization can be easily observed experimentally, even when $|\delta\psi| \sim 10^{-3}$ rad. Moreover, the measurement of $\delta\psi$ can have certain advantages for NLCs as compared with the relatively small quantity ε_a

The effect that we have been discussing of rotation of the plane of polarization of light with the angle $\delta\psi \sim |E|^2$ can be called the effect of "nonlinear optical activity." The right-left asymmetry here is ensured by the very geometry of the vectors, $\mathbf{e}_z$, $\mathbf{n}$, and $\mathbf{k}$ (the vector $\mathbf{e}_z$ is directed from the "rigid" to the "soft" wall). If we write $\delta\psi$ in the form $\delta\psi = GLP$, we obtain $G \approx 6\times10^{-2}$ rad·cm/W as the value of the constant G for $\alpha \sim 30°$, $\beta = 45°$, $\varepsilon_a \approx 0.5$, and $L = 10^{-2}$ cm. As is known, the order of magnitude of the constant G for electronic nonlinearity amounts to $G \approx 10^{-3}$ rad·cm/W. If the anisotropy of the surface tension σ_a has the other sign, then we have $\xi < 0$. That is, the surface $z = L$ tends to orient the director homeotropically, rather than planarly. However, as long as $-1 < \xi$, the influence of the rigid surface $z = 0$ maintains the planar orientation throughout the thickness. Only when $\xi < -1$ does the homogeneous planar orientation become unstable. Besides, this can also be seen from Eq. (3.18), which diverges when $\xi \to -1$.

There is a number of theoretical and experimental indications[22] that the dependence of the surface energy on the orientation of the director has an appreciably more complicated form than the Rapini potential that we have been using. Hence it is of especial interest to study the orienting influence of the surface by the methods of nonlinear optics, in particular in the regime of large perturbations of the director. There are grounds for expecting a rather high accuracy of experiment, owing to the possibility of calibrating the geometric and power characteristics of the light beam in the cell containing the same NLC, but with rigidly orienting surfaces.

3.4. Cells with inhomogeneous orientation

Very interesting and specific effects should arise in the action of a light wave on a cell in which the initial orientation of the director is inhomogeneous throughout the thickness. The propagation of light in such cells under typical conditions is described in terms of independent o-and e-waves whose polarization adiabatically tracks the direction of the optic axis, i.e., the direction of the director. The quantitative criterion for such adiabatic tracking has the form $\omega|n_e - n_o|/c \gg |\partial n_i/\partial x_k|$. Upon estimating $\partial \mathbf{n}/\partial \mathbf{r}$ as $\sim 10^3$ cm^{-1}, $n_e - n_o \sim 0.1$, we obtain that, when $\omega/c \sim 10^5$ cm^{-1} ($\lambda_{vac} \sim 0.6\,\mu$m), this condition is fulfilled with much room to spare. If the cell is inhomogeneous only in thickness, $\mathbf{n} = \mathbf{n}(\mathbf{r})$, then in the propagation of light in such a medium the transverse component of the wave vector is exactly conserved. If we neglect the moderate changes ($\Delta k/k \sim (n_e - n_o)/n$) in the length of the wave vector $\mathbf{k} = k\mathbf{m}$, then the direction of propagation $\mathbf{m}$ is also conserved. In this approximation the polarization unit vectors for the ordinary and extraordinary waves can be written in the form

$$\mathbf{e}_o(z) = \frac{[\mathbf{mn}(z)]}{|[\mathbf{mn}(z)]|}, \quad \mathbf{e}_e(z) = [\mathbf{me}_o(z)]. \qquad (3.19)$$

First let us study a cell having a so-called hybrid orientation, for which the director is rigidly fixed: on one wall in the plane $\mathbf{n}(z = L) = \mathbf{e}_z$, and perpendicular to it on the other wall, $\mathbf{n}(z = 0) = \mathbf{e}_x$.

The distinguishing feature of such a cell is that always, i.e., for any direction of the wave vector $\mathbf{m}$ of the incident e-wave, regions are found for which the director and the polarization unit vector form an oblique angle. Therefore GON, i.e., an effect linear in the intensity of the incident light, always occurs here. The order of magnitude of the nonlinear phase shift here is the same as for homogeneous (i.e., planar or homeotropic) cells under their optimal conditions. We shall not take up the derivation of the corresponding expressions; see Refs. 23 and 24.

In a hybrid cell the unperturbed director lies in the xz plane:

$$\mathbf{n}(z) = \mathbf{e}_x \sin\theta(z) + \mathbf{e}_z \cos\theta(z). \qquad (3.20)$$

Here we have $\theta(z = 0) = 0$, $\theta(z = L) = \theta_L$. In the single-constant approximation, the relationship $\theta(z) = pz$ is a solution of the equations of equilibrium and the conditions of attachment to the walls yield $p = \theta_L/L$.

In the absence of a light field, small perturbations $\delta\theta$ relax to zero for any value of the parameter θ_L. In contrast to this, with respect to perturbations δn_y that correspond to moving the director out of the xz plane, the system becomes softer with increasing θ_L and loses stability at a certain crtical value of θ_L. The threshold of this instability can be easily found in the single-constant case, where it is θ_L (threshold) $= \pi$ (cell with both walls planar and with kinking of the director).

Near the instability threshold the response δn_y to the action of a light field increases according to a law of the Curie-Weiss type $(\theta_L(\text{threshold}) - \theta_L)^{-1}$. However, it is essential that the electric field of the e-wave must have a y component to excite δn_y. To do this it must have a y component of the wave vector. That is, the light must be incident outside the xz plane of the director.

Another interesting case is presented by twist cells, i.e., cells with a twisted nematic or cholesteric LC. Here the distribution of the director has the form

$$\mathbf{n}(z) = (\mathbf{e}_x \cos\psi + \mathbf{e}_y \sin\psi)\cos\zeta + \mathbf{e}_z \sin\zeta. \qquad (3.21)$$

Here we have $\psi = pz$ and $\zeta = 0$ in the unperturbed state, and the conditions for rigid planar attachment to the walls have the form $\zeta(0) = \zeta(L) = 0$, $\psi(0) = 0$, $\psi(L) = \psi_L$, $p = \psi_L/L$. Here the system is also stable with respect to small perturbations $\delta\psi$. Yet the situation is more complicated with respect to small perturbations $\delta\zeta$. We shall write the linearized system of equations for $\delta\psi$ and $\delta\zeta$ for a cholesteric with the helix parameter q_0, which in the general case does not coincide with p:

$$\gamma\frac{\partial\zeta}{\partial t} = K_1\frac{\partial^2\zeta}{\partial z^2} + [(2K_2 - K_3)p^2 - 2K_2 q_0 p]\zeta - \frac{\varepsilon_a \sin\alpha\cos\alpha\sin(pz+\beta)|E|^2}{8\pi}, \qquad (3.22a)$$

$$\gamma\frac{\partial\delta\psi}{\partial t} = K_2\frac{\partial^2\delta\psi}{\partial z^2} - \frac{\varepsilon_a\sin^2\alpha\sin 2(pz+\beta)|E|^2}{16\pi}. \qquad (3.22b)$$

When $p \approx q_0$, the system is stable to all small perturbations. If the nematic is twisted ($q_0 = 0$), then the instability of the perturbations of ζ, which draw the director out of the xy plane, is realized when $(\psi_L/\pi)^2 > K_1/(2K_2 - K_3)$. For comparison with the experiment of Ref. 25, we shall restrict the treatment to the case $q_0 = p$. Without writing explicitly the solutions of Eqs. (3.22), we point out that the characteristic scale of the inhomogeneity of the perturbations is $\Delta z \sim p^{-1}$. Hence the order of magnitude of the elastic energy amounts to Kp^2. Therefore, when $pL \gg 1$, the optical nonlinearity proves to be substantially suppressed as compared with the case of a homogeneous cell containing an NLC. Let us present here the final answer for the contribution to the nonlinear phase shift,[25] which is proportional to the thickness L of the layer:

$$\delta\phi = \frac{\omega}{cn_e}\frac{\varepsilon_a^2\sin^2\alpha|E|^2 L}{8\pi q^2\cos\alpha}\left(\frac{\sin^2\alpha}{16K_2} + \frac{\cos^2\alpha}{K_1+K_3}\right). \qquad (3.23)$$

An experiment[25] was set up using the nematic 5CB (*p*-amyl-*p*-cyanobiphenyl), to which was added from 0.1 to 1% by weight of cholesteryl chloride to create the required pitch of the helix satisfying the condition $q_0 L = \pi n$. Here n is an integer and $L = 60\,\mu$m is the thickness of the cell. The radiation of an argon laser was employed, $\lambda = 0.51\,\mu$m, with power up to 200 mW. We can conveniently introduce the parameter η, which characterizes the ratio of the nonlinear phase shift in a CLC cell to the shift in an NLC cell of the same thickness for the same values of the power, K_i, $n_\parallel$, $n_\perp$, and the angle of incidence α_{air}. Figure 6 shows the experimental dependence of the parameter η on the reciprocal helix pitch h^{-1}. The solid line is drawn by Eqs. (3.23) and (3.7). We observe very good agreement.

We have discussed above the orientational nonlinearity of a CLC involving the change in the structure of the orientation within the limits of a single period. This nonlinearity is weak in comparison with GON, since it contains the small parameter $(h/L)^2$. It is of interest to elucidate whether a form of GON can be realized for a CLC where the direction of the *axis* of the cholesteric helix undergoes major variations in space for the case opposite to the adiabatic Mauguin limit. Calculation shows that for wide beams ($a \gg L$) a GON effect of this type in a CLC having the constant $\varepsilon_2 \approx \varepsilon_a^2 L^2/K_i$ cannot occur if the director is rigidly attached, even to one of the surfaces. The point here is that bending of the axis of the

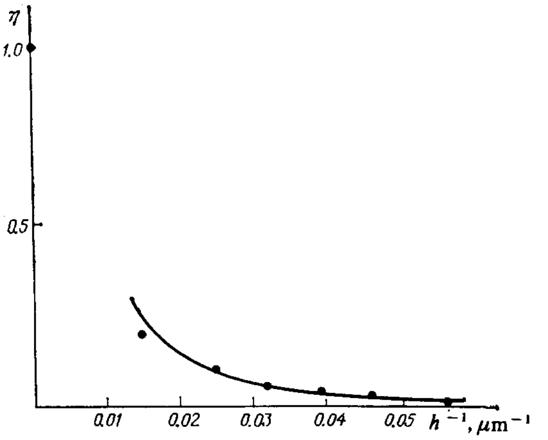

FIG. 6. Dependence of the parameter η on the reciprocal pitch of the helix h^{-1}.[25]

helix under these conditions unavoidably leads to local changes in the pitch δh to give rise to a large elastic energy $F \sim (\delta h/h)^2 (K/h)^2$. The highly interesting problem of GON for CLCs in narrow beams with $a \lesssim L$ requires separate treatment.

3.5. C smectics

The deformation energy of smectics (SLCs) consists of a large term, $0.5B_0(\partial u/\partial z)^2$, involving the change in the interlayer distance, and relatively "soft" terms that describe the different forms of deformation that do not affect the distance between the layers. Here $u(z)$ is the displacement of the layer and the quantity B_0 (erg/cm^3) can be estimated as $B_0 \sim K/a_m^2$. With the molecular dimension $a_m \sim 10^{-7}$ cm (which approximately coincides with the distance between layers), we have $B_0 \sim K \cdot 10^{14}$ cm$^{-2} \sim 10^8$ erg/cm^3. We are interested in the large effects of orientational nonlinearity. They correspond only to "soft" orientational modes with fixed interlayer distance.

Since experimentation on C smectics is still in the qualitative stage, we shall not write out the corresponding expressions explicitly (see Refs. 26 and 27), but shall restrict the treatment only to estimates and discussion of the qualitative features. Let the smectic layers be planar and directed perpendicular to the z axis (Fig. 7). In this case the molecules of the SLC are oriented along the director **n**, which makes the fixed angle θ with the z axis:

$$\mathbf{n}(\mathbf{r}) = \mathbf{e}_z \cos\theta + \mathbf{c}(\mathbf{r}) \sin\theta,$$
$$\mathbf{c}(\mathbf{r}) = \mathbf{e}_z \cos\psi(\mathbf{r}) + \mathbf{e}_y \sin\psi(\mathbf{r}). \qquad (3.24)$$

Here $\mathbf{c}(\mathbf{r})$ is a unit vector lying in the plane of the layers. In the undeformed state the **c**-director (the vector **c**) is constant throughout the volume. We can write the deformation energy in a quadratic form in the spatial derivatives of the vector $\mathbf{c}(\mathbf{r})$.

If the angle θ is small, then one can consider the SLC-C medium to be optically uniaxial as in the case of a nematic. The greatest nonlinearity is reached when the unperturbed director **n** and the polarization unit vector $\mathbf{e}_e$ of the extraordinary wave lie about 45° apart. Its magnitude and time of establishment must be about the same as for the corresponding nematic. When the direction of the normal to the smectic layers, which we have denoted as $\mathbf{e}_z$ is fixed, the director **n** can lie only within the limits of the cone of (3.24) with a fixed value of θ. Hence, to obtain the nonlinear response of the polarization unit vector $\mathbf{e}_e$, the director **n** and the normal $\mathbf{e}_z$ to the layers cannot lie in one plane. Moreover, when the value of the parameter θ is small, the maximal value of the variation of the tensor $\hat{\varepsilon}$, which is obtained at $\delta\psi = 180°$, will be of the order of $\varepsilon_a^2 \sin^2\theta$. This means that, when $\theta \ll 1$, the orientational nonlinearity of the SLC-C is saturated at a power smaller by a factor of θ^{-2} than for a nematic having the same parameters.

Experiments on the action of laser radiation on SLC's have been performed in Refs. 27 and 28. Unfortunately the results of these studies are qualitative in nature.

Qualitatively new effects must arise in a SLC-C when one takes into account the fact that they are biaxial in their optical properties. In particular, an orientational nonlinearity must arise here even for the polarization that corresponds to the o-wave in a nematic. Besides, it will be rather weak, and in fact proportional to the square of the difference of the eigenvalues of the tensor $\hat{\varepsilon}$ that coincide in the nematic or in the smectic.

Analogously to the case of a nematic, when one employs a cell containing a SLC-C with one free surface, nonlinear optical activity must arise, i.e., rotation of the plane of polarization induced by light.

In A smectics the orientation of the molecules (the **n**-director) is parallel to the normal to the smectic layers, and the degree of freedom corresponding to the **c**-director in an SLC-A is absent. Therefore the reorientation of the director in an SLC-A must generally be accompanied by deformation of the layers. Owing to the practical incompressibility of the interlayer distance, such a deformation propagates in an SLC-A to great distances, up to the boundaries of the cell. If the orientation of the director (or layers) is rigidly fixed at the boundaries of the cell, then the effect of GON must be absent in the SLC-A.

4. LIGHT-INDUCED FRÉEDERICKSZ TRANSITION (LIFT)

The Fréedericksz effect in static or radiofrequency fields (electric, $\vec{\mathscr{E}}$, or magnetic, $\vec{\mathscr{H}}$) is well known. In the

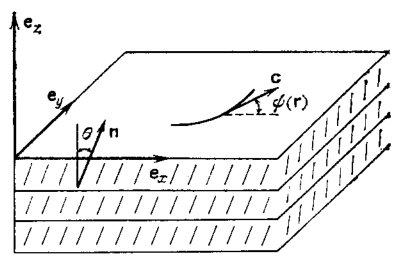

FIG. 7. Plane layered structure of a C smectic. The normal $\mathbf{e}_z$ to the layers is constant throughout the volume, and the orientation **n** of the molecular axes makes the angle θ with the normal $\mathbf{e}_z$. The projection of the vector **n** on the plane of the layers characterizes the c-director; the angle between the c-director and a certain unperturbed direction is denoted by $\psi(\mathbf{r})$.

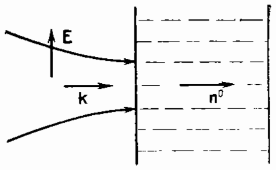

FIG. 8. A geometry in which the reorientation of the director occurs in threshold fashion.

broad sense, one takes the Fréedericksz effect to mean any reorientation of the director by external fields on the scale of the entire cell[29] (see Sec. 4.2). When the directions of the director and the reorientation field do not coincide and are not strictly perpendicular to one another, even in weak fields a perturbation $\delta\theta \sim \mathscr{E}^2$ or $\delta\theta \sim \mathscr{H}^2$ arises from the Fréedericksz effect. However, in the very important special case in which the unperturbed direction of the director corresponds to the maximum energy of interaction with the field (e.g., $\mathbf{n}^0 \| \vec{\mathscr{H}}$ when $\chi_a < 0$), reorientation arises only beyond some threshold value of the square of the field. In this case the Fréedericksz effect is of threshold type.

As we showed above, the light field also reorients the director of the LC. When the light field is incident oblique to the director, this reorientation occurs even in weak fields with $\delta\theta \sim |E|^2$. The effect is easily observable at an intensity ~ 50 W/cm^2.

A threshold effect of reorientation of the director by a light field has been experimentally detected[17] upon using a far more powerful light beam ($\sim 10^3$ W/cm^2). This effect is observed in normal incidence of the light beam on a homeotropically oriented cell containing an NLC (Fig. 8). At such a high power density P, even a moderate relative excess over the threshold, $P - P_{\text{thr}} \sim 0.3 P_{\text{thr}} \sim 300$ W/cm^2, leads to very strong reorientation, and as a consequence, to a very great self-focusing increase in the angular divergence of the transmitted beam. The threshold effect found in Ref. 17 has been termed a light-induced Fréedericksz transition (LIFT). According to the tradition that has been established by now, this name is applied to denote precisely the threshold effect. However, a reorientation of the director that is linear in the intensity of a weak field is called giant orientational nonlinearity.

In this section we shall first present the results of the theory with the aim of using them later is discussing experiment.

4.1. Broad beams: threshold behavior

Let us study the case of strictly normal incidence of a light wave with the polarization $\mathbf{e}_x$ on a homeotropic cell (see Fig. 8). For a broad beam, $a \gg L$, one can restrict the treatment to the problem homogeneous in the xy plane. Then, from symmetry considerations the vector of the field throughout the cell will remain in the xz plane: $\mathbf{E}(z) = \mathbf{e}_x E_x(z) + \mathbf{e}_z E_z(z)$. The appearance of a z component arises from the action of the nondiagonal components of the tensor $\varepsilon_{xz} = \varepsilon_{zx}$ that appear when the director is deflected from the unperturbed state. We shall write the field of the director in the form of (3.21): $\mathbf{n}(z,t) = \mathbf{e}_z \cos\theta + \mathbf{e}_x \sin\theta$; $\theta = \theta(z,t)$. The equations for θ have the form

$$\gamma \frac{\partial\theta}{\partial t} = (K_1 \sin^2\theta + K_3 \cos^2\theta) \frac{\partial^2\theta}{\partial z^2} - (K_3 - K_1)\sin\theta\cos\theta \left(\frac{\partial\theta}{\partial z}\right)^2 + \frac{\varepsilon_a}{16\pi}\left[\sin 2\theta\,(|E_x|^2 - |E_z|^2) + \cos 2\theta\,(E_x E_z^* + E_x^* E_z)\right]. \quad (4.1)$$

Equation (4.1) and the corresponding Maxwell equations have the exact solution $\theta \equiv 0$, $\mathbf{E} = \mathbf{e}_x E \exp(i\omega n_\perp z/c)$. To determine the threshold, one must linearize Eq. (4.1) in the small deflection θ. The modification of the field by the perturbation of the director can be determined with the required accuracy from the equation

$$\operatorname{div}\mathbf{D} = \frac{\partial D_z}{\partial z} = \frac{\partial(\varepsilon_{zx}E_x + \varepsilon_{zz}E_z)}{\partial z} = 0.$$

This equation yields $E_z = -\varepsilon_{zx}/E_x/\varepsilon_{zz}$, so that we have the following in an approximation linear in θ: $E_z \approx -\varepsilon_a \theta E_x/\varepsilon_\|$. As a result the linearized problem is described by the equation

$$\gamma \frac{\partial\theta}{\partial t} = K_3 \frac{\partial^2\theta}{\partial z^2} + \frac{\varepsilon_a}{8\pi}\left(1 - \frac{\varepsilon_a}{\varepsilon_\|}\right)|E_x|^2\theta. \quad (4.2)$$

The correction term $\varepsilon_a/\varepsilon_\|$ in (4.2) corresponds to taking into account the z-component of the light field that arises on perturbing the director; this correction amounts to ~ 0.3 when $\varepsilon_a \sim 0.9$, $\varepsilon_\| = 3$. Precisely this correction distinguishes the exact calculation of the LIFT threshold from the simple replacement in the corresponding expressions for the rf electric field. The solution of Eq. (4.2) with the boundary conditions $\theta(z=0,t) = \theta(z=L,t) = 0$ has the form

$$\theta(z,t) = \sum_{m=1}^{\infty} c_m \exp(-\Gamma_m t)\sin\frac{m\pi z}{L}, \qquad \Gamma_m = \frac{K_3}{\gamma}\left(\frac{m\pi}{L}\right)^2 - \frac{\varepsilon_a}{8\pi\gamma}\left(1 - \frac{\varepsilon_a}{\varepsilon_\|}\right)|E_x|^2. \quad (4.3)$$

The smoothest perturbation, $m = 1$, becomes unstable ($\Gamma < 0$) before the others. This corresponds to the threshold condition $\varepsilon_a |E|^2[1 - (\varepsilon_a/\varepsilon_\|)](8\pi)^{-1} = K_3(\pi/L)^2$:

$$P_{\mathrm{Fr}}\,(\text{erg/cm}^2\text{s}) = \frac{c n_\perp |E|^2}{8\pi} = \frac{c\varepsilon_\| K_3}{\varepsilon_a \varepsilon_\perp^{1/2}}\left(\frac{\pi}{L}\right)^2. \quad (4.4)$$

Near the threshold the characteristic times of development (or decay) of perturbations are retarded:

$$\exp(-\Gamma t) = \exp\left[-\frac{\pi^2}{\gamma L^2}\left(1 - \frac{P}{P_{\mathrm{Fr}}}\right)t\right]. \quad (4.5)$$

Just as for Fréedericksz transitions in quasistatic fields (P is the power density of the radiation incident on the NLC).

To determine the LIFT threshold in a beam of arbitrary polarization state, we shall write the perturbation of the director in the form

$$\mathbf{n}(z,t) \approx \mathbf{e}_z + \mathbf{e}_x \delta n_x(z,t) + \mathbf{e}_y \delta n_y(z,t).$$

Moreover, to an accuracy linear in δn_x, δn_y, we find from the equation $\operatorname{div}\mathbf{D} = 0$ that $E_z \approx -(\varepsilon_a/\varepsilon_\|)(\delta\mathbf{n}\cdot\mathbf{E})$. Consequently the equation for the perturbation of the director acquires the form

$$\gamma \frac{\partial\,\delta n_i}{\partial t} = K_3 \frac{\partial^2 \delta n_i}{\partial z^2} + \frac{\varepsilon_a}{8\pi}\left(1 - \frac{\varepsilon_a}{\varepsilon_\|}\right)\frac{E_i E_k^* + E_i^* E_k}{2}\,\delta n_k, \quad (4.6)$$

Here the subscripts i and k run through the two values x and y. Thus the LIFT threshold is determined by the symmetrized matrix composed of the x- and y-components of the unperturbed field.

While referring to the original study[30] for the details, we shall present the final result for the LIFT threshold:

$$P_{\mathrm{Fr}}\,(\mathrm{erg/cm^2 s}) = \frac{c\varepsilon_\perp^{1/2}\langle EE^*\rangle_{\mathrm{thr}}}{8\pi} = \frac{\pi^2 c\varepsilon_\parallel K_3}{\varepsilon_a\varepsilon_\perp^{1/2}L^2}\,\frac{2}{1+(\xi_1^2+\xi_3^2)^{1/2}}. \tag{4.7}$$

Here $\xi_i = (\xi_1, \xi_2, \xi_3)$ is the so-called Stokes vector[31] that characterizes the polarization state of the radiation. If the radiation is fully depolarized ($\xi_1 = \xi_2 = \xi_3 = 0$) or if it is circularly polarized ($\xi_1 = \xi_3 = 0$, $|\xi_2| = 1$), then the threshold proves to be twice as high as that for linearly polarized radiation. In the case $\xi_1 = \xi_3 = 0$, the eigenvectors of the symmetrized matrix $E_iE_k^* + E_i^*E_k$ are not defined. Hence the direction of the perturbation in an established superthreshold state must be governed by more subtle effects (e.g., by the transverse distribution of the intensity in the beam).

4.2. Broad beams: superthreshold structure

To determine the superthreshold stationary structure, we need a more accurate solution of the Maxwell equations (2.9) in the inhomogeneous medium. The complex amplitude of the field varies, so to speak, by 100% if the phase shifts by the amount $\delta\phi \sim 1$ radian. For normal incidence of the light, this corresponds to a deflection of the director $\theta \sim [\lambda/2\pi L(n_\parallel - n_\perp)]^{1/2}$. For $\lambda \sim 0.5\,\mu$m and $L \sim 100\,\mu$m, this yields the estimate $\theta \sim 0.05$ radian. Actually, in the case of a Fréedericksz transition in the field of the ordinary wave (see below, Sec. 5.4), the nonlinearity of the system of equations involving the strong modification of the field *vector* takes effect, even at such small distortions of the director. Here we can neglect the "elastic" nonlinearity of the LC. However, for a broad, linearly polarized beam, one can treat the problem as homogeneous in the xy plane; the field vector remains in the xy plane. The action of the light on the liquid crystal is determined only by the modulus and the direction of the vector E. The strong phase change of the field (of the order of $\omega(n_\parallel - n_\perp)L\theta^2/c$) in this case does not affect its modulus in any way. The direction of the complex vector E changes only by an amount of the order of θ, approximately in the same way as for a Fréedericksz transition in a static field.

For a broad, linearly polarized beam, the problem of the stationary superthreshold structure of the director for LIFT has been solved in Refs. 32 and 30. While referring the reader to these studies for the details, we shall present here the results for the case of a small excess of the flux density P over the threshold value P_{Fr}:

$$\theta(z) = \theta_1 \sin\frac{\pi z}{L} + \theta_3 \sin\frac{3\pi z}{L} + \ldots, \tag{4.8}$$

$$\theta_1 = \pm u^{-1/2}\left(2\,\frac{P-P_{\mathrm{Fr}}}{P_{\mathrm{Fr}}}\right)^{1/2}, \tag{4.9}$$

$$u = 1 - \frac{9}{4}\frac{\varepsilon_a}{\varepsilon_\parallel} - \frac{K_3-K_1}{K_3}, \tag{4.10}$$

$$\theta_3 = \frac{1}{48}\theta_1^3\left(1 - \frac{9}{4}\frac{\varepsilon_a}{\varepsilon_\parallel} - 3\,\frac{K_3-K_1}{K_3}\right). \tag{4.11}$$

Equation (4.9) yields an important conclusion: the quantity $\theta_1 \approx \theta(z = L/2)$ increases very rapidly ($\propto (P - P_{\mathrm{Fr}})^{1/2}$) as the power exceeds the threshold value. For example, for MBBA we have $u = 0.28$, so that we obtain $\theta_1 \approx 0.5$ radian $\approx 30°$, even when $(P - P_{\mathrm{Fr}})/P_{\mathrm{Fr}} \approx 4\%$. For OCBP the parameter u is 0.24. In the same approximation the nonlinear phase shift is

$$\delta\phi = \frac{\omega}{2c}\,L\,\frac{\varepsilon_\perp^{1/2}\varepsilon_a}{\varepsilon_\parallel u}\,\frac{P-P_{\mathrm{Fr}}}{P_{\mathrm{Fr}}}. \tag{4.12}$$

The numerically small quantity u characterizes the compliance of the system toward the superthreshold action, $\theta_1 \propto [(P - P_{\mathrm{Fr}})/u]^{1/2}$. In a simplified treatment of LIFT, i.e., in the single-constant approximation and without taking into account the reverse influence of the reorientation of the director on the field ($\varepsilon_a \to 0$), this parameter would be unity.

Very importantly, for certain NLCs the value of the parameter u can prove even to be negative. For example, for PAA at $T = 125$°C we have $u = -0.12$. In this case the relationship (4.9) has no region of applicability at all. One can show from the more exact solution that LIFT in broad beams for NLCs with $u < 0$ will show hysteresis. As the intensity increases from zero, the transition will occur at the power density p defined by the relationship (4.9), Fig. 9. However, θ_m will increase jumpwise in the transition. As one goes backward in power, the transition to the state $\theta = 0$ also will occur jumpwise, but at a lower value of the power.[1)]

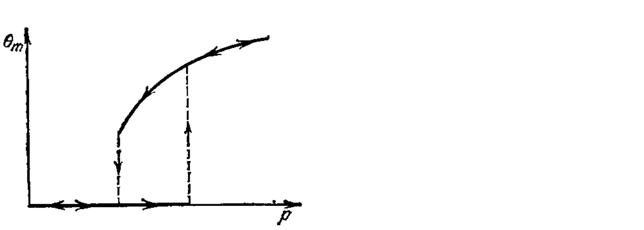

FIG. 9. Hysteresis of a light-induced Fréedericksz transition (the arrows indicate the direction of change of the power of the light field).

4.3. Effects of transverse finiteness of the beam

For a beam of limited transverse dimensions, the Frank energy acquires an additional term $\delta F = K/l^2$, where l is the transverse dimension of the perturbation. Hence a crude estimate of the LIFT threshold in this case has the form

$$|E|^2_{\mathrm{thr}} \geqslant \frac{8\pi K}{\varepsilon_a}\left(\frac{\pi^2}{L^2} + \frac{1}{l^2}\right) \quad (?). \tag{4.13}$$

Just as in the case of GON, the form of the exact answer proves to depend substantially on whether we are dealing with a one- or two-dimensional problem with respect to the transverse coordinates. Namely, if the dimension of the beam is small, $a \ll L$, then in the one-dimensional case the dimension l of the perturbation proves to be of the order of $l \sim (aL)^{1/2}$, so that $|E|^2_{\mathrm{thr}} \sim K/aL$. In contrast, in the two-dimensional problem an estimate of the form of (4.13) is valid with logarithmic accuracy when $l = a$.

For more details on the effects of transverse finiteness of the beam in LIFT, see Refs. 4, 5, and 30. The analogy of the equations for the transverse distribution of the director with the stationary Schrödinger equation is employed in Ref. 30, while Ref. 4 uses the Ritz variational procedure to find the threshold. As regards the superthreshold structure in LIFT in narrow beams, it is difficult to expect here a good analytic theory, since usually the light wave strongly alters both the direction of propagation and the polarization, even with a small excess over the threshold and even within the limits of the cell.

4.4. Experimental studies of LIFT

A light-induced Fréedericksz transition has been found[17] upon illuminating a homeotropic cell containing octylcyanobiphenyl (OCBP) with the radiation from a transverse single-mode argon laser, $\lambda = 0.5145\,\mu$m, power up to 0.2 W, radius of the constriction at the focus of the lens $\sim 5\times 10^{-3}$ cm, and power density up to 2×10^3 W/cm^2. In agreement with the results obtained earlier experimentally and theoretically for a planar cell, with inclined incidence of the extraordinary wave one observes threshold-free self-focusing arising from GON. With normal incidence of the light wave on the cell, the wave vector is parallel to the unperturbed direction of the optic axis (the director), and at low intensity self-focusing was absent. However, starting at an intensity of the order of 70–80 mW (for a cell 150-μm thick), after a rather considerable time, from 10 s to 3 min, a very strong angular divergence of the transmitted beam $\sim 10°$ arose, accompanied by a characteristic pattern with a large number $N\sim 20$–50 of aberrational rings.

LIFT was recorded in Ref. 33 for a circularly polarized beam. The threshold power proved to be about two times higher than for linearly polarized radiation. This agrees well with the theory.

For broad beams the threshold power density P (W/cm^2) must decline as L^{-2} upon increasing the thickness of the cell. This statement has not been verified in its pure form. However, the absolute values of the experimentally measured threshold power densities agree well with the theory.

For narrow beams the dependence of the threshold intensity on the transverse dimension of the beam has been studied experimentally and theoretically in Ref. 4. The results obtained agree well with the theory presented in Refs. 4, 30, and 32.

The dependence of the LIFT threshold on the temperature, according to the theory, is mainly determined by the factor $P_{\rm Fr} \propto \varepsilon_{\rm a}^{-1}K_3$. Since K_3 declines as $T\to T_{\rm c}$ as the square of the order parameter, $K\propto S^2$, while $\varepsilon_{\rm a}\propto S$, $P_{\rm Fr}\propto S$ must also decrease along with decreasing S with increasing temperature. In the experiment[34] for LIFT in an OCBP cell, a lowering of the threshold was measured from $P = 51$ mW to $P = 24.5$ mW as the temperature varied from 34 °C to 39.4 °C ($\lambda = 0.515\,\mu$m, cell thickness 150 μm, transverse radius of the beam 18.6 μm at the constriction). In this same study LIFT was measured by using the radiation of a neon-helium laser ($\lambda = 0.628\,\mu$m) and two lines of an argon laser ($\lambda = 0.476\,\mu$m and $\lambda = 0.515\,\mu$m).

Under conditions above the LIFT threshold the polarization of the transmitted light proves to be inhomogeneous, depending on the angle of incidence even for a linearly polarized incident beam. There are two different causes of the deviation of the polarization from the initial direction. Both causes are based on the fact that the polarization unit vector of the extraordinary wave under conditions of large birefringence tracks the local direction $\mathbf{n}$ of the optic axis and the local direction $\mathbf{k}$ of propagation: $\mathbf{e}_{\rm e} = {\rm const}\cdot\mathbf{k}\times(\mathbf{k}\times\mathbf{n})$.

In one of the mechanisms[30] one takes into account the fact that the perturbations of the director $\mathbf{n}$ are nonplanar in type when $K_2\neq K_1$: $\mathbf{n} = \mathbf{e}_z + \mathbf{e}_x\delta n_x + \mathbf{e}_y\delta n_y$. Here we have $\delta n_y\neq 0$, even with x-polarization of the incident light.

The other mechanism[35,36] substantially rests on the fact that, even inside the medium, the rays undergo a considerable self-focusing deviation ($\sim 10°$–$20°$). Owing to the change in the vector $\mathbf{n}$, the polarization vector, which adiabatically tracks it, must also change.

While referring to the original studies[30,36] for the details, we point out that experimentally the rotation and ellipticity of the polarization of light transmitted in LIFT have been studied in Refs. 17 and 36. When the threshold is slightly exceeded (small deviations of the rays), the depolarization pattern approximately corresponds to the first mechanism, while in the case of large divergence of the beam (considerably exceeding the threshold), the experimental results[36] agree with the second mechanism.

The problem is very interesting of the dynamics of nonstationary processes of establishment and relaxation of the orientation upon turning the field on and off. For broad beams the relaxation $\delta\mathbf{n}\propto\exp(-\Gamma t)$ is determined by the relationship $\Gamma = K_3(\pi/L)^2/\gamma$, which is valid both for GON and for LIFT (and generally for a Fréedericksz effect of any type). For narrow beams ($a\ll L$), the major part of the perturbation relaxes with the constant $\Gamma\sim Ka^{-2}/\gamma$ as the field is turned off to zero. This statement also holds both for GON and LIFT.

Near the threshold of LIFT instability (both above and below it), all the processes are retarded by a factor of approximately $P_{\rm thr}/|P - P_{\rm thr}|$; cf. Eqs. (4.3) and (4.5). This statement is valid both for broad and narrow beams. The situation here, just as for a Fréedericksz transition in nonoptical fields, recalls a second-order phase transition.

The concrete expressions depend on the form of the transverse intensity distribution, on the relationships between the constants K_2/K_1, K_2/K_3, and $\varepsilon_{\rm a}/\varepsilon_\perp$, and on the initial and final power. We must acknowledge that a quantitative theory of the effects near the established state above the LIFT threshold for narrow beams does not exist, since one must solve in self-consistent fashion the entire 100%-nonlinear three-dimensional problem for the Frank and Maxwell equations, even when the threshold is very slightly exceeded, owing to the strong saturation.

The exponential growth of small perturbations upon turning on the power above the threshold and the decay again of the small deviations of the director from the direction $\mathbf{n}^0 = \mathbf{e}_z$ upon reducing the power to values below the threshold are of greatest interest. We recall that, when $\theta_m\ll 1$ and $\theta(z) = \theta_m\sin(\pi z/L)$, the phase shift at the center of the beam for normal incidence of the light wave on a homeotropic cell amounts to

$$\delta\phi = 2\pi\frac{L}{\lambda}\frac{\varepsilon_a\varepsilon_\perp^{1/2}}{4\varepsilon_\parallel}\theta_m^2 \equiv 2\pi\frac{L}{\lambda}(n_\parallel - n_\perp)\frac{n_\perp(n_\parallel+n_\perp)}{4n_\parallel^2}\theta_m^2. \tag{4.14}$$

Hence $\delta\phi \propto \theta_m^2$ must vary according to the law $\exp(-2\Gamma t)$. In this relation, which is characteristic of LIFT, normal incidence yields a result differing from the case of GON, where $\delta\phi \propto \theta \propto \exp(-\Gamma t)$. In other words, for LIFT the given relaxation constant or increment Γ of the director yields twice as large a constant 2Γ for the number of rings $N = \delta\phi/2\pi$.

In practically all studies, the experimental investigation of the dynamics of reorientation in LIFT has been performed by observing the LIFT. Just as in a nonoptical Fréedericksz transition, the increment of the perturbations $\delta\theta(t) = \delta\theta_0\exp(|\Gamma|t)$ starts from the small level of the initial fluctuations $\delta\theta_0$. The sign of the resulting perturbation is determined by the sign of $\delta\theta_0$ and varies by a random law. The time for reaching the stationary state is given by the expression

$$\tau \approx \frac{1}{|\Gamma|}\ln\frac{1}{|\delta\theta_0|} \sim \frac{1}{P-P_{\rm thr}}\ln\frac{1}{|\delta\theta_0|}.$$

This time is rather large, both owing to the logarithmic factor (which, moreover, fluctuates from experiment to experiment) and owing to the retardation of the processes near the threshold. Under typical experimental conditions it varies from seconds to tens of minutes.

The experimental study of the growth and decay of small perturbations in Refs. 37 and 38 showed the following: 1) an exponential character of the dependence of the number of rings on the time is obeyed in the initial period to good accuracy; 2) a near-threshold retardation of the rate of relaxation and growth of perturbations occurs with the relationship $|\Gamma| \approx \text{const}/|P - P_{\rm thr}|$; 3) the absolute value of the relaxation constants $|\Gamma|$ for LIFT, as previously for GON, agrees reasonably with the data on the viscosity constants γ and elasticity constants K_i obtained from nonoptical experiments.

We note an interesting experimental method.[6] In it LIFT in a cell containing the NLC 5CB 250 μm-thick was excited with a broad beam from an argon laser ($\lambda = 0.51$ μm). The induced perturbations were measured from the phase differences of the ordinary and extraordinary waves for a weak, very narrow beam of a helium-neon laser ($\lambda = 0.63\,\mu$m). In the adiabatic approximation the o-wave should not sense the distortion of the director at all.

5. STIMULATED SCATTERING (SS) AND GRATING ORIENTATIONAL NONLINEARITY (GRATING ON)

Stimulated light scattering is one of the most beautiful phenomena of nonlinear optics. It consists in the fact that a beam of coherent laser radiation of sufficiently high power begins to be intensely scattered by the elementary perturbations of the medium. Here it yields a resultant beam of shifted frequency, while the amplitude of the elementary perturbations increases in self-consistent fashion under the action of the light fields.

Let us explain the essence of the wave processes that occur here. Let two waves propagate through the medium:

$$E = E_1\exp(i\mathbf{k}_1\mathbf{r} - i\omega_1 t) + E_2\exp(i\mathbf{k}_2\mathbf{r} - i\omega_2 t). \tag{5.1}$$

Their interference acts on the medium, including its dielectric permittivity ε, so that in the first nonvanishing approximation we can write

$$\delta\varepsilon(\mathbf{r}, t) = (A' + iA'')E_1^*E_2 \times \exp[-i(\mathbf{k}_1 - \mathbf{k}_2)\mathbf{r} + i(\omega_1 - \omega_2)t] + \text{c.c.} \tag{5.2}$$

The concrete mechanisms can be most varied; thus, we shall study below the variation of the tensor $\hat\varepsilon$ owing to reorientation of the director by light fields. The complex coefficient $A' + iA''$ describes the possible phase shift of the response of the medium as compared with the phase of the original interference pattern. The scattering of the wave $E_1\exp(i\mathbf{k}_1\cdot\mathbf{r} - i\omega_1 t)$ by the running space-time grating $\delta\varepsilon(\mathbf{r}, t)$ yields by (5.2) additional terms in the displacement[2)]

$$\delta D = (A' + iA'')|E_1|^2E_2\exp(i\mathbf{k}_2\mathbf{r} - i\omega_2 t) + (A' + iA'')E_1E_1E_2^*\exp[i(2\mathbf{k}_1 - \mathbf{k}_2\mathbf{r}i(2\omega_1 - \omega_2)t]. \tag{5.3}$$

Usually the second term in (5.3) stongly fails to satisfy the wave equation. Therefore it practically does not excite propagating waves. On the contrary, the first term gives rise to radiation of additional waves in the direction $\mathbf{k}_2$ at the frequency ω_2. One can say that the Bragg condition for scattering by the corresponding running grating $\delta\varepsilon(\mathbf{r}, t)$ is automatically satisfied. In order to understand whether the newly radiated waves $\exp(-i\omega_2 t + i\mathbf{k}_2\cdot\mathbf{r})$ attenuate, amplify, or only shift the phase of the wave E_2 already existing in the medium, it is convenient to interpret this first term in (5.3) as a change in the effective dielectric permittivity of the medium for the wave E_{2i}. Simultaneously the wave vector $\mathbf{k}_2$ also varies:

$$\delta\varepsilon(\omega_2) = (A' + iA'')|E_1|^2, \quad \delta k_2 = \frac{\omega_2}{2c\varepsilon^{1/2}}(A' + iA'')|E_1|^2. \tag{5.4}$$

When $A'' = 0$, $A' \neq 0$, the scattering $(\omega_1\mathbf{k}_1 \to \omega_2\mathbf{k}_2)$ by the grating alters the phase of the wave E_2:

$$E_2(z) = \exp(i\delta k_2 z), \quad \delta k_2 = \frac{\omega A'}{2c\varepsilon^{1/2}}|E_1|^2. \tag{5.5}$$

When $A'' \neq 0$, it also alters its intensity according to the law

$$|E_2(z)|^2 \sim \exp(gz), \quad g = -\frac{\omega_2}{c\varepsilon^{1/2}}A''|E_1|^2. \tag{5.6}$$

Exponential amplification of the wave E_2 with the coefficient g, which is proportional to the "pumping" intensity $|E_1|^2$, as is realized when $A'' < 0$, characterizes the process of stimulated light scattering (SS). One can call the change in the phase of the wave E_2, which is proportional to the intensity $|E_1|^2$, cross-focusing by analogy with self-focusing, while the nonlinearity itself is called grating orientational nonlinearity (also grating ON). Thus grating ON and SS are manifestations of the same physical process of excitation of running gratings in the medium; see, e.g., Ref. 39.

5.1. Theory of orientational SS in nematic LCs

After these general remarks, let us proceed to discuss the concrete features of orientational SS and grating ON in nematic liquid crystals (NLCs). To do this, we must substitute into the usual dynamic equations of NLCs the stimulating force from the interference of the two waves, find the response, and calculate the result of scattering by the running grating of perturbation of the director. This procedure, while extremely simple in principle, leads to rather unwieldy expressions. While referring to the original study[40] for the details, we shall write the equations for the concrete case for which SS and grating ON were found and studied experimentally.

Let a wave propagate through a homeotropic cell at the small angle α to the director. In the general case it contains both polarizations and is nonmonochromatic. Upon choosing the z axis along the unperturbed director, we shall write the field in the form

$$E_{\text{real}}(x, z, t) = \frac{1}{2}[\mathbf{e}_y E_y + \mathbf{e}_x E_x(z)\exp(i\Omega t + i\mu z)] \times \exp[ik(x\alpha + z\cos\alpha) - i\omega t] + \text{c.c.} \quad (5.7)$$

Here $k = \omega n_\perp / c$ is the wave number of the o-wave $\mathbf{e}_y E_y$, which is the "pump" in our case. The signal is the e-wave $\mathbf{e}_x E_x$ with the shifted frequency Ω. The quantity $\mu = (\omega n_\perp / 2c)[1 - (n_\perp^2/n_\parallel^2)]\alpha^2$ is the difference of the z components of the wave vectors of the o- and e-waves. The linearized equation for the perturbation of the director $\mathbf{n} = \mathbf{e}_z + \mathbf{e}_x\theta_x + \mathbf{e}_y\theta_y$ has the form

$$\gamma\frac{\partial\theta_y}{\partial t} - K_3\frac{\partial^2\theta_y}{\partial z^2} = -\frac{\alpha\varepsilon_a}{16\pi}[E_x E_y^* \exp(i\Omega t + i\mu z) + \text{c.c.}]. \quad (5.8)$$

The right-hand side in (5.8) is written in the first nonvanishing approximation in ε_a.

If the rate of variation of the slow amplitude $E_x(z)$ is small in comparison with the quantity μ, then the stationary solution of Eq. (5.2) neglecting boundary effects has the form

$$\theta_y(z, t) = -\frac{\alpha\varepsilon_a E_0^* E_x(z)}{16\pi K_3 \mu^2 (1 + i\Omega/\Gamma)}\exp(i\Omega t + i\mu z) + \text{c.c.} \quad (5.9)$$

Here $\Gamma = K_3 q^2/\gamma$ is the relaxation constant (s^{-1}). The complex conjugate term in (5.9) does not yield accumulating effects owing to lack of fulfillment of synchronism conditions. If we omit it, we can derive truncated equation for the amplitude of the signal:

$$\frac{dE_x}{dz} = -i\mu\alpha^{-1}\theta_y(z, t)E_0\exp(-i\Omega t + i\mu z) = \frac{i\varepsilon_a |E_0|^2}{16\pi K_3\mu^2(1 + i\Omega|\Gamma)}E_x. \quad (5.10)$$

Equation (5.10) implies that the greatest amplification is reached for the Stokes signal with $\Omega = \Gamma$, so that $|E_x|^2 \propto \exp(gz)$:

$$g = \frac{\lambda n_\parallel^2 |E_0|^2}{16\pi K_3 n_\perp \alpha^2}, \quad \Omega_{\text{opt}} = \frac{\pi^2 n_\perp^2 [1 - (n_\perp^2/n_\parallel^2)]\alpha^4 K_3}{\lambda^2\gamma}. \quad (5.11)$$

The time for establishment of the maximum intensity of SS amounts to $\tau \approx gz/2\Gamma$ (see Refs. 41–43). Under the experimental conditions we have $gz \approx 1$–2, so that $\tau \approx \Gamma^{-1}$.

The use of a small angle between the director and the direction of propagation leads to two effects. First, according to the right-hand side of Eq. (5.8) the action of the light on the director when $\theta \approx 0$ is attenuated by the small factor $\alpha \ll 1$. The action of the perturbation θ on the interaction of the waves is also attenuated by the factor α (right-hand side of Eq. (5.10)). All this yields the small numerator $\sim\alpha^2$ in the expression for the nonlinearity. At the same time the denominator, which corresponds to the Frank energy $K_3\mu^2$, is proportional to α^4, since $\mu \sim \alpha$ for a small angle of refraction. Hence the amplification coefficient g of SS (along with the grating ON constant) depends on the angle of refraction α as $g \propto \alpha^{-2}$; see (5.11).

5.2. Observation of SS in nematic LCs

Stimulated scattering in NLCs was found in Ref. 44. The o-wave was incident at a small angle on a homeotropically oriented layer of the NLC 5CB 110-μm thick. An argon laser ($\lambda = 0.49\,\mu$m) was used. A certain fraction of the radiation was spontaneously scattered into the e-wave with a frequency shift close to optimal. This fraction was amplified exponentially in the volume of the NLC, and its intensity was measured at the output of the cell. Figure 10 shows the dependence of the power of the scattered e-wave W_S on the power of the incident o-wave W_L.

The frequency shift was measured as follows. A polarizer was placed at the output of the cell so that the o- and e-waves in the transmitted radiation interfered. Then the time course of the interference beats could be directly recorded in an oscillogram.

The functional dependences and absolute values of the measured quantities ($g, \Omega_{\text{opt}}, W_S/W_L$) on the pump intensity and the angle of refraction agreed well with the theory; see the original paper for the details.

The observed results included one not fully understood at present. After 10–15 s of the normal SS pattern, the intensity of the e-wave fell almost to zero. This can involve a homogeneous reorientation of the director by the e-wave based on GON, whereby the angle between the optic axis and the direction of propagation becomes unfavorable for SS.

A study[73] has been performed very recently in which

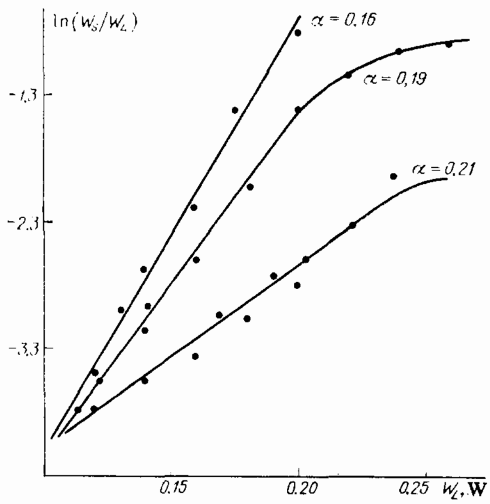

FIG. 10. Dependence of the power of the scattered e-wave on the power of the incident o-wave.

forward SS was experimentally determined with normal incidence of the o-wave on a planar cell containing the nematic 5CB. The exciting radiation at the wavelength $\lambda = 0.69\ \mu$m was generated with a ruby laser; its duration of 8×10^{-4} s was appreciably shorter than the relaxation time $\tau = \gamma/Kq^2 \approx 2.5\times10^{-3}$ s of orientation gratings having the period $\Lambda = 2\pi/q = \lambda/(n_{\parallel} - n_{\perp})$. For this reason the nonstationary theory of SS was used to describe SS from the o-wave into the e-wave (see, e.g., Ref. 74). The experimental results agree well with the theory, both in functional dependences and in absolute values for the increments.

5.3. Observation of static gratings and four-wave mixing

Insofar as we know, grating orientational nonlinearity (grating ON) in the mesophase of an NLC has not been observed. However, a related nonlinear-optical process has been studied, involving the recording and readout of static gratings $\delta\mathbf{n}(z)$ in NLCs, namely, four-wave mixing.[45] The radiation of an argon laser was directed in this study into a homeotropic cell 100-μm thick containing the nematic 5CB. It had a polarization such that an o-wave and a signal e-wave of lower power (by a factor of about 9) were simultaneously excited. The interference of these waves produced a volume static grating. The transmitted o-wave was isolated with a polarizer and reflected exactly backward with a plane mirror. Owing to scattering of the forward-propagating o-wave by the grating of the director, an e-wave was excited that also propagated in the opposite direction to the original laser beam. Both the absolute value and the functional dependences of the intensity of the resulting e-wave and time of establishment on the angle of incidence and the power of the incident radiation agreed well with the theory. A coefficient of pumping from the o-wave into the e-wave of $\sim 70\%$ (in intensity) was attained at relatively low levels (~ 500 W/cm^2) of power density of the reference beams.

5.4. LIFT in the field of the ordinary wave

With strictly normal incidence of a broad beam on a homeotropic cell, the absolute direction of polarization is inessential, since the problem is symmetric about the z axis. With strongly inclined incidence, $\mathbf{k} = k(\mathbf{e}_z \cos\alpha + \mathbf{e}_x \sin\alpha)$, $\alpha \sim 1$, one must study the behavior of the system under two possible types of perturbations of the director.[30] If the perturbations are smooth (which is favorable from the standpoint of the deformation energy), then the incident wave is split into o- and e-waves, with the polarization of each of them tracking the local direction of the director. Then the e-wave gives rise to the usual giant reorientation, while for the o-wave the free energy does not depend on the orientation of the director, and hence it has no effect on the LC. If the perturbations have a small spatial scale, $\sim\lambda/(n_e - n_o)$ or smaller, then a nonadiabatic transfer of energy from the o-wave to the e-wave can occur in the process of propagation of the light. Under these conditions a lowering of the light component of the free energy can occur also for an incident wave of the o-type. However, the deformation energy sharply rises for small-scale perturbations.

For these reasons LIFT is practically impossible for strongly inclined incidence ($\alpha \sim 1$) of waves of o-type under ordinary conditions in which $\omega(n_{\parallel} - n_{\perp})L/c \sim 10^2$. In fact the compromise between the deformation energy and the energy of interaction with light would be attained at such a large power that the system would prove to be considerably above the threshold of other nonlinear processes, e.g., stimulated scattering.

LIFT in the field of a weakly inclined ($\alpha \ll 1$) o-type wave is of great interest. While referring for the details to Ref. 46, in which this problem was studied both theoretically and experimentally, we note only the following. The theoretical dependence of the threshold on the angle of refraction α has the form

$$P_{\text{thr}}(\alpha) = P_{\text{thr}}(\alpha = 0)\left[1 + \alpha^4\left(\frac{L}{\lambda}\,\frac{\varepsilon_a\varepsilon_{\perp}^{3/2}}{\varepsilon_{\parallel}}\right)^2 + \dots\right]. \tag{5.12}$$

In Fig. 11, which is taken from Ref. 46, the dots show the experimentally measured dependence of the LIFT threshold on α^4. The solid line is the straight line drawn through these points. The dotted line corresponds to the theoretical dependence with the parameter $(L\varepsilon_a\varepsilon_{\perp}^{3/2}/\lambda\varepsilon_{\parallel})$ obtained from observation of the conoscopic pattern.

As V. F. Kitaeva has kindly reported to us, she and her associates have recently studied in detail strictly periodic and chaotic oscillations upon exciting LIFT with an ordinary weakly-inclined wave.

6. ACTION OF LIGHT ON THE SURFACE LAYER OF A LIQUID CRYSTAL

This Section 6 will discuss a series of problems in which a light wave acting on an LC is localized in a relatively thin layer Δz at the surface. At first glance the corresponding effects are weaker than those of bulk type by a factor of $L/\Delta z$, where L is the thickness of the cell. However, in concrete problems a number of favorable factors exists that allow compensation of the weakening effect of the small parameter $\Delta z/L$. Importantly, when one studies surface actions, one

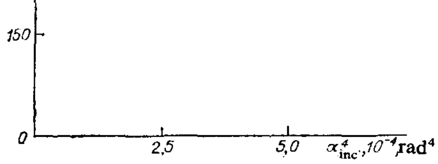

FIG. 11. The dots indicate the experimentally determined dependence of the LIFT threshold on the fourth power of the angle of refraction α^4. The solid line is the straight line drawn through these points. The dotted line corresponds to the theoretical dependence $P_{\text{thr}} = P_0[1 + (\mu L/\pi)^2]$ with μ determined from experiment.

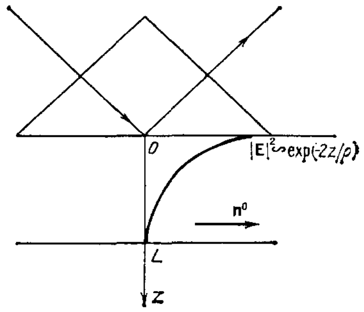

FIG. 12. The plate of the NLC cell in the plane $z = 0$ is a prism at whose boundary with the NLC total internal reflection of light occurs.

can study an LC in the presence of strong bulk scattering and in a region of light frequencies at which strong absorption exists.

6.1. GON and LIFT in the field of a surface light wave

Let us study the orienting action on an NLC of a nonpropagating light wave localized near the boundary $z = 0$. Such a wave can arise when light is incident on an NLC from a medium having a large refractive index n_i owing to the effect of total internal reflection (TIR) (Fig. 12). Let the cell have a planar orientation ($\mathbf{n}^0 = \mathbf{e}_x$) caused by rubbing of the surface $z = L$, while the surface $z = 0$ does not affect the orientation. We shall take the wave vector of the incident wave in the form

$$\mathbf{k} = \frac{\omega}{c} n_i (\mathbf{e}_x \sin\alpha\cos\beta + \mathbf{e}_y \sin\alpha\sin\beta + \mathbf{e}_z\cos\alpha).$$

For concreteness, let us examine the case in which the wave is polarized perpendicular to the plane of incidence, $\mathbf{e}_{\rm inc} \propto \mathbf{k}\times(\mathbf{k}\times\mathbf{e}_z)$. In this case the field in the NLC has only the components E_x and E_y and the equation for the profile of the director $\mathbf{n} = \mathbf{e}_x \cos\theta(x) + \mathbf{e}_y \sin\theta(z)$ has the form

$$-\gamma\frac{\partial\theta}{\partial t} + K_2\frac{\partial^2\theta}{\partial z^2} + \frac{\varepsilon_a}{16\pi} \times [\sin 2\theta(|E_y|^2 - |E_x|^2) + \cos 2\theta(E_x E_y^* + E_x^* E_y)] = 0. \quad (6.1)$$

When the anisotropy is small, $\varepsilon_a \ll \varepsilon_\perp$, the field inside the NLC can be taken as the same as for an isotropic medium. Then, employing the usual relations of the theory of TIR, we obtain

$$\mathbf{E}(z) = E_{\rm inc}\mathbf{e}_{\rm inc}\frac{2\exp(-z/2\rho)}{1 + i[1 - (\sin\alpha_{\rm TIR}/\sin\alpha)^2]^{1/2}}. \quad (6.2)$$

Here $\alpha_{\rm TIR}$ is the angle of total internal reflection, $\sin^2\alpha_{\rm TIR} = n^2/n_i^2$, and

$$\rho = \frac{\lambda_{\rm vac}}{4\pi n_i \sin\alpha}\left[1 - \left(\frac{\sin\alpha_{\rm TIR}}{\sin\alpha}\right)^2\right]^{-1/2}. \quad (6.3)$$

LIFT arises when the unperturbed direction of the director lies in the plane of incidence of the wave, i.e., $\beta = 0$, $\mathbf{e}_{\rm inc} = \mathbf{e}_y$. Upon linearizing Eq. (6.1), we obtain

$$-\frac{\gamma}{K_2}\frac{\partial\theta}{\partial t} + \frac{\partial^2\theta}{\partial z^2} + b^2\theta\exp\left(-\frac{z}{\rho}\right) = 0. \quad (6.4)$$

Here we have $b^2 = \varepsilon_a|E|^2_{\rm inc}/2\pi K_2$. (If we do not assume ε_a to be small, then the reorientation of the director leads to modification of the conditions of refraction and will excite a field component $E_x \neq 0$. It turns out that an accurate treatment of these effects[47] does not alter the qualitative conclusions and gives rise only to a certain redefinition of the quantities b^2 and ρ.)

One must solve the problem of finding the threshold of instability from Eq. (6.4) with the boundary conditions $\theta(z = L) = 0, (d\theta/dz)_{z=0} = 0$. The latter is the condition for free orientation of the director at $z = 0$. Upon employing the small parameter $\rho/L \sim 10^{-2}$ for $\rho \sim 1\ \mu$m, $L \sim 100\ \mu$m, we can describe the action of the term $b^2\theta\exp(-z/\rho)$ by introducing the boundary condition

$$\left(\frac{d\theta}{dz} + C\theta\right)_{z=0} = 0, \qquad C = \int_0^\infty b^2\exp\left(-\frac{z}{\rho}\right)dz = b^2\rho. \quad (6.5)$$

Here we can treat the equation itself without the term $\propto b^2$. The instability threshold is determined by the appearance of a nontrivial solution having a zero time increment. Then we have $\theta(z) = {\rm const}\cdot(z - L)$ and $CL = 1$ at the threshold, whence we obtain the following expression for the threshold power density in the incident wave:

$$P_{\rm thr} = \frac{cn_i|E_{\rm inc}|^2}{8\pi} = \frac{cn_iK_2}{4\pi\varepsilon_a L\rho} = \frac{c\varepsilon_i(\sin^2\alpha - \sin^2\alpha_{\rm TIR})^{1/2}}{\varepsilon_a L\lambda_{\rm vac}}. \quad (6.6)$$

Comparison of Eq. (6.6) with the threshold power density for bulk LIFT in a homeotropic cell of the same thickness (Eq. (4.4)) yields

$$\frac{P(\text{Surf. LIFT})}{P(\text{LIFT})} = \frac{\varepsilon_i}{\varepsilon_\perp^{1/2}}\frac{K_2}{K_3}\frac{L(\sin^2\alpha - \sin^2\alpha_{\rm TIR})^{1/2}}{\pi^2\lambda_{\rm vac}}. \quad (6.7)$$

Thus (6.7) contains the large dimensional factor $L/\Delta z = L(\sin^2\alpha - \sin^2\alpha_{\rm TIR})^{1/2}/\lambda$, which involves the fact that the light action occurs in the small fraction $\Delta z/L$ of the cell. However, the ratio K_2/K_3 proves to be a favorable small factor, which usually amounts to about 0.3, and the numerical coefficient π^{-2} is ≈ 0.1. All this shows that the LIFT threshold in the field of a surface light wave can prove to be of the order of the threshold of ordinary LIFT for a homeotropic cell of the same thickness.

To determine the superthreshold structure in the same approximation with $\rho \ll L$ and $\varepsilon_a \ll \varepsilon_\perp$, one can use a boundary condition of the form $[d\theta/dz + 0.5\sin 2\theta]_{z=0} = 0$. Hence we have $\theta(z) = \theta_1(1 - z/L)$, and the quantity θ_1 is determined by solving the equation

$$\frac{\sin 2\theta_1}{2\theta_1} = \frac{P_{\rm thr}}{P}. \quad (6.8)$$

If the angle $\beta \neq 0$, then analogously to GON, a reorientation of the director arises even in the first order in the light intensity. Upon taking $|E_x| = |E|\cos\beta$ and $|E_y| = |E|\sin\beta$, where $|E|$ is defined in (6.2) and solving Eq. (6.1) in the first order in θ, we obtain

$$\theta(z) = \left(1 - \frac{L}{z}\right)\frac{P}{P_{\rm thr}}\frac{\sin 2\beta}{2}. \quad (6.9)$$

The time of establishment for GON amounts to $\tau = 4\gamma L^2/\pi^2 K_2$, while for LIFT it is even larger, especially near the threshold.

The discussed effects must occur also in the case when we take a cholesteric instead of a nematic. All of the derived formulas are also valid for a CLC as well if we consider $\theta(z) - q_0(z-L)$ instead of $\theta(z)$. Qualitatively the same effect, but with a certain modification of the formulas, should also occur for a SLC-C cell in which the layers are parallel to the walls of the cell.

If the surface $z=0$ has the preferential orientation $\theta = 0$, then we can introduce the surface energy F_{surf} (erg/cm^2) $= 1/2\sigma_a \sin^2\theta(z=0)$. This corresponds to an attachment length $R = K_2/\sigma_a$; cf. Eq. (3.16). Then the threshold power density is increased by the factor

$$P_{\text{thr}}(R) = \frac{1+(L/R)}{1+(\rho/R)} P_{\text{thr}}(R=0), \tag{6.10}$$

which has been derived under the assumption that $R \gg \rho$. When R is decreased to the value $R \lesssim L$, the threshold begins to increase as R^{-1}. After R has become less than ρ, the threshold again ceases to depend on R and is determined by Eq. (6.6) with the substitution $L \to \rho$.

An experiment[48] has been performed according to the diagram of Fig. 12 using the radiation of an argon laser ($\lambda = 0.51\,\mu$m) and the nematic MBBA. The deviation of the director was determined from the rotation of the plane of polarization of a probe beam of a neon-helium laser transmitted through the NLC from the side of the rigidly orienting surface $z = L$. The LIFT in this geometry was recorded at a power density of the incident wave $P_{\text{thr}} \approx 1.2$ kW/cm^2 for a cell thickness of $50\,\mu$m. We point out that the value of ρ was about 25×10^{-5} cm. The angle of rotation observed by using the test beam was about 25° for $P/P_{\text{thr}} \sim 5$.

The absolute value of P_{thr} agrees reasonably well with the theoretical estimate by Eq. (6.6). The superthreshold angle θ_1 had different signs in different experiments, in accord with the general views on the development of instability in LIFT from initial random fluctuations. The magnitude of $|\theta_1|$ above the threshold strongly deviates from Eq. (6.8). This can involve the fact that the condition for TIR was not satisfied for the extraordinary wave (the refractive index of the prism $n_i = 1.76$ is close to the value $n_\parallel \approx 1.75$). The times of establishment for the intensity near the threshold amounted to ~120–150 s. A threshold-free effect was observed for $\beta \neq 0$, an analog of GON.

6.2. Interaction of surface plasmons with liquid crystals

Localized electromagnetic waves can propagate near the boundary of two media having the dielectric permittivities ε_1 and ε_m—surface plasmons; see, e.g., Refs. 49 and 50. Their existence requires that one of the media (usually a metal) has a negative ε so that $-\varepsilon_m = |\varepsilon_m| > \varepsilon_1 > 0$. Let us denote by $\mathbf{e}_z$ the normal to the boundary, and by $\mathbf{m}$ the unit vector in the direction of propagation of the surface plasmon, $\mathbf{m} = \cos\beta\mathbf{e}_x + \sin\beta\mathbf{e}_y$. Also let $\mathbf{v} = \mathbf{e}_z \times \mathbf{m} = \mathbf{e}_y \cos\beta - \mathbf{e}_x \sin\beta$. Then, if we assume that $\varepsilon = \varepsilon_1$ when $z>0$ and $\varepsilon = -|\varepsilon_m|$ when $z<0$, we can write the following expression[50] *for the electric field of the surface plasmon (SP):*

$$\mathbf{E}(\mathbf{r}) = Ae^{ik\mathbf{m}\mathbf{r}}\left\{\mathbf{e}_z\begin{bmatrix} e^{-\varkappa_1 z} \\ -\frac{\varepsilon_1}{|\varepsilon_M|}e^{\varkappa_2 z}\end{bmatrix} + \mathbf{m}\begin{bmatrix} -i\frac{\varkappa_1}{k}e^{-\varkappa_1 z} \\ -i_1\frac{\varepsilon_1}{|\varepsilon_M|}\frac{\varkappa_2}{k}e^{\varkappa_2 z}\end{bmatrix}\right\}. \tag{6.11}$$

In Eq. (6.11) the upper row pertains to the region $z>0$, and the lower row to $z<0$. The quantities $(2\varkappa_1)^{-1}$ and $(2\varkappa_2)^{-1}$ characterize the dimension of the region of localization of intensity of the SP respectively when $z>0$ and $z<0$, where we have

$$\varkappa_1 = \left[k^2 - \left(\frac{\omega}{c}\right)^2\varepsilon_1\right]^{1/2}, \quad \varkappa_2 = \left[k^2 + \left(\frac{\omega}{c}\right)^2|\varepsilon_M|\right]^{1/2}. \tag{6.12}$$

For example, when $z>0$, the square of the modulus of the field declines as $\exp(-z/\rho)$, where $\rho = (2\varkappa_1)^{-1}$. Moreover, the equation $\varkappa_1/\varepsilon_1 = \varkappa_2/|\varepsilon_m|$ is implied by the continuity of the fields at the boundary; it gives rise to the dispersion equation, or connection between ω and $\mathbf{k}$:

$$k^2 = \left(\frac{\omega}{c}\right)^2\frac{\varepsilon_1|\varepsilon_M|}{|\varepsilon_M| - \varepsilon_1}. \tag{6.13}$$

We can express the quantity $|A|^2$ in terms of the component P_m of the Poynting vector $\mathbf{P}$. If we introduce the specific energy flux,

$$W(\text{erg/cm}\cdot\text{s}) = \int_{-\infty}^{+\infty}(P_m)\,dz,$$

then we have

$$W = |A|^2\rho\frac{c\varepsilon_1}{8\pi}\left(1 - \frac{\varepsilon_1^2}{\varepsilon_M^2}\right)\left(\frac{|\varepsilon_M| - \varepsilon_1}{\varepsilon_1|\varepsilon_M|}\right)^{1/2}, \tag{6.14}$$

Here the term $-\varepsilon_1^2/\varepsilon_m^2$ corresponds to a counterflux of energy flowing in the medium having a negative dielectric permittivity.

We shall be interested in the problem in which the SP propagates along the phase boundary of a solid having $\varepsilon = \varepsilon_m < 0$ and a layer of a LC adjoining it. Strictly speaking, we must solve the problem of SPs taking into account the anisotropy of the tensor $\hat{\varepsilon}$ for the LC. For estimates of the order of magnitude of the orientational effects, we shall use the formulas given above that were derived for SPs at the boundary of isotropic media.

The action of the field of the SP on the orientation of the LC has roughly the same character as for the nonpropagating surface wave in TIR discussed in Sec. 6.1. In particular, the SP can cause a twisting deformation in the geometry of Fig. 12 in threshold fashion (when $\mathbf{m}\perp\mathbf{n}^0 = \mathbf{e}_x$) or threshold-free (when $0 < |\mathbf{m}\cdot\mathbf{n}^0| < 1$). Instead of the prism, we must assume here a medium having $\varepsilon = \varepsilon_m < 0$.

Rather often for metals we find $|\varepsilon_m| \gg 1$, even at frequencies in the optical range. Then we have $k^2 \approx (\omega^2\varepsilon_1/c^2)[1 + (\varepsilon_1/|\varepsilon_m|)]$, the dimension of localization in the intensity in the LC is small, and $\rho = \lambda_{\text{vac}}|\varepsilon_m|^{1/2}/4\pi\varepsilon_1$. Therefore $|\mathbf{E}\cdot\mathbf{e}_z|$ is larger than $|\mathbf{E}\cdot\mathbf{m}|$ by a factor of about $(|\varepsilon_m|/\varepsilon_1)^{1/2}$. Under these conditions the interaction of an SP with *an NLC in the geometry of Fig. 12 can be of interest, but with* account taken of the possibility of S-deformation. If the director at the surface $z = 0$ is not attached at all, then one can consider perturbations of the form $\mathbf{n}(z, t) = \mathbf{e}_x \cos\theta(z, t) + \mathbf{e}_y \sin\theta(z, t)$. Let the SP propagate in the direction $\mathbf{k} = \mathbf{e}_y k$ perpendicular to the director. Then the linearized equation for $\theta(z, t)$ acquires the form

$$-\frac{\gamma}{K_3}\frac{\partial\theta}{\partial t} + \frac{\partial^2\theta}{\partial z^2} + b^2\theta \exp\left(-\frac{z}{\rho}\right) = 0. \qquad (6.15)$$

Here we have $b^2 = \varepsilon_a |A|^2/2\pi K_3$. Treating this equation in the same way as in Sec. 6.1, we find that the threshold condition on the reorientation of the layer by the field of the SP has the form $b^2\rho K = 1$, whence we obtain the following expression for the specific threshold power W_{thr} (erg/cm·s) of the SP:

$$W_{\text{thr}} = \frac{\omega\varepsilon_1 K_3}{4K\varepsilon_a L}\left(1 - \frac{\varepsilon_1^2}{\varepsilon_M^2}\right) \approx \frac{K_3 c\varepsilon_1^{1/2}}{4L}. \qquad (6.16)$$

Applicability of the one-dimensional approximation used above requires that the dimension of the SP packet along the y coordinate must not be smaller than L. Thus we obtain the condition for the total power $W_{\text{thr}}L$ (erg/s) transported by the SP. Upon assuming that $\varepsilon_1/|\varepsilon_m| \ll 1$, $k \approx \omega\sqrt{\varepsilon_1}/c$, $\varepsilon_a \sim 1$, $K_3 \sim 10^{-6}$ dyne, we obtain the estimate $W_{\text{thr}}L \sim 10^{-3}$ W. This value proves independent of the thickness L. When $L \sim 10^{-2}$ cm the specific threshold flux amounts to $W_{\text{thr}} \sim 10^{-1}$ W/cm·s.

Above in Secs. 6.1 and 6.2 we have studied the influence of surface waves on the orientation of NLCs. In turn, the altered orientation of the NLC exerts a reverse influence on the amplitude and phase of the same light waves—the reflected wave in TIR or the propagating surface plasmon. For example, effects can occur for SPs such as self-focusing owing to GON and LIFT, grating nonlinearity in the interaction of two SPs, and stimulated scattering of one SP in the field of another. Effects of rescattering from an SP bound at a boundary into a bulk wave and vice versa owing to nonlinear orientational interaction are highly interesting. This field is only beginning to develop at present.

6.3. Variation of pitch of the cholesteric helix

The strongest influence of a light field on the pitch of a cholesteric helix involves the trivial effect of heating of the CLC by the light field. With a heat-conductivity coefficient $\sim 10^{-3}$ cm^2/s and a cell thickness $L = 50\,\mu$m, the time for establishment of a stationary temperature profile amounts to $\tau = (L/\pi)^2/\chi = 2.5\times 10^{-3}$ s. For a moderately pure LC, one can adopt the estimate of the coefficient of true absorption $\alpha \sim 1$ cm^{-1}. Then, with a power density of the incident light of $\sim 10^3$ W/cm^2, we obtain the energy release in the time τ of the order of 2.5 J/cm^3. This corresponds to a temperature elevation of about 2 °C. We shall defer a detailed discussion of the thermal effects to Sec. 7.

In this section we shall completely neglect thermal effects and discuss how a light field can alter the pitch of a CLC owing to direct dynamic action on the director. It turns out that the greatest effect is attained under conditions of Bragg scattering $(\omega n/c) \approx q_0$ of a normally incident light wave from a planar Grandjean structure. For CLCs the magnitude of ε_a is usually small, $\varepsilon_a \sim 0.03$–0.3. However, this smallness is compensated by the fact that the wave being reflected penetrates into the CLC to a depth $\Delta z \sim \lambda/\varepsilon_a$. Consequently, for thick enough specimens (totally reflecting), the moment of the force exerted by the light wave on the helix does not depend on ε_a.

In the stationary case the Maxwell and Euler-Lagrange equations imply the relationship

$$\frac{dM}{dz} = 0,$$

$$M(z) \equiv -K_2\left(\frac{d\theta}{dz} - q_0\right) + \frac{c^2}{16\pi\omega^2}\left[\left(iE_+\frac{\partial E_+^*}{\partial z} - iE_-\frac{\partial E_-^*}{\partial z}\right) + \text{c.c.}\right]. \qquad (6.17)$$

Here the director is defined by the relationship $\mathbf{n} = \mathbf{e}_x \cos\theta + \mathbf{e}_y \sin\theta$; E_+ and E_- are the circularly polarized components of the electric field of the light wave

$$\mathbf{E}(z) = \frac{\mathbf{e}_x + i\mathbf{e}_y}{\sqrt{2}}E_+(z) + \frac{\mathbf{e}_x - i\mathbf{e}_y}{\sqrt{2}}E_-(z).$$

It expresses the law of conservation of angular momentum of the system CLC + field. Although one can easily verify (6.17) indirectly, we have derived the original expression by using Noether's theorem on the connection between the law of conservation of angular momentum and the invariance of the Lagrangian with respect to the rotation group about the z axis.

While referring to the original study[51] for further details, we shall formulate its fundamental results. The wave E_-e^{ikz} is totally reflected from a "right-handed" CLC ($q_0 > 0$) when the Bragg condition is satisfied, while transferring its energy to the wave E_+e^{-ikz}. The moment of the "recoil" forces arising here is transferred to the helix so as to alter its pitch. If the director is free on the side on which the light is incident and reflected, and fixed at the opposite boundary of the cell, then the pitch of the helix is shortened by the action of the field *throughout the volume* (and not only in the layer where light is present). At equilibrium we have

$$\frac{d\theta}{dz} = q_0 + \frac{2}{\omega K_2}P_0.$$

Here P_0 is the power density of the reflected light (erg/cm^2 s). Yet if the director is fixed specifically to the input boundary and free at the opposite boundary, then the pitch of the helix increases by about the same amount, but only in the region where the reflection process takes place.

We can liken the situation of the change in the pitch of a CLC to the problem of the change of the pitch of a steel spring to which we apply a twisting moment to a certain small region $0 < z < \Delta z$. If the spring is free at $z = 0$ and fixed at $z = L$, then its pitch is altered practically throughout the length. Yet if the spring is fixed at $z = 0$ and free at $z = L$, then the pitch is altered only in the layer $0 < z < \Delta z$, and in the sense opposite to the former case. That is, the pitch remains unchanged through most of the cell, while the change in the angle $\delta\theta(z = L)$ amounts to about

$$\delta\theta \sim -q_0 L\frac{2P_0 n}{cK_2 q^2}\frac{\rho}{L}. \qquad (6.18)$$

None of our conclusions have rested on any concrete form of the dependences $\theta(z)$ and $P^{(z)}(z)$, and have employed only the fact that the waves can be distinguished both in direction of propagation and in circular polarization when $\varepsilon_a \ll \varepsilon$.

The problem that we have treated shows how useful it is to employ a unitary Lagrangian for obtaining the total system of equations and finding the conservation laws. Owing to such a solution of this problem,[51] the qualitatively new conclusion presented above was obtained of the *nonlocal* dependence of the helical pitch on the intensity of the light waves; see also Ref. 75 (previously this problem had been treated incorrectly in Ref. 52 and in our own Ref. 21).

7. NONLINEARITIES INVOLVING ABSORPTION

Above we have treated the orientational nonlinearity of transparent LCs in which the light quanta are not absorbed. The natural question remains of the source of the energy that goes into reorienting the director. The answer is that on the average the light quanta are slightly red-shifted in the nonlinear processes. For example, the light transmitted through a GON cell when the intensity is abruptly turned on has the time-dependence

$$E_{\text{trans}}(t) = E_{\text{inc}}(t) \exp[i\phi_0 + i\delta\phi_{\text{nonlin}} f(t) - i\omega_0 t]. \tag{7.1}$$

Here we have $f(t) = 1 - \exp(-\Gamma t)$. Since $\delta\phi_{\text{nonlin}} > 0$, then Eq. (7.1) implies that the instantaneous frequency $\omega_{\text{inst}} = -\partial\phi/\partial t$ is shifted by the amount

$$\omega_{\text{inst}} - \omega_0 = -\Gamma e^{-\Gamma t}\delta\phi_{\text{nonlin}} < 0. \tag{7.2}$$

Precisely this quantum defect $\hbar(\omega_0 - \omega_{\text{inst}})$ is spent on establishing the equilibrium orientation. Under ordinary conditions this is an extremely small quantity: when $\delta\phi_{\text{nonlin}} \sim 20 - 2\pi \sim 120$ and $\Gamma \sim 10^{-1}\ \text{s}^{-1}$, the relative frequency shift $|\delta\omega|/\omega$ amounts to less than 10^{-14}. If we multiply the typical power density $10^3\ \text{W/cm}^2 = 10^{10}\ \text{erg/cm}^2\ \text{s}$ by the time $\tau \sim 10$ s and by the factor 10^{-14}, we obtain an energy expenditure of about $10^{-3}\ \text{erg/cm}^2$. And actually, the Frank energy for 50% reorientation of the director in a cell of thickness $L = 100\ \mu\text{m}$ is $FL = LK(\pi/L)^2 \approx 10^{-3}\ \text{erg/cm}^3$.

These estimates show that a quite insignificant absorption, $1 - \exp(-\sigma L) \sim 10^{-13}$ would suffice to make the light beam release a tenfold larger energy in the medium. This implies that if we can find a suitable mechanism transforming the absorbed energy into a change in the refractive index, then nonlinearities caused by absorption can prove to be far stronger than all the "dynamic" nonlinearities treated in the previous sections of our review.

The simplest group of effects involves the heating of the medium upon absorption of light; we shall start with them.

7.1. Thermal self-focusing and self-defocusing

A distinctive feature of the mesophase of an LC is the very strong dependence of the refractive index on the temperature. While in an isotropic phase we have $\partial n/\partial T \approx (\partial n/\partial\rho)(\partial\rho/\partial T) \approx -(10^{-4}\text{–}10^{-5})\ \text{deg}^{-1}$, for the mesophase of MBBA at $T = 33\ ^\circ\text{C}$ (i.e., $T - T_c \approx 10\ ^\circ\text{C}$) we have $\partial n_\parallel/\partial T \approx -4\times 10^{-3}\ \text{deg}^{-1}$. The quantity $\partial n_\perp/\partial T$ is positive but more than five times smaller in modulus. Consequently, in propagating through a weakly absorbing NLC, the extraordinary wave will undergo thermal self-defocusing, while the ordinary wave will undergo self-focusing. In order of magnitude one can write

$$\delta n = \sigma \frac{cn|E|^2}{8\pi\rho C_p} \frac{\partial n_i}{\partial T} \Gamma^{-1}(1 - e^{-\Gamma t}). \tag{7.3}$$

Here σ is the absorption coefficient (cm^{-1}), ρC_p (erg/cm^3 deg) is the heat capacity per unit volume, Γ^{-1} is the time for establishment of a stationary temperature profile, with $\Gamma \sim \chi[a^{-2} + (\pi^2/L^2)]$; $\chi \sim 10^{-3}\ \text{cm}^2/\text{s}$ is the heat-conductivity coefficient, a is the transverse dimension of the beam, and L is the thickness of the cell with well-cooled walls. Effects of this type were observed experimentally[53] as early as 1974. Both the power and the duration of the radiation of the argon laser were sufficient for the detection of GON. To do this, it was necessary to incline the cell with respect to the beam, which unfortunately was not done. From our viewpoint, this example convincingly shows how nontrivial it was to acknowledge the fact that the light field can reorient the director in a quite appreciable fashion.

For the extraordinary wave, the thermal effects lead to defocusing, and GON to self-focusing. In principle, one can compensate these nonlinear effects by choosing the angle of incidence.

7.2. Stimulated temperature scattering

If two waves of somewhat differing frequencies are propagating in a weakly absorbing medium, the interference term in the temperature perturbation is phase-shifted with respect to the intensity pattern. Consequently a stimulated temperature scattering of the light (STS) arises (cf. Sec. 5), caused by absorption (A). For isotropic liquids it has been studied in Refs. 54–56 and many other papers. Temperature SS in the mesophase of an LC has been discussed in Refs. 40 and 57; the essential point consists in the large magnitude and the anisotropic character of the derivative $\partial\hat{\varepsilon}/\partial T$. For example, for STS of opposing extraordinary waves propagating perpendicular to the director, with $\lambda_{\text{vac}} = 0.5\ \mu\text{m}$, $\partial n_\parallel/\partial T = -4\times 10^{-3}\ \text{deg}^{-1}$, $\chi \approx 10^{-3}\ \text{cm}^2/\text{s}$, and $\rho C_p \approx 1.5\ \text{J/cm}^3\cdot\text{deg}$, we have the threshold power density for STS $P_{\text{thr}} \approx 6\times 10^5\ \text{W/cm}^2$, $\Gamma \approx 2\times 10^8\ \text{s}^{-1}$. If we make the absorption not too small, $\sigma \approx 5\ \text{cm}^{-1}$, $P \approx 20 P_{\text{thr}}$, then at the length $L = 0.1$ cm of the medium, the amplification of the wave $|E_2|^2$ amounts to $\exp(gz) \approx e^{10}$ times. In the time $\tau \sim 7\Gamma^{-1} \approx 3\times 10^{-8}$ s, the medium having these parameters is heated by the beam $|E_1|^2$ by about 1.5 °C. Thus the observation of STS-A in NLCs using pulsed lasers seems quite realistic.

7.3. Thermal reorientation

In a homogeneous specimen of a nematic, a change in temperature leads to a local change in the refractive indices $n_\parallel$ and $n_\perp$, but does not alter the homogeneous orientation of the director. If we are dealing with an inhomogeneous nematic, e.g., in a hybrid cell (see Sec. 3.4) or a twist cell, the heating of the specimen upon absorption must lead to a change in the profile of the director in the volume owing to the temperature-dependence of the Frank constant. We can expect this mechanism to manifest effects of self-focusing, defocusing, grating ON, SS, etc.

We can expect interesting phenomena in cholesterics, where the equilibrium parameter $q_0 = d\theta/dz$ depends on the temperature. For example, far from Bragg resonance a linearly polarized wave can propagate through the CLC (with a smoothly rotating direction of polarization). Owing to dichroism, the heat release in the CLC proves to be modulated with a period equal to the period of the helix. Then, owing to the temperature-dependence of q_0, the course of the angle $\theta(z)$ must be distorted within the limits of a period. In turn, this can lead to the appearance of higher Bragg orders of reflection at normal incidence.

In C smectics, in addition to the thermal effects discussed above, another effect exists: a temperature-dependence of the magnitude of the angle ζ between the **n**-director and the normal to the smectic layers. If we can assume the orientation of the layers to be fixed in the employed geometry, then heating must lead to a local rotation of the optical axis with all the consequences for nonlinear optics that stem from this. Effects of bistability based on thermal orientational nonlinearity have been discussed in Refs.75 and 76.

7.4. Nonlinearity due to photostimulated transitions

A new, substantially nonthermal mechanism of large optical nonlinearity due to absorption has been found.[58–60] A planar cell containing a mixture of cyanobiphenyls 10-μm thick or containing MBBA 65-μm thick[59,60] was illuminated with the field of two plane waves incident normal to the director and forming a small angle $\Delta\theta$ (up to several degrees) with one another. The interference of these waves inscribed a refractive-index grating with the spatial period $\Lambda = \lambda_{vac}/\Delta\theta$. The intensity of diffraction by the grating being formed of a third (probe) beam, as well as that of the inscribing waves, was recorded. The experiment had a geometry such that the orientational nonlinearity could make no appreciable contribution to this process. The authors[58–60] also convinced themselves that the thermal nonlinearities under the experimental conditions would yield a nonlinearity smaller by several orders of magnitude and faster by a factor of about 10^3 than that experimentally observed.

A model of reversible transitions of the molecules of the LC in the mesophase upon absorbing light was proposed as the mechanism in the cited studies. The greatest nonlinearity, $|\varepsilon_2| \sim 5$ cm^3/erg, was measured at the wavelength $\lambda = 0.44$ μm (helium-cadmium laser), for which the absorption coefficient of MBBA amounted to $\sigma \approx 25$ cm^{-1}. This nonlinearity declined by about ten times in going to the wavelength $\lambda = 0.48$ μm, for which $\sigma \approx 18$ cm^{-1}. If we assume that the modified state of the molecules persists in the homogeneous medium for the time T and diffuses with the coefficient D (cm^2/s), then we can write the following phenomenological equation for $\delta\varepsilon$:

$$\frac{\partial\,\delta\varepsilon}{\partial t} + \frac{1}{T}\,\delta\varepsilon - D\Delta\,\delta\varepsilon = \frac{1}{2T}\,\varepsilon_2\,|E|^2. \tag{7.4}$$

Here the constant ε_2 is proportional to the absorption coefficient. The measurements[60] of the time course of $\delta\varepsilon$ and the dependence of $\delta\varepsilon$ on the angle $\Delta\theta$ between the interfering waves enabled determining the value of the parameters from Eq. (7.4): $T \approx 1.6$ s, $D_\perp = 1.3\times 10^{-7}$ cm^2/s. The authors also convinced themselves that the strongest nonlinearity arises upon absorption of the extraordinary wave. Moreover, a result of the process is preferentially the change in the value of $\varepsilon_\parallel$, while the value of $\varepsilon_\perp$ varies more weakly by a factor of about two.

It is interesting to compare all these results with those from the earlier study,[61] where the processes of deactivation and diffusion of the dye "methylene red" mixed into the mesophase of MBBA were studied. In Ref. 61 about the same technique was applied of inscribing the amplitude-phase gratings with the radiation of an argon laser. However, owing to strong absorption in the green region of the spectrum, the beam of a helium-neon laser, $\lambda = 0.628$ μm, was used for readout of the grating. The values obtained there amounted to about several seconds for the time T, and $D_\parallel \approx 1.5\times 10^{-7}$ cm^2/s, $D_\perp \approx 2.4\times 10^{-7}$ cm^2/s. We note that also in Ref. 61 all the experimental conditions existed to enable detecting GON by choice of the inclination of the beams.

Returning to Ref. 60, let us present the estimate from it for the relative change in the polarizability β of an individual MBBA molecule upon phototransformation: $\Delta\beta/\beta \sim 10^{-1}$. The methodology of Ref. 60 did not allow determining the signs of $\delta\varepsilon_\parallel$ and $\delta\varepsilon_\perp$, i.e., ascribing the effect to self-focusing or self-defocusing. An extremely important experimental fact determined in Ref. 60 was the total absence of that type of nonlinearity in the isotropic phase of the same specimens. Moreover, at a light power density of the order of 4 W/cm^2, saturation of the nonlinear increment to the refractive index was observed. Actually, with an absorption $\sigma \sim 25$ cm^{-1} and a flux $I \sim 1.5\times 10^{19}$ quanta/cm^2 in the time $T \approx 1.5$ s, about 6×10^{20} photoexcitation events/cm^3 occur. This constitutes an appreciable fraction of the total density $N_0 \sim 3\times 10^{21}$ cm^{-3} of MBBA itself. Therefore, despite the large value of the constant ε_2, it proves impossible to obtain a considerable ($\delta\Phi \gtrsim 2\pi$) nonlinear phase shift based on the studied mechanism in cells containing MBBA of thickness $L \lesssim 60$ μm, owing to saturation.

In our opinion studying this type of nonlinearity in the mesophase of other LCs is of great interest.

8. ON THE HISTORY OF THE PROBLEM

The orientational nonlinearity of the isotropic phase of an LC near the transition point to the mesophase was studied in the early papers of G. Wong and Y. R. Shen,[62,63] Rao and Jayaraman,[64] and others; see also the review by Shen.[65] In that work it was possible to obtain experimentally a nonlinear constant of the order of 2×10^{-9} cm^3/erg. It was not possible to approach closer to the transition point and thus to elevate the constant, owing to turbidity of the specimen. A study by B. I. Lembrikov[66 3)] has treated theoretically the effect of mutual modulation of the phase in the normal propagation of the o- and e-waves in a nematic (grating ON in our terms). Herman and Serinko[67] have treated theoretically the process of reorientation of the director of an NLC by a pair of interfering light waves. Here the cell was supposed to be brought into a state near the Fréedericksz transition by using an external magnetic field. The process was studied of amplification of a weak wave by self-diffraction of a strong wave, and the low level of power required for observing effects was noted, especially near the FT threshold. B. I. Lembrikov[68] has treated theoretically stimulated scattering (SS) by second sound in a SLC-A with an amplification constant of the same order as for Mandel'stam-Brillouin stimulated scattering in ordinary liquids. B. Ya. Zel'dovich and N. V. Tabiryan[57] have calculated the cubic nonlinearity of an SLC-C and the corresponding SS processes owing to orientational, thermal, and orientational-thermal mechanisms. The estimates made indicated a high value of the nonlinearity constants. B. Ya. Zel'dovich and N. V. Tabiryan[40] have calculated the cubic susceptibility tensor of an NLC in light fields owing to reorientation of the director, have studied processes of orientational and temperature SS in NLCs, and noted anomalously high values of the amplification coefficients. B. Ya. Zel'dovich, N. F. Pilipetskiĭ, A. V. Sukhov, and N. V. Tabiryan[12] were the first to discover experimentally the giant orientational nonlinearity (GON) of the mesophase of an NLC—the light analog of the Fréedericksz effect with inclined orientation of the field with respect to the director. An experiment on self-focusing of low-power radiation, $\lesssim 10^{-2}$ W, was set up to test the theoretical predictions of the present authors, which were being developed by them independently of the studies of Lembrikov and of Herman and Serinko. In our study[12] these predictions were fully confirmed. Khoo and Zhuang[69] have carried out an experimental test of the predictions of the study of Herman and Serinko[67] by observing the amplification in the case of self-diffraction. The theoretical studies of B. Ya. Zel'dovich and N. V. Tabiryan[26,70,71] contained a detailed discussion of the processes of GON, grating ON, SS, and wave-front conjugation based on a mechanism of orientational nonlinearity of the mesophase of SLCs, NLCs, and CLCs. In an experimental test of the results of Ref. 12 on the existence of GON-self-focusing with inclined incidence of the light, A. S. Zolot'ko, V. F. Kitaeva, N. Kroo, N. N. Sobolev, and L. Chillag[17] used a homeotropic cell. Here they were able to detect experimentally a threshold reorientation of the director for a normally incident beam with power of the order of 0.1 W—a light-induced Fréedericksz transition (LIFT) in the narrow sense. S. G. Odulov, Yu. A. Reznikov, O. G. Sarbeĭ, M. S. Soskin, E. K. Frolova, and A. I. Khizhnyak[58] have reported observing a large nonthermal absorption nonlinearity of the mesophase of an NLC by the self-diffraction method. A theory of threshold LIFT, including its qualitative differences from the theory of the FT in quasistatic fields, has been presented by B. Ya. Zel'dovich, N. V. Tabiryan, and Yu. S. Chilingaryan.[32]

A large number of interesting theoretical studies has been performed in the short period since 1980 on the optical nonlinearity of liquid crystals. A bibliography approximately up to 1983 can be found in Ref. 9.

In closing these remarks on the history of the problem, we wish to note the following. The possibility of observing giant nonlinear effects in thin (~ 0.01 cm) layers of an LC at a very low power level ($\sim 10^{-2}$–10^{-2} W) was not at all obvious at the time of appearance of the first theoretical studies. As an example, we draw attention to the detailed review on nonlinear optics of LCs of the "preorientational period."[77] Apparently the impetus to a broad study of orientational nonlinearity in the socialist countries were Refs. 12 and 17, and in the Western Hemisphere, Refs. 67 and 69.

CONCLUSION

In essence the study of orientational optical nonlinearity is just beginning. In a number of cases the nonlinear effects enable one to determine parameters of the LC that are difficult to measure by other methods. A distinctive feature of the optical actions on an LC is their high spatial localization, down to dimensions $\Lambda/2\pi = \lambda_{\text{vac}}/4\pi n \approx 0.02$ μm. Moreover, the nonlinear effects in the mesophase of an LC enable one to create a model for many phenomena of interest to coherent optics using low-power lasers. The role of the possible nonlinear effects (harmful in some cases) must be taken into careful account in building LC devices designed to transmit light fluxes of even moderate power.

In our opinion a detailed experimental verification of the numerous predictions of the theory is highly desirable. We are almost convinced that many unexpected things will be discovered along this line, perhaps even more interesting that what we have been concerned with for the past several years and in which we are so delighted now.

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1) The condition for existence of hysteresis of LIFT in a broad beam and the qualitative behavior of $\theta(\rho)$ have been found in Ref. 32. Unfortunately the diagram given in this paper inaccurately conveyed the character of the correct analytic expressions derived in this same study. H. L. Ong[8] has pointed out this situation, but has employed an erroneous procedure of deriving the variational equations that in principle does not allow one to describe GON.

2) Transl. editor's note: The second term in Eq. (5.3) appears to be incorrect as reproduced here from the Russian original, but, since the authors *claim that it is in any event unimportant, no effort was made to correct* the error.

3) The studies cited below are listed in an order corresponding to the dates of submission for publication to the corresponding journals.

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