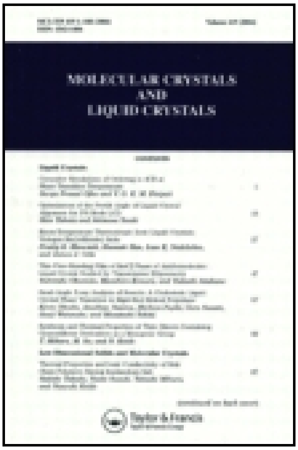

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

Orientational Optical Nonlinearity of Liquid Crystals

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Version of record first published: 20 Apr 2011.

To cite this article: N. V. Tabiryan , A. V. Sukhov & B. YA. Zel'dovich (1986): Orientational Optical Nonlinearity of Liquid Crystals, Molecular Crystals and Liquid Crystals, 136:1, 1-139

To link to this article: http://dx.doi.org/10.1080/00268948608074569

Orientational Optical Nonlinearity of Liquid Crystals

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As a result of development of optics during many centuries the concept was formed that a medium can strongly influence on the light propagation, but the light itself cannot have backward influence on the medium. That concept was refuted only with appearing of coherent and high power radiation of lasers, see, e.g., Nobel Lecture by N. Bloembergen in 1981. A new field in physics—nonlinear optics—formed as a result. The main part of laser investigations concerns with nonlinear optical phenomena, nowadays. First of all these phenomena are of extraordinary interest and beauty and provide for new methods of physical investigations. Secondly, the knowledge of laws governing the interaction of a powerful light with a medium is necessary for the correct utilization of laser beams. Finally, a great number of devices of up-to-date laser optics is totally based on utilization of nonlinear optical effects; generation of higher harmonics of light, stimulated scattering of light, optical phase conjugation, self-focusing and optical bistability are amongst. To realize the nonlinear optical effects, however, it was nearly always necessary to use powerful lasers pulses.

A new direction in liquid crystals (LC) physics—orientational optical nonlinearity of LC (mesophase)—appeared and is rapidly developing recently. Now it became obvious to everyone that the electric field of the light reorients the LC's director as well as a static magnetic or electric field do. Till the first experiment, however, the answer to a simple question was far from being obvious, namely, whether the transparency of mesophases would be enough to transmit the light of sufficient power to a length sufficient for observation of optical effects of orientational nonlinearity. The answer turned out to be positive, fortunately, and that yielded the necessity to write the present review.

The orientational nonlinearity of mesophases turned out to be $10^6 - 10^{10}$ orders of magnitude greater than the nonlinearity of "usual" media. That allowed to observe those effects using low-power c.w. lasers.

High spatial locality (up to the size 200Å) and temporal resolution (up to 10^{-12}s) of laser beams are promising for nonlinear optical effects to become a unique method for investigations of LC's physical properties. Nowadays such devices based on LC as electrooptical shutters and optically controlled light valves are largely used for the control of light beams and in optical processing of information.

1 GIANT OPTICAL NONLINEARITY (GON) OF NEMATICS: EXPERIMENT AND SIMPLE ESTIMATES

1.1 Experiment[8]

The radiation of a He-Ne laser ($\lambda = 0.628$ μm), having a power from 0 to 20 mW, was focused by a lens with focal length 25 cm on a 60-μm-thick cell with the nematic liquid crystal (NLC), in planar orientation, Figure 1. The cell was inclined with respect to the beam so that the polarization unit vector of the extraordinary wave inside the LC made an angle α with the director, i.e. with the optical axis. The angular structure and the divergence of the transmitted radiation was registered in the far zone. The angular distribution of the transmitted

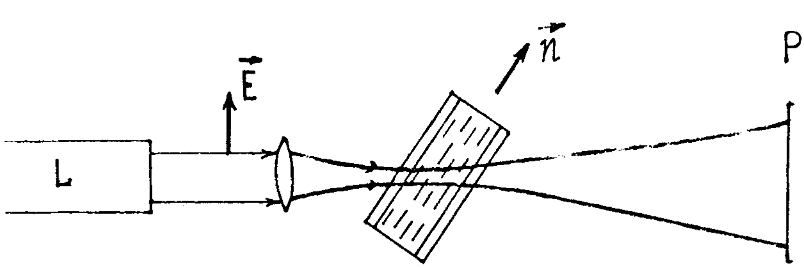


FIGURE 1 Experimental scheme for the GON observation, *L*-laser, *LC*-*NLC*-cell, *P*-screen.

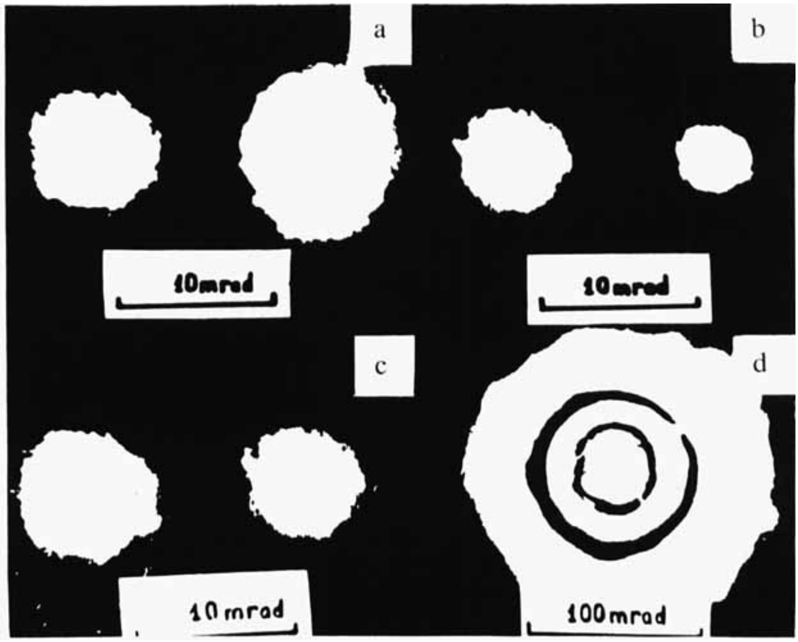


FIGURE 2 Angular distribution of incident beams and the one transmitted through the cell for different cases: a) incident beam (left) and broadened due to self-focusing one (right), $W \approx \approx 3$ mW; b) incident beam (left) and the one with smaller divergence after self-focusing in the cell, the latter placed behind the focal waist (right); c) coincidence of angular spectra of incident and transmitted beams in cases of ordinary-polarized wave and normal incidence of extraordinary wave; d) aberration fringe pattern of self-focusing in transmitted beam ($W \approx 20$ mW, ten-fold scale).

wave was practically the same as in the absence of the cell for the low power level, $W \leqslant 3$ mW, Figure 2. The angular divergence of the transmitted beam increased with increasing power. In subsequent experiments, for a higher power $W \sim 20$ mW, the angular distribution in the far zone took the peculiar form of circular fringes. The effect increased with increasing angle α (maximal value of α was 32° in the experiment). The divergence did not increase but even decreased, relative to its initial value, when the power was increased from zero to 10 mW, if the cell was moved to the region beyond the waist of the focused beam, i.e. to the region of the diverging wave. This means that the cell plays the role of a positive lens focusing the diverging wave—Figure 3. Thus, the self-focusing effect was registered for a very low power level, $W \sim 3$ to 10 mW; the corresponding power density on the cell was about 50 W/cm^2. The establishment time of the effect was about 10 s. The control experiments showed

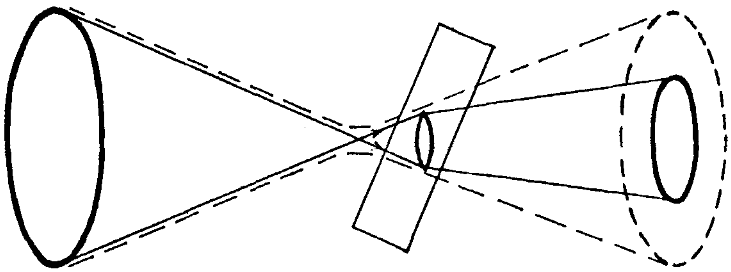


FIGURE 3 The effect of the collecting (self-focusing) lens on the light beam.

that, in correspondence with the theory, the effect was absent for a wave incident normal to the cell, $\alpha = 0$, and for an ordinary wave at any orientations of the cell.

1.2 Estimates

Detailed measurements of the dependence of the nonlinear lens force on the angle α and on the light intensity were carried out in these experiments. The detailed discussions of the results of the above as well as of the subsequent experiments and comparison with theory is postponed to Chapter 4. Here we shall confine ourselves to the simplest estimates confirming that in the experiment we had found just the effect we were seeking, i.e. the orientational optical nonlinearity.

The strength of the light wave electric field in a beam of 50 W/cm^2 in power is $|E| = 0.5$ esu $= 1.5.10^2$ V/cm. The anisotropic part of the field interaction energy with a NLC equals $U_E = -(\varepsilon_a/16\pi)|E|^2\cos^2(\alpha - \delta\theta)$, where $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$ is the anisotropy of the NLC dielectric constant at the light frequency; $\varepsilon_a = n_\parallel^2 - n_\perp^2 = 1.71^2 - 1.51^2 = 0.64$ in the experiment. For example, the director reorientation to an angle $\delta\theta > 0$ at $\alpha = 45°$ brings the optical axis nearer to the direction of the field E and decreases the energy by the quantity $\delta U_E(\text{erg/cm}^3) = -(\varepsilon_a/16\pi)\cdot|E|^2\delta\theta$. However, the planar orientation is fixed on the cell walls, so that the perturbation of $\delta\theta$ is maximum in the middle of the cell and vanishes on the walls. The corresponding density of the inhomogeneous deformation energy is $\delta U_d = K(\delta\theta/L)^2$, where L is the cell thickness, K(erg/cm) $\sim 10^{-6}$ is the Frank constant. Minimizing the sum $\delta U_E + \delta U_d$ we obtain the director reorientation angle $\delta\theta = \varepsilon_a|E|^2L^2/32\pi K \approx 6.10^{-2}$ rad in the experiment. Such a deflection of the optical axis leads, at $\alpha = 45°$, to a change in the extraordinary wave refraction index by the quantity $\delta n \approx (n_\parallel - n_\perp)\,\delta\theta \approx 1{,}2.10^{-2} > 0$. An additional phase shift $\delta\varphi =$

$2\pi z\delta n/\lambda \approx 10$ rad arises on the distance $z = L/\cos\alpha \approx 1.4L$ as a result. This value corresponds to the beam centre, where the intensity $|E|^2$ is maximum. In the beam edge $\delta\varphi \approx 0$. The wavefront of the central region of the beam turns out to be retarded with respect to the peripheries as a result of $\delta n > 0$, which means self-focusing of the light. Self-focusing effect in nonlinear optics is usually described in terms of the dependence of the medium dielectric permittivity ε at the light 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 medium—liquid carbon bisulphide CS_2—this constant is equal to $\varepsilon_2 = 10^{-10}$ cm^3/erg (see below, Chapter 2).

The value of ε_2 calculated with the use of the results of the experiment under consideration is $\varepsilon_2 \approx 0.07$ cm^3/erg, i.e. it is about 10^9 times greater than for CS_2. In this context the orientational optical nonlinearity has been referred to as "giant" (GON).

The order of magnitude of Frank constants, as is known, can be obtained proceeding from the assumption that at 100 per cent deformation over the scale of the order of molecular size a_m the disturbed free energy density Ka_m^{-2} coincides with Nk_BT, where $N \sim a_m^{-3}$ is the density, k_BT is temperature in energy units; this gives the following estimate $K \sim Na_m^2k_BT \sim k_BT/a_m$. On the other hand there is the following estimate for the orientational nonlinearity constant for an isotropic liquid consisting of optically anisotropic molecules: $\varepsilon_2(\text{IL}) \sim (Nk_BT)^{-1}$. We come as a result to the conclusion that if $\varepsilon_a \sim 1$ in the mesophase, the corresponding constant $\varepsilon_2(\text{GON}) \sim L^2/K$ is greater than $\varepsilon_2(\text{IL})$ by a factor

$$\frac{\varepsilon_2(\text{GON})}{\varepsilon_2(\text{IL})} \approx \left(\frac{L}{a_m}\right)^2$$

Actually that factor is about 10^9 in accordance with the experimental results if $L = 5.10^{-3}$ cm, $a_m \sim 10^{-7}$ cm. The establishment time of the nonlinearity increases practically to the same factor. This means that nearly the same value of $|E|^2t_p$ (t_p is the light pulse duration) is necessary to obtain a given value of the disturbance $\delta\varepsilon$ both in isotropic liquid, and in a mesophase.

More complicated deformations of LC will be considered in the forthcoming parts of the review. The scale of the light waves interference pattern will act there instead of the cell thickness L. With account of this replacement the above obtained estimate for the advantage factor works in general case.

2 BASIC NONLINEAR OPTICS

2.1 Electronic nonlinearity

In a strong enough light field

$$E_{\text{real}}(\mathbf{r},t) = \frac{1}{2}[E(\mathbf{r})e^{-i\omega t} + E^*(\mathbf{r})e^{i\omega t}] \tag{2.1}$$

the polarization P, i.e. the unit volume dipole moment, can contain terms of higher orders of E:

$$P_{\text{real}} = \chi^{(1)}E_{\text{real}} + \chi^{(2)}E^2_{\text{real}} + \chi^{(3)}E^3_{\text{real}} + \cdots \tag{2.2}$$

For simplicity we consider here a monochromatic field and do not write down explicitly the indices of the vectors and tensors E, P and $\chi^{(i)}$. In linear approximation the electric induction equals $D = E + 4\pi P = \varepsilon E$, where $\varepsilon = 1 + 4\pi\chi^{(1)}$, and $\chi^{(1)}$ is the linear polarizability (dimensionless in the esu system). It can be estimated by considering an electron wave function disturbance in the molecule in the first order of the dipole Hamiltonian of the interaction $U = -dE$. Indeed, $|\delta\Psi_i\rangle \sim (d_{io}E/\hbar\omega_{io})|\Psi_o\rangle$, where d_{io} is the transition dipole moment matrix element, $\hbar\omega_{io}$ is the energy difference. As a result, the value of the induced dipolar moment is $\langle d\rangle = \langle\Psi_o|\hat{d}|\delta\Psi_i\rangle = |d_{io}|^2(\hbar\omega_{io})^{-1}E = \beta E$, where $\beta = |d_{io}|^2/\hbar\omega_{io}$ is the molecular polarizability (having the dimensions of cm^3). Here we have assumed that the light frequency ω lies considerably below the frequencies ω_{io} of the main dipole-active transitions. Taking into account that $|d_{io}| \sim ea_{el}$, where e is the charge of the electron, a_{el} is the size of an electronic orbit, and considering that $\hbar\omega_{io} \sim e^2/a_{el}$ i.e. $\hbar\omega_{io}$ is of the order of the Coulomb energy of an electron with respect to the charged framework, we obtain the estimate for β, $\beta \approx a^3_{el}$. Designating the number of molecules in cm^3 by N, we have $P = N\langle d\rangle$, $\chi^{(1)} = N\beta$; $\varepsilon = 1 + 4\pi N\beta$. In typical dielectrics $a_{el} \approx 10^{-8}$ cm, $\beta \approx 10^{-24}$ cm^3, $N \approx 10^{23}$ cm^{-3}, $\chi^{(1)} \approx 10^{-1}$, $\varepsilon \approx 2$, so that the refractive index value $n = \sqrt{\varepsilon}$ is about 1,5.

From the previous consideration the important conclusion is that the parameter determining the weakness of the electron wave function perturbation is $d_{io}E/\hbar\omega_{io}$, which can be rewritten in the form E/E_{mol}. Here $E_{\text{mol}} = \hbar\omega_{io}/d_{io}$ is the typical strength of the intramolecular field, for which one can estimate that $E_{\text{mol}} \approx e/a^2_{el} \sim 0.5.10^7$ esu $\approx$ $1.5.10^9$ V/cm.

To estimate the medium nonlinear polarizabilities $\chi^{(2)}$, $\chi^{(3)}$ one must take into account that every additional degree of the external field E leads, in the framework of the perturbations theory, to the appearance of an additional degree of E_{mol} in the denominator, so that

$$\chi^{(2)} = c_1\chi^{(1)}/E_{\mathrm{mol}}, \qquad \chi^{(3)} = c_2\chi^{(1)}/E^2_{\mathrm{mol}} \tag{2.3}$$

Here c_1, c_2 are dimensionless coefficients of the order of 1. Note, that the relation $P_i = \chi^{(2)}_{ikl}E_kE_l$ changes the sign under the operation of the coordinate system inversion. Therefore $\chi^{(2)} \equiv 0$ (i.e. $c_1 = 0$) for media (or molecules) with a centre of symmetry. In crystals without the centre of symmetry the typical values of quadratic polarizabilities $\chi^{(2)}$ equal $\chi^{(2)} \approx 10^{-9} - 10^{-8}$ esu, which corresponds to $c_1 \sim$ 0,05 – 0.5.

The cubic nonlinearity $\chi^{(3)}$ is not constrained and, therefore, is different from zero in every medium. For glasses, crystals and other condensed media far from resonance $\chi^{(3)} \sim 10^{-15} - 10^{-14}$ cm^3/erg (in esu units), which corresponds to $c_2 \sim$ 0,75 – 2,5. The response of a system (nonlinear as well as linear) is practically instantaneous on a scale of a light wave period $T = 2\pi/\omega$, if the frequencies ω_{io} of the strongest transitions in the molecules lie in the ultraviolet, and the light frequency ω lies in the visible, i.e. if $\omega_{io} >> \omega$. Then the quadratic term $\chi^{(2)}E^2_{\mathrm{real}}$ describes two effects—appearance of polarization $P_{2\omega}$ at doubled frequency

$$P_{2\omega}(\mathbf{r})e^{-2i\omega t} = \chi^{(2)}E_\omega(\mathbf{r})E_\omega(\mathbf{r})e^{-2i\omega t} \tag{2.4}$$

i.e. the second harmonic generation, and appearance of the static polarization $P_0(\mathbf{r}) = \chi^{(2)}E_\omega(\mathbf{r})E^*_\omega(\mathbf{r})$ (optical rectification). The cubic term $\chi^{(3)}E^3_{\mathrm{real}}$ gives the third harmonic of the light

$$P_{3\omega}(\mathbf{r})e^{-3i\omega t} = \chi^{(3)}E_\omega(\mathbf{r})E_\omega(\mathbf{r})E_\omega(\mathbf{r})e^{-3i\omega t} \tag{2.5}$$

In addition the cubic term gives rise to an additional polarization at the initial frequency ω:

$$P_\omega(\mathbf{r})e^{-i\omega t} = \chi^{(3)}E_\omega(\mathbf{r})E_\omega(\mathbf{r})E^*_\omega(\mathbf{r})e^{-i\omega t} \tag{2.6}$$

We can say that in the relation $D_\omega\exp(-i\omega t) = \varepsilon E_\omega\exp(-i\omega t)$ the medium dielectric permittivity was changed by a quantity proportional to the light field intensity $|E|^2$:

$$\varepsilon = \varepsilon_0 + 0.5\, \varepsilon_2 |E|^2; \qquad \varepsilon_2 = 8\pi\chi^{(3)} \tag{2.7}$$

(the multiplier 0.5 in the first relation (2.7) is inserted by established convention). The electronic contribution to the constant ε_2 of condensed media is $\varepsilon_2 \sim 10^{-13} - 10^{-14}$ cm^3/erg. The quantity ε_2^{-1}, of dimension erg/cm^3, characterizes the intramolecular energy density of the order of $N\hbar\omega_{io}$, with which the light field is forced to "fight" (having the energy density of interaction with the medium of the order of $(\varepsilon - 1)|E|^2/8\pi$) when this light field "tries" to enlarge the electronic orbits.

2.2 Orientational nonlinearity of isotropic liquids

Most molecules have anisotropic polarizabilities at light frequency. For example, the polarizability tensor of carbon bisulphide is $\beta_{ik} = \beta_{\parallel} n_i n_k + \beta_{\perp}(\delta_{ik} - n_i n_k)$, where $\mathbf{n}$ is a unit vector along the molecular axis and $\beta_{\parallel} = 15.10^{-24}$cm^3, $\beta_{\perp} = 5.10^{-24}$cm^3. The interaction energy of the field with a molecule, averaged over a few light periods, has the form $U_E = -\beta_{ik} E_i E_k^*/4$, i.e. contains a part

$$\delta U_E = -(\beta_{\parallel} - \beta_{\perp})(\mathbf{nE})(\mathbf{nE}^*)/4 \tag{2.8}$$

which depends on the mutual orientation of the vectors $\mathbf{E}$ and $\mathbf{n}$. The initial isotropic distribution of the molecular axes in the presence of the field is modified by the Boltzmann factor

$$j(\mathbf{n}) \sim \exp[-\delta U_E(\mathbf{n})/k_B T]$$

where $k_B T$ is the temperature in energy units. The medium dielectric permittivity tensor $\varepsilon_{ik} = \hat{1} + 4\pi N \int \alpha_{ik}(\mathbf{n}) j(\mathbf{n}) dO_{\mathbf{n}}$ is also changed as a result. Omitting the indicies of the tensors one can write $\varepsilon = \varepsilon_0 + 0.5\varepsilon_2|E|^2$ in a first nonvanishing approximation by $|E|^2$ where the following estimate is obtained for ε_2:

$$\varepsilon_2 \sim 4\pi \left(\frac{\beta_{\parallel} - \beta_{\perp}}{\beta} \right)^2 \frac{(\varepsilon_0 - 1)^2}{N k_B T} \tag{2.9}$$

The comparison of the electronic and the orientational contributions to the constant ε_2 shows that

$$\frac{\varepsilon_{2\text{orient}}}{\varepsilon_{2\text{electr}}} \sim \frac{\hbar\omega_{io}}{k_B T}\left(\frac{\beta_{\parallel} - \beta_{\perp}}{\beta}\right)^2 \tag{2.10}$$

The advantage factor of the orientational nonlinearity $\hbar\omega_{io}/k_B T$ is of the order of 100, if $(\beta_{\parallel} - \beta_{\perp})^2/\beta^2 \sim 1$, $T \sim 300°K$, $k_B T \sim 2.6 \cdot 10^{-2} eV$, $\hbar\omega_{io} \sim 3eV$. Actually, for carbon bisulphide the orientational contribution to ε_2 is the main part, $\varepsilon_2 \sim 1.2 \cdot 10^{-10}$ cm^3/erg. One can say that the energy density "resisting" the orientational action of the light field is $Nk_B T$ (erg/cm^3).

For the advantage factor we have to pay by a longer establishment time. The orientational mechanism of nonlinearity responds with a time of the order of the orientation relaxation constant, $\tau \sim 10^{-12} - 10^{-10}$ s. This means that, in any case, the orientational nonlinearity does not allow the generation of the third harmonic of the light (there is no term $P \sim EEEe^{-3i\omega t}$ due to the orientational contribution).

The orientational nonlinearity of the LC isotropic phase increases as $(T - T_c)^{-1}$, i.e. by the Curie-Weiss law near the point of transition to the mesophase. The increase of the constant ε_2 by about a factor of 10 has been observed experimentally in this way for $T - T_c \sim 3°C$ with the relaxation time increasing by about the same factor.[1,2] It is difficult to investigate by traditional methods the optical nonlinearity for smaller values of $T - T_c$ because of the existence of strong opalescence.

2.3 Phase matching in nonlinear interaction of waves

The Maxwell equations can be approximately reduced to an D'Alambert type equation (we temporarily neglect the vectoral character of the field)

$$\Delta E - \frac{1}{c^2}\frac{\partial^2 D}{\partial t^2} = 0; \qquad D = \varepsilon E + 4\pi P^{NL} \tag{2.11}$$

The incident monochromatic wave in the problem of second harmonic generation can be taken in the form

$$E_1 \exp(ik_1 z - i\omega t); \qquad k_1 = (\omega/c)\sqrt{\varepsilon(\omega_1)} \tag{2.12}$$

Then, for the field $E_2(\mathbf{r})\exp(-2i\omega t)$ at frequency $\omega_2 = 2\omega$ one can obtain from (2.11)

$$\Delta E_2 + k_2^2 E_2 = -\frac{16\pi\omega^2}{c^2}\chi^{(2)} E_1 E_1 e^{2ik_1 z}; \qquad k_2 = \frac{2\omega}{c}\sqrt{\varepsilon(\omega_2)} \quad (2.13)$$

The solution of this equation, depending on z only, has the form

$$E_2 = c_1 e^{ik_2 z} + c_2 e^{-ik_2 z} + \frac{16\pi\omega^2 E_1 E_1}{c^2[(2k_1)^2 - k_2^2]} e^{2ik_1 z} \quad (2.14)$$

where the constants c_1 and c_2 are chosen so as to satisfy the conditions on the boundaries of the nonlinear medium. It is seen from (2.14) that the highest efficiency of the generation is achieved when $k_2^2 = (2k_1)^2$. This can be interpreted in quantum-mechanical language as the energy and momentum conservation laws in an elementary act of the three photon process

$$\hbar\omega + \hbar\omega \rightarrow \hbar(2\omega); \qquad \hbar\mathbf{k}_1 + \hbar\mathbf{k}_1 = \hbar\mathbf{k}_2 \quad (2.15)$$

In the language of classical waves the condition of the second harmonic effective generation

$$(\mathbf{k}_1 + \mathbf{k}_1')^2 = (2\omega)^2\,\varepsilon(2\omega)/c^2 \quad (2.16)$$

is usually called the phase matching condition. In general, the exciting field can consist of two plane waves with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_1'$. The Eq. (2.16) means that the second harmonic elementary waves excited in various points of the medium are added in phase during further propagation.† Phase matching condition for the third harmonic generation has approximately the same character: $(3\omega)^2\varepsilon(3\omega)/c^2 = |\mathbf{k}_1 + \mathbf{k}_1' + \mathbf{k}_1''|^2$. Special attention should be paid to the following term in nonlinear polarization

$$P(\mathbf{r})e^{-i\omega t} = \chi^{(3)}E(\mathbf{r})E^*(\mathbf{r})E(\mathbf{r})e^{-i\omega t} \quad (2.17)$$

The polarization (2.17) automatically satisfies the phase matching condition if the incident wave is plane $E \sim \exp(i\mathbf{k}_1\mathbf{r})$ with $|\mathbf{k}_1| =$

† Usually $\varepsilon(2\omega) > \varepsilon(\omega)$ because of dispersion and therefore, for phase matching one must use waves of various polarization types in anisotropic crystals (see any textbook on nonlinear optics).

$\omega\sqrt{\varepsilon}/c$, since $P(\mathbf{r}) \sim \exp(i\mathbf{k}_1\mathbf{r})$. An additional phase modulation of the plane wave appears

$$\varphi(z) = kz, \qquad k = \frac{\omega}{c}\sqrt{\varepsilon_0 + 0.5\varepsilon_2|E|^2} \approx k_0 + \frac{\omega\varepsilon_2}{4c\sqrt{\varepsilon_0}}|E|^2,$$

$$\delta\varphi(z) = \frac{\omega\varepsilon_2}{4c\sqrt{\varepsilon_0}}|E|^2 z \tag{2.18}$$

Consider another important case, when the incident monochromatic field consists of two plane waves

$$E(\mathbf{r})e^{-i\omega t} = (E_1 e^{i\mathbf{k}_1\mathbf{r}} + E_2 e^{i\mathbf{k}_2\mathbf{r}})e^{-i\omega t} \tag{2.19}$$

These waves induce in the medium (due to the orientational nonlinearity, for example) a disturbance of the dielectric permittivity of the form

$$\delta\varepsilon(\mathbf{r}) = \frac{1}{2}\varepsilon_2(|E_1|^2 + |E_2|^2)$$

$$+ \frac{1}{2}\varepsilon_2[E_1E_2^* e^{i(\mathbf{k}_1-\mathbf{k}_2)\mathbf{r}} + E_1^*E_2 e^{-i(\mathbf{k}_1-\mathbf{k}_2)\mathbf{r}}] \tag{2.20}$$

consisting of the homogeneous base $|E_1|^2 + |E_2|^2$ and the periodical volume grating caused by interference of the fields E_1 and E_2. Let us write explicitly the nonlinear part of the induction D, arising as a result of the scattering of the wave (2.19) on the disturbance (2.20). We have

$$D^{NL}(\mathbf{r})e^{-i\omega t} \equiv \delta\varepsilon(\mathbf{r})E(\mathbf{r})e^{-i\omega t} = \frac{1}{2}\varepsilon_2 M(\mathbf{r})e^{-i\omega t}; \tag{2.21a}$$

$$M(\mathbf{r}) = (|E_1|^2 + |E_2|^2)(E_1e^{i\mathbf{k}_1\mathbf{r}} + E_2e^{i\mathbf{k}_2\mathbf{r}}) \tag{2.21b}$$

$$+ (E_1E_2^*)E_2e^{i\mathbf{k}_1\mathbf{r}} + (E_1^*E_2)E_1e^{i\mathbf{k}_2\mathbf{r}} \tag{2.21c}$$

$$+ E_1^2E_2^*e^{i(2\mathbf{k}_1-\mathbf{k}_2)\mathbf{r}} + E_2^2E_1^*e^{i(2\mathbf{k}_2-\mathbf{k}_1)\mathbf{r}} \tag{2.21d}$$

If the waves E_1 and E_2 propagate at considerable angle relative to each other, the quantities $|2\mathbf{k}_1 - \mathbf{k}_2|$ and $|2\mathbf{k}_2 - \mathbf{k}_1|$ strongly differ from k, see Figure 4. One can say that the terms (2.21d) do not satisfy the phase matching conditions (i.e. the Bragg conditions for the scattering on corresponding gratings) and, therefore, are unimportant. The terms (2.21b) have a trivial nature—the correction to the medium refractive index due to the homogeneous base. It is remarkable that the complicated enough process—the recording of the dielectric permittivity interference disturbance $\delta\varepsilon \sim E_1 E_2^* \exp[i(\mathbf{k}_1 - \mathbf{k}_2)\mathbf{r}]$ and the subsequent scattering of the wave $E_2 \exp(i\mathbf{k}_2\mathbf{r})$—gives the induction

$$\delta D e^{-i\omega t} = \frac{1}{2}\,\varepsilon_2 |E_2|^2 E_1 e^{i\mathbf{k}_1\mathbf{r} - i\omega t} \tag{2.22}$$

automatically satisfying the phase matching condition. The expression (2.22) has the character of a correction to the refractive index of the wave E_1, but proportional to "another's" intensity $|E_2|^2$ only. Analogous statement can be made also for the second term in (2.21c). As a result, for the length of wave vectors k_1 and k_2 we can write

$$\begin{aligned} k_1^2 &= \left(\frac{\omega}{c}\right)^2 \left[\varepsilon_0 + \frac{1}{2}\,\varepsilon_2(|E_1|^2 + |E_2|^2) + \frac{1}{2}\,\varepsilon_2 |E_2|^2\right], \\ k_2^2 &= \left(\frac{\omega}{c}\right)^2 \left[\varepsilon_0 + \frac{1}{2}\,\varepsilon_2(|E_1|^2 + |E_2|^2) + \frac{1}{2}\,\varepsilon_2 |E_1|^2\right] \end{aligned} \tag{2.23}$$

Let us emphasize once again the surprising character of the result (2.23). Gratings of the dielectric permittivity are really recorded in

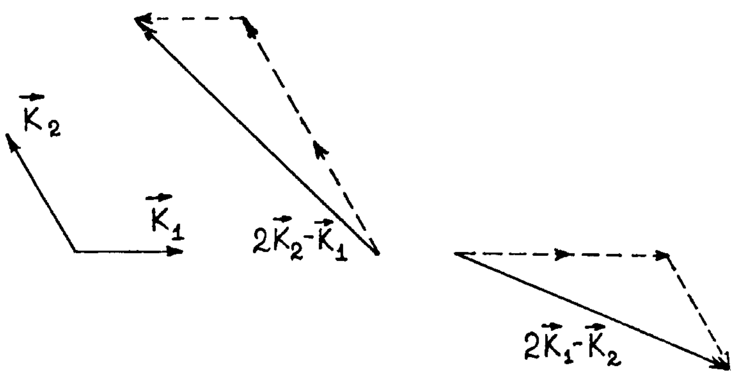


FIGURE 4 The vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ have their length determined by the wave equation, $|\mathbf{k}_1| = |\mathbf{k}_2| = \omega n/c$. The lengths of the vectors $2\mathbf{k}_1 - \mathbf{k}_2$ and $2\mathbf{k}_2 - \mathbf{k}_1$ do not satisfy the wave equation.

the medium. The waves satisfying the Bragg condition for scattering on these gratings are really propagating in the medium. However, for purely real $\varepsilon_2 = \varepsilon_2^*$ this scattering does not yield the real energy transfer from one wave to the other, but only modifies the effective refractive indices of the waves. The contribution of the "grating" processes under consideration is described by the last terms in the formulae (2.23).

Still one important case corresponds to the incidence of three waves on the medium:

$$E(\mathbf{r}) = E_1 e^{i\mathbf{k}_1\mathbf{r}} + E_2 e^{i\mathbf{k}_2\mathbf{r}} + E_3 e^{i\mathbf{k}_3\mathbf{r}}$$

Here, besides the above considered self-action processes as well as two-wave interaction, specific terms in nonlinear induction arise proportional to all the three wave amplitude's product; for example,

$$P_4(\mathbf{r})e^{-i\omega t} = 2\chi^{(3)} E_1 E_2 E_3^* \exp[i(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3)\mathbf{r} - i\omega t] \quad (2.24)$$

For arbitrary mutual orientation of the wave vectors $\mathbf{k}_1$, $\mathbf{k}_2$ and $\mathbf{k}_3$, the phase matching condition

$$(\omega/c)^2\varepsilon = (\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3)^2 \quad (2.25)$$

is not satisfied. Note a very important particular case. Let the waves E_1 and E_2 belong to the same polarization type and propagate exactly in the mutually opposite directions. Then $\mathbf{k}_1 + \mathbf{k}_2 = 0$, and the condition (2.25) is transformed to $(-\mathbf{k}_3)^2 = \omega^2\varepsilon/c^2$, i.e. turns out to be fulfilled. The polarization

$$P_4(\mathbf{r})e^{-i\omega t} = 2\chi^{(3)} E_1 E_2 E_3^* \exp[-i\omega t + i(-\mathbf{k}_3)\mathbf{r}] \quad (2.26)$$

effectively and coherently generates the wave E_4 with wave vector $\mathbf{k}_4 = -\mathbf{k}_3$ exactly opposite to the incident wave E_3 independent of the direction of $\mathbf{k}_3$. This means that, for a signal of complicated spatial structure $E_3(\mathbf{r})$, the wave $E_4(\mathbf{r})$ has the form

$$E_4(\mathbf{r})e^{-i\omega t} \sim E_3^*(\mathbf{r})e^{-i\omega t} \quad (2.27)$$

i.e. corresponds to the phase conjugated field. Generation of phase conjugated fields is one of the most active points in modern quantum electronics and nonlinear optics, see [3,4].

2.4 Wave picture of external self-focusing and self-diffraction

Assume that a plane monochromatic wave with intensity distribution $I(x,y) = |E_0(x,y)|^2$ inhomogeneous in transverse cross section (x,y) is incident on a nonlinear medium layer. Dielectric permittivity disturbance $\delta\varepsilon(\mathbf{r}) = 0.5\,\varepsilon_2\,|E(\mathbf{r})|^2$ arises in the medium and distorts the field phase distribution (wavefront). We shall assume for simplicity that the layer is not too thick so that deflection of the rays because of the wavefront distortion would not change considerably the distribution of amplitude $|E(\mathbf{r})|$ within the limits of the layer. Then the field at the exit from the layer of thickness L can be written in the form

$$E(z = L,x,y) = E_0(x,y)\exp\left[i\frac{\omega}{c}\sqrt{\varepsilon_0}L + i\frac{\omega\varepsilon_2 L}{4c\sqrt{\varepsilon_0}}|E_0(x,y)|^2\right] \qquad (2.28)$$

Further, it is necessary to solve the problem of wave propagation in a linear medium (more often—in vacuum or, which is approximately the same, in the air) along the positive direction of the z-axis with the initial condition (2.28) in the section $z = L$. We shall confine the discussion mainly to the form of this solution in the far zone, i.e. when $z \to \infty$. Experimentally, instead of the field distribution in the far zone, one sometimes observes the equivalent field distribution in the lens' focal plane.

Consider first the case where $E_0(x,y)$ represents a plane wave with a bell-shaped distribution of intensity, Figure 5a. Then the wavefront

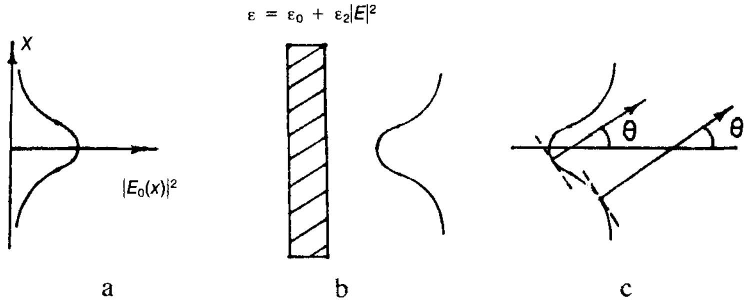


FIGURE 5 Intensity transverse distribution $|E_o(x)|^2$ in the incident beam (a), the form of the transmitted beam (b) and geometrooptical rays propagating to the far zone at the given angle θ (c).

of the radiation transmitted through the layer will be concave in the central region if $\varepsilon_2 > 0$, see Figure 5b. The ray deflection angle from the axis in the geometrical optics approximation is determined by the normal to the wavefront and can be calculated by the formula

$$\boldsymbol{\theta} = (\theta_x, \theta_y) = \frac{\varepsilon_2 L}{4\sqrt{\varepsilon_0}}\left(\frac{\partial}{\partial x}, \frac{\partial}{\partial y}\right)|E_0(x,y)|^2 \qquad (2.29)$$

Maximum deflections carry the rays corresponding to the intensity transverse profile bend point. One can estimate

$$\theta_{max} \sim \frac{\varepsilon_2 L|E_0|^2}{4\sqrt{\varepsilon_0}\, a_\perp} \qquad (2.30)$$

where $a_\perp$ is the transverse size of the beam with a bell-shaped profile.

Two points of wavefront (two beams) radiate simultaneously in the direction with a given angle $\boldsymbol{\theta}$, smaller than θ_{max}; it is not difficult to see that from a simple geometrical construction, Figure 5c. The corresponding points of the initial wavefront diverge when θ decreases from θ_{max} to zero, and when $\theta \to 0$ one point moves to the center of the beam and the other to the far periphery. The fields of the two beams add coherently in the far zone forming the typical structure of angular distribution in the form of interference circular fringes (see Figure 2). It is not difficult to understand that the total number of fringes corresponds to the phase difference in the periphery and in the beam center divided by 2π:

$$N = [\varphi(r = 0) - \varphi(r \to \infty)]/2\pi = \varepsilon_2 L|E_0|^2/4\sqrt{\varepsilon_0}\,\lambda \qquad (2.31)$$

Note that the first part of this formula is correct also in general case, when the simple relation (2.28) is not obeyed.

The intensity in the beam center may be represented in the form $|E(\mathbf{r})|^2 = |E(0)|^2(1 - 2r^2/a_\perp^2 + \cdots)$; then from (2.28) it follows for the nonlinear lens focal length

$$f^{-1} = \frac{\varepsilon_2|E(0)|^2}{\sqrt{\varepsilon_0}}\frac{L}{a_\perp^2} \qquad (2.32)$$

This formula, of course, is related only to the central part of the nonlinear lens; this lens possesses very strong aberrations with the latter giving rise to the fringe interference structure.

Consider now another important problem on light wave transformation by a nonlinear medium layer. Let the field $E(z = 0,x,y)$, incident on the layer, consist of two plane waves directed at small angles relative to the z-axis:

$$E_0(x) = E_1 e^{iq_1 x} + E_2 e^{iq_2 x}$$

where $|q_{1,2}/k| = |\sin\alpha_{1,2}| << 1$. We will assume that the intensity distribution is the same along the whole layer thickness:

$$I(x) = I_1 + I_2 + 2\sqrt{I_1 I_2}\cos(qx + \Delta\varphi)$$

where (2.33)

$$I_i = |E_i|^2, \qquad q = q_1 - q_2, \qquad \Delta\varphi = \arg(E_1 E_2^*) .$$

In the same approximation the layer can be considered as transparent with a given phase profile of transmission:

$$E(x,z = L) = E_0 \exp[i\varphi_0 + i\eta\cos(qx + \Delta\varphi)] ,$$

$$\eta = \frac{\omega\varepsilon_2 L}{2c\sqrt{\varepsilon_0}}\sqrt{I_1 I_2}, \qquad \varphi_0 = L\left[k_0 + \frac{\omega\varepsilon_2}{4c\sqrt{\varepsilon_0}}(I_1 + I_2)\right] \tag{2.34}$$

It is not difficult to obtain the expansion of the transmitted field into elementary plane waves using the known equality $\exp(i\eta\cos\gamma) = \sum_{-\infty}^{\infty} i^n J_n(\eta)\exp(in\gamma)$, where J_n is the Bessel function of index n. Further, in free space, the wave $E(z = L,x) = \exp(ipx)$ propagates, according to the wave equation, by the law $\exp[ipx + i(z - L)\sqrt{k_0^2 - p^2}]$. As a result, the field after the nonlinear medium layer has the form

$$E(x_1 z) = E_1 e^{i\varphi_0}\sum_{-\infty}^{\infty} c_n \exp[i(q_1 + nq)x + n\Delta\varphi + iz\sqrt{k_0^2 - (q_1 + nq)^2}];$$

$$c_n = [J_n(\eta) + iJ_{n+1}(\eta)\sqrt{I_1/I_2}] \cdot$$

$$\cdot \exp[-iL\sqrt{k_0^2 - (q_1 + nq)^2}] \tag{2.35}$$

Thus, beside the initial waves with the transverse components of wave vector q_1 and $q_2 = q_1 - q$, additional waves arise corresponding to higher order diffraction on the phase transparency with sinusoidal relief.

In the previous sect. 2.3 such "higher orders" of diffraction—the terms (2.21d)—were omitted as non-satisfying the phase matching condition. But here the violation of the phase matching condition is small due to the assumption that the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$ is small. The small thickness helps to compensate the remaining small discrepancy in the wave vector by the uncertainty relation $|\Delta k_z| \gtrsim L^{-1}$.

For simplicity we have assumed that the light waves are incident almost normally to the layer (incidence angle $\alpha_{air} << 1$). If α_{air} is not small, then it is necessary to use the length $L/\cos\alpha$ of the path that the wave passes through the nonlinear medium.

The remarkable advantage of the self-diffraction method is its high sensitivity. Namely, even for very small value of the parameter η, new waves are radiated in the directions $k_x = q_1 + q = 2q_1 - q_2$ and $k_x = q_1 - 2q = 2q_2 - q_1$. Their intensities, in this case, are small enough, of the order of $I_1\eta^2/4$ and $I_2\eta^2/4$, respectively. It is important, however, that there are no waves in these directions without nonlinearity. Thus, registration of a signal at a level $\sim 10^{-4}$ from incident (which is not difficult) allows one to find the existence of a nonlinear interference with the parameter $\eta \sim 2.10^{-2}$. The sensitivity of the method can be further increased by using the heterodyne technique to register the diffracted beams.

Fortunately, the nonlinearities of LC turn out to be so great that there was no need to improve the sensitivity.

If the intensity of one of the waves (for example $|E_2|^2$) is very small, the corresponding expressions for the intensities of various diffracted beams can be obtained either from (2.35) or directly. The latter method is simpler and is presented here. Let $E_{in} = E_1(\mathbf{r}) + E_2(\mathbf{r})$ and $t(r) = \exp[iC_1(|E_1|^2 + |E_2|^2) + iC_2E_1E_2^* + iC_3E_1^*E_2]$; the previous consideration corresponds to the particular case $C_1 = C_2 = C_3 = \partial\varphi/\partial|E|^2$. Then the transmitted field $E_t = E_{in}t(\mathbf{r})$ to a linear accuracy by E_2 or E_2^* can be represented in the form

$$E_t \approx e^{iC_1|E_1|^2}[E_1 + E_2(1 + iC_3|E_1|^2) + iE_2^*(C_2E_1^2)] \quad (2.35b)$$

In other words, the weak wave $|E_2|^2$ is amplified by a factor $|1 + iC_3|E_1|^2|^2$, and a complex conjugate wave $E' = E_2^*(iC_2E_1^2)$ appears.

2.5 What is stimulated light scattering?

Above in § 2.3 we considered recording of a space grating of the dielectric permittivity when two waves E_1 and E_2 of the same frequency interfere. Qualitatively different situations can arise if their frequencies differ and the nonlinearity establishes during the time $\tau = \Gamma^{-1}$, where $\Gamma(s^{-1})$ is the relaxation constant. Thus, let

$$E(\mathbf{r},t) = E_1 e^{i\mathbf{k}_1\mathbf{r} - i\omega_1 t} + E_2 e^{i\mathbf{k}_2\mathbf{r} - i\omega_2 t} \tag{2.36}$$

The establishment of perturbations of $\delta\varepsilon$ can be described with the aid of the simplest relaxation equation

$$\frac{1}{\Gamma}\frac{\partial\delta\varepsilon}{\partial t} + \delta\varepsilon = \frac{1}{2}\varepsilon_2[|E_1|^2 + |E_2|^2 + E_1^* E_2 e^{-i\mathbf{q}\mathbf{r} + i\Omega t} + E_1 E_2^* e^{i\mathbf{q}\mathbf{r} - i\Omega t}] \tag{2.37}$$

where $\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2$, $\Omega = \omega_1 - \omega_2$. Unlike the case with equal frequency waves, the interference gratings $\delta\varepsilon(\mathbf{r},t)$ are generated here with different amplitude and phase relatively to the base:

$$\delta\varepsilon_{\text{int}}(\mathbf{r},t) = \frac{1}{2}\varepsilon_2 \frac{E_1^* E_2}{1 + i\Omega/\Gamma} e^{i\mathbf{q}\mathbf{r} - i\Omega t} + C.C. \tag{2.38}$$

The (2.21d) type terms do not play a role due to their non-phase-matched character. The scattering of the wave $E_1\exp(i\mathbf{k}_1\mathbf{r} - i\omega_1 t)$ on the first term in (2.38) automatically satisfies the Bragg condition for the generation of the wave $E_2\exp(i\mathbf{k}_2\mathbf{r} - i\omega_2 t)$, i.e. gives induction

$$\delta D = \delta\varepsilon_{ef}(\omega_2)E_2 e^{i\mathbf{k}_2\mathbf{r} - i\omega_2 t}; \qquad \delta\varepsilon_{ef} = \frac{1}{2}\varepsilon_2|E_1|^2 \frac{1 - i\Omega/\Gamma}{1 + \Omega^2/\Gamma^2} \tag{2.39}$$

It is very important that the medium effective dielectric permittivity for the wave E_2 get an imaginary addition meaning amplification (or absorption). Actually $k_2 = \omega_2\sqrt{\varepsilon_0 + \delta\varepsilon_{ef}}/c \approx \omega_2\sqrt{\varepsilon_0}/c + \omega_2\delta\varepsilon_{ef}/2c\sqrt{\varepsilon_0}$ so that $|E_2|^2 \sim \exp(gz)$,

$$g = -\frac{\omega_2}{c}\frac{\mathcal{Im}\,\delta\varepsilon_{ef}}{\sqrt{\varepsilon_0}} = \frac{\omega_2\varepsilon_2|E_1|^2}{4c\sqrt{\varepsilon_0}}\frac{2\Omega/\Gamma}{1 + \Omega^2/\Gamma^2} \tag{2.40}$$

The gain coefficient g (cm^{-1}; by intensity) of the wave E_2 turns out to be proportional to the power density of the wave $|E_1|^2$. The process of E_2-wave amplification (for $\Omega = \omega_1 - \omega_2 > 0$) is conditioned by energy transfer from the wave E_1. The wave E_1 usually is called the "pump" and the wave E_2 the "signal." The process of energy transfer goes in opposite direction when $\Omega < 0$ and does not take place, according to the results of § 2.3, when $\Omega = 0$.

The process under consideration is called stimulated scattering of the orientational wing of the Rayleigh line, if the equation (2.37) describes relaxation establishment of orientational nonlinearity in an initially isotropic liquid. It can be shown that any kind of light scattering on equilibrium fluctuations of a medium dielectric permittivity (or on quantum fluctuations of individual molecules' polarizability) has its stimulated analog.

By analogy to the Einstein relation between spontaneous and stimulated emission of photons, there is a connection between the amplification g for light stimulated scattering and the extinction coefficient $d^2R/do d\omega$ ($s \cdot$ steradian$^{-1} \cdot$ cm^{-1}), characterizing spontaneous scattering:

$$g(\omega_2) = \frac{k_2^2}{(2\pi)^3} \frac{d^2R(\omega_1 \rightarrow \omega_2)}{do_2 d\omega_2} \cdot \frac{S_1}{\hbar\omega_1}\left\{1 - \exp[-\hbar(\omega_1 - \omega_2)/k_B T]\right\}; \qquad (2.41)$$

$$S_1 = c\sqrt{\varepsilon_0}|E_1|^2/8\pi \qquad (2.42)$$

Here S_1(erg/cm^2s) is the pump power density. The relation (2.41) can be written in the form $g = GS_1$, where G is a constant characterizing medium properties. Pump power density in nonlinear optics is usually expressed in Megawatts per cm^2, and then $[G]$ = cm/MW. Let us give some typical figures. For stimulated Brillouin scattering in carbon bisulphide $G \approx 0.15$ cm/MW, the constant of sound relaxation $\Gamma \approx 1.8.10^8\ s^{-1}$.

Usually the signal wave E_2 arises as a result of extremely weak ($dR/do \sim 10^{-7}$ cm^{-1}) spontaneous scattering of the pump E_1, so that very strong amplification $\exp(GS_1z) \sim e^{20}$ to e^{30}, is required for stimulated scattering observation, i.e. it is necessary to satisfy the threshold condition $GS_1z \gtrsim 20$–30.

The gain coefficient of light stimulated scattering characterizes the imaginary part of the corresponding component of the medium polarizability cubic tensor $\chi^{(3)}$. The knowledge of $J_m\chi^{(3)}(\Omega)$ allows one to reconstruct $Re\chi^{(3)}(\Omega)$ by dispersion relations of the Kramers-Kronig type. Thus the cubic polarizability tensor $\chi^{(3)}(\Omega)$ can be expressed in terms of the light spontaneous scattering cross-section. We do not write down the corresponding (complicated enough) formulae. Only the general conclusion is important for us; the media with strong thermal-equilibrium light scattering possess proportionally stronger cubic optical nonlinearity. It was just this statement that brought the authors of the present review to the studies of liquid crystal optical nonlinearity.

The thermal-equilibrium spontaneous light scattering in mesophases is known to be very large, $dR/dO \sim 10$ to $100\ \text{cm}^{-1}$, therefore the threshold condition for stimulated scattering here is $GS_1z \gtrsim 3$–5.

3 DERIVATION OF THE BASIC EQUATIONS OF THE THEORY

3.1 Euler-Lagrange-Rayleigh variational equations

The equations of the equilibrium state of liquid crystals are usually obtained from the variational principle according to which the free energy $\mathcal{F} = \int F d^3\mathbf{r}$ at a constant temperature in the established state must have the minimal value. If the free energy density F (erg/cm^3) depends on some number m of independent variables $U_m(\mathbf{r})$ and on their derivatives $\delta U_m/\delta x_j$, then use of the variational calculus standard methods gives the system of equilibrium equations

$$\frac{\partial}{\partial x_j}\frac{\delta F}{\delta(\partial U_m/\partial x_j)} - \frac{\delta F}{\delta U_m} = 0 \tag{3.1}$$

(the Euler-Lagrange equations). To describe the establishment processes it is necessary also to introduce the dissipative function density $R(\dot{U}_m)$, such that the rate of energy relaxation to heat will be equal to $2R$ (erg cm^{-3}s^{-1}). The dot in $\dot{U}_m$ means here the time derivative. Then, instead of (3.1) one must write

$$\frac{\partial}{\partial x_j}\frac{\delta F}{\delta(\partial U_m/\partial x_j)} - \frac{\delta F}{\delta U_m} - \frac{\delta R}{\delta \dot{U}_m} = 0, \tag{3.2}$$

the Euler-Lagrange-Rayleigh equations.

Usually densities of free energy F and dissipative function R are chosen by phenomenological considerations, with proper account of the requirements of invariance under groups of translation, rotation and so on. At first sight, it would be possible to write directly the phenomenological equations satisfying the same requirements.

The use of variational principle really gives no advantage, if we had only one independent function U. However in the presence of several variables (for example, Cartesian components of the director **n**) the use of the variational principle allows one to take account of reciprocity relations, which may turn out to be difficult to obtain directly from the written equations. For example, the "force" $f_m = \partial F/\partial U_m$ satisfies the symmetry relation $\partial f_m/\partial U_n = \partial f_n/\partial U_m$, since both derivatives are equal to $\partial^2 F/\partial U_m \partial U_n$. By analogy, when the dissipative "forces" f_m are obtained by differentiation of a single function R, they automatically ensure the fulfillment of the Onsager's principle of symmetry of the kinetic coefficients $\partial f_m/\partial \dot{U}_n = \partial f_n/\partial \dot{U}_m = \partial^2 R/\partial \dot{U}_n \partial \dot{U}_m$.[151,152]

Besides, the presence of special type symmetries of the function F, playing the role of a Lagrangian, allows one directly to write the conservation laws by the E. Noether theorem, see e.g. [102].

3.2 Elastic (Frank) part of the free energy

Nematics and cholesterics are defined by the unit vector of the director **n**, $|\mathbf{n}| = 1$, **n** and $-\mathbf{n}$ are equivalent. The density F of the distorted state free energy is taken in the form

$$F = \frac{1}{2} K_1(\mathrm{div}\mathbf{n})^2 + \frac{1}{2} K_2(\mathbf{n}\mathrm{rot}\mathbf{n} + q)^2 + \frac{1}{2} K_3[\mathbf{n}\mathrm{rot}\mathbf{n}]^2 \quad (3.3)$$

Here $q = 2\pi/h$, where h is the cholesteric helix equilibrium pitch, K_1, K_2, K_3 are the Frank constants having the dimensions of dyne; $q = 0$ for nematics. The dissipation function density will be taken in its simplest form

$$R = \frac{1}{2} \gamma \dot{\mathbf{n}}^2 \quad (3.4)$$

where γ (poise) is the constant of orientational viscosity. Note that here we neglect the interaction of the director with the hydrodynamic degrees of freedom, see in detail.[5,151,152] It is impossible to use directly eqs. (3.1) or (3.2) and the free energy (3.3) since the three quantities n_x, n_y, n_z are connected by the relation $n_x^2 + n_y^2 + n_z^2 = n^2 = 1$. There are two ways to deal with the problem. First, one can explicitly exclude the dependent variable: to take $n_z = (1 - n_x^2 - n_y^2)^{1/2}$, for example, or to use spherical coordinates θ, φ, or some other two independent variables. But if it is desirable for some reasons to preserve the form which symmetrically treats the three Cartesian components, n_x, n_y, n_z, it is possible to use the method of the Lagrange indefinite multiplier $\lambda(\mathbf{r})$, see [5]. The term $0.5\lambda(\mathbf{r})(n^2 - 1)$ is added to the free energy density in this method, so that the variational equations take the form

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

To determine the multiplier $\lambda(\mathbf{r})$ it is sufficient to multiply both sides of this equation by n_k and to take into account the relation $n_k n_k = 1$. This immediately shows that the equations obtained, with the relation $\mathbf{n}^2 = 1$, have the form

$$\Pi_{iK}\left[\frac{\partial}{\partial x_j}\frac{\delta F}{\delta(\partial n_k/\partial x_j)} - \frac{\delta F}{\delta n_k} - \frac{\delta R}{\delta \dot{n}_k}\right] = 0; \qquad \Pi_{ik} = \delta_{ik} - n_i n_k$$

These differ from the "u̯sual" equations (3.1), (3.2) by the multiplication by the operator $\hat{\Pi}$ of projecting on the plane perpendicular to the local direction of $\mathbf{n}(\mathbf{r})$.

Smectics A and C are defined by a more complicated set of variables. We discuss corresponding contributions to the free energy directly when we consider corresponding nonlinear-optical effects.

3.3 Electromagnetic part of the free energy (of the Lagrangian)

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

$$F_{\mathscr{E}} + F_{\mathscr{H}} = -\frac{1}{8\pi}(\varepsilon^0_{ik}\mathscr{E}_i\mathscr{E}_k + \mu^0_{ik}\mathscr{H}_i\mathscr{H}_k) \tag{3.5}$$

where ε^0_{ik} and μ^0_{ik} are the tensors of dielectric and magnetic permittivity. In liquid crystals ε^0_{ik} and μ^0_{ik} are usually uniaxial tensors having the form

$$\begin{aligned} \varepsilon^0_{ik} &= \varepsilon^0_{\perp}\delta_{ik} + \varepsilon^0_a n_i n_k \\ \mu_{ik} &= (1 + 4\pi\chi^-)\delta_{ik} + 4\pi\chi_a\left(n_i n_k - \frac{1}{3}\delta_{ik}\right) \end{aligned} \tag{3.6}$$

where $\chi_a \sim 10^{-7}$ is the anisotropy of magnetic permittivity

For light fields it is necessary to take into account that in the electromagnetic wave $|\mathbf{E}| \sim |\mathbf{H}|$. Since the anisotropy of the magnetic polarizability is 7 orders of magnitude smaller than the anisotropy of the dielectric permittivity, the influence of the magnetic field of the electromagnetic waves (including light waves) on the director orientation can be neglected. Besides, the square of real field E^2_{real} must be replaced by 0.5 EE^*, where E is the complex amplitude defined by formula (2.1); this way the terms at doubled light frequency are omitted. The symmetric, real tensor of the dielectric permittivity at light frequency, is also denoted by ε_{ik}, and then

$$F_E = -\frac{1}{16\pi}\varepsilon_{ik}(\mathbf{r})E_i(\mathbf{r})E^*_k(\mathbf{r}) \tag{3.7}$$

Variation of the sum of the expressions (3.3), (3.5) and (3.7) by variables $\mathbf{n}$ at fixed $\mathscr{E}(\mathbf{r})$, $\mathscr{H}$, $\mathbf{E}(\mathbf{r})$, $\mathbf{E}^*(\mathbf{r})$ and the expression (3.7) by variables $\dot{n}$, allows one to obtain the Euler-Lagrange-Rayleigh equations for the director relaxation rate or for the steady state case.

It is necessary to add to these equations the equations of electrostatics $\text{rot}\mathscr{E} = 0$, $\text{div}(\hat{\varepsilon}^0\mathscr{E}) = 0$ and the Maxwell equations for the monochromatic wave $\sim\exp(-i\omega t)$:

$$-i\frac{\omega}{c}\hat{\varepsilon}\mathbf{E}(\mathbf{r}) = \text{rot}\mathbf{H}, \qquad i\frac{\omega}{c}\mathbf{H} = \text{rot}\mathbf{E} \tag{3.8}$$

It is convenient to reduce (3.8) to a single equation

$$\text{rotrot}\mathbf{E} - \frac{\omega^2}{c^2}\hat{\varepsilon}(\mathbf{r})\mathbf{E}(\mathbf{r}) = 0 \tag{3.9}$$

We can use the Maxwell equations for monochromatic fields even for describing the transient effects of light interaction with the LC, since the light propagation time through the sample $\sim L/c \lesssim 10^{-11}s$ is many orders of magnitude shorter than any time for the LC orientation process.

It is remarkable that the equations (3.9) also can be obtained from the variational principle, if one takes this free energy (i.e. the Lagrangian with minus sign)

$$F_{\text{light}} = \frac{1}{16\pi}\left[\frac{c^2}{\omega^2}\left(\frac{\partial E_i}{\partial x_k}\frac{\partial E_i^*}{\partial x_k} - \frac{\partial E_i}{\partial x_k}\frac{\partial E_k^*}{\partial x_i}\right) - \varepsilon_{ik}(\mathbf{r})E_i(\mathbf{r})E_k^*(\mathbf{r})\right] \quad (3.10)$$

Here the quantities $\mathbf{n}(\mathbf{r})$ together with $\varepsilon_{ik}(\mathbf{r})$ must be considered fixed in the variation, and $\mathbf{E}(\mathbf{r})$ and $\mathbf{E}^*(\mathbf{r})$ must be taken as independent variables.

Thus, the whole set of the required equations can be obtained by variation of a single free energy—the sum of expressions (3.3), (3.5), (3.10)—by the independent variables $\mathbf{n}(\mathbf{r})$, $\mathbf{E}(\mathbf{r})$, $\mathbf{E}^*(\mathbf{r})$.

For example, consider the problem of the orientation of the NLC sample with initially homogeneous distribution of the director $\mathbf{n} = \mathbf{n}^0$ by the light with complex amplitude $\mathbf{E}(\mathbf{r})$. Consider also that there is a magnetic field $\mathcal{H} = \mathcal{H}\mathbf{n}^0$ applied to the sample parallel to the undisturbed director. The Euler-Lagrange-Rayleigh variational equations in linear approximation yield, by the perturbation $\delta\mathbf{n}(\mathbf{r},t) = \mathbf{n}(\mathbf{r},t) - \mathbf{n}^0$,

$$\gamma\frac{\partial\delta n_i}{\partial t} + K_2[\nabla_i(\nabla\delta\mathbf{n}) + (\mathbf{n}^0\nabla)^2\delta n_i - \Delta\delta n_i] - K_1\nabla_i(\nabla\delta\mathbf{n})$$
$$- K_3(\mathbf{n}^0\nabla)\delta n_i + (K_1 - K_2)n_i^0(\mathbf{n}^0\nabla)(\nabla\delta\mathbf{n}) + \chi_a\mathcal{H}^2\delta n_i$$
$$= \frac{\varepsilon_a}{16\pi}(\delta_{il}n_m^0 + \delta_{im}n_l^0 - 2n_i^0n_l^0n_m^0)E_lE_m^* \quad (3.11)$$

Here the condition $(\mathbf{n}^0\delta\mathbf{n}) = 0$ has been taken into account.

Since the wrong statements concerning the variational principle for a LC in light fields were published by some authors, we will discuss that question in more detail.

The energy density in a light wave consists of the "electrical" term $\varepsilon|E|^2/16\pi$ and the "magnetic" term $|H|^2/16\pi$. To simplify the consideration we neglect here the tensoral character and the dispersion of the quantity ε. As it is known, $\varepsilon|E|^2 = |H|^2$ in a traveling wave, so that the total density of the electromagnetic energy is twice as large as the "electrical" term only. The additional contribution to the energy of the dielectric in the presence of a light wave, as a result, is given by the expression

$$\overline{U}_E = 2\,\frac{\varepsilon|E|^2}{16\pi} \tag{3.12}$$

where the bar designates time averaging. Below we will use the well-known effect of electrostriction as an example. A medium is attracted into the region with the higher light intensity, and the corresponding additional pressure δP is negative and equals

$$\delta P = -\rho\,\frac{\partial\varepsilon}{\partial\rho}\,\frac{|E|^2}{16\pi} \tag{3.13}$$

This expression has been repeatedly checked theoretically and verified by experiments on stimulated scattering of light on hypersound waves. At first sight, Eq. (3.13) contradicts (3.12), since δP should be equal to

$$\delta P = \rho\,\frac{\partial U}{\partial\rho} \tag{3.14}$$

Here U is the energy density, ρ is the mass density. Actually, the variation of (3.12) at the fixed field strength E yields the expression twice as large as the correct one (3.13), and, what is even more important, with the wrong sign!

The settlement of this seeming contradiction was given by L. P. Pitaevski in a paper;[6] see also § 81 in the book.[7] The point is that for the adiabatic, i.e. sufficiently slow change of the dielectric permittivity ε, neither the field amplitude $|E|$ nor the induction amplitude $|D| = |\varepsilon E|$ nor the energy density $\varepsilon|E|^2/8\pi$ nor, at last, the Pointing vector $|\mathbf{S}| = c\sqrt{\varepsilon}|E|^2/8\pi$ or any of its components, are conserved. The quantity which is actually conserved is the adiabatic invariant, i.e. the ratio of the energy to the radiation frequency, which coincides with the number of quanta times Planck constant $\hbar$. In other words, the number of quantum state (i.e. the number of quanta) is conserved while we change adiabatically the parameters of the medium. On the other side, there is a functional dependence $\omega \sim \varepsilon^{-1/2}$ for the given type of oscillation (for the given mode of the electromagnetic field in space). That dependence can be easily derived for the example of a resonator of length L with refractive index $\sqrt{\varepsilon}$, where the frequency of the m-th mode is determined by the condition $L\omega_m\sqrt{\varepsilon}/c = m\pi$. We obtain the result that the frequency is virtually changed by the quantity $\delta\omega = -0.5\omega\delta\varepsilon/\varepsilon$ with the virtual change of ε. The virtual change of the energy δU, therefore, can be obtained, with the use of the conservation of the adiabatic invariant $A = U/\omega$, as

$$\delta U = \delta(\omega A) = A\delta\omega = -0.5A\omega\delta\varepsilon/\varepsilon = -0.5U\delta\varepsilon/\varepsilon \quad (3.15)$$

The use of (3.15) for variation in (3.14) leads to the expression (3.13) both with the right sign and with the right coefficient! That result was formulated [6] as the following. The force acting on the dielectric in the a.c. field can be obtained by variation of the electrical part of the energy only, taken with the $(-)$ sign, at the fixed field E:

$$\delta U = \delta(-\varepsilon|E|^2/16\pi)_{E=\text{const}} = -(|E|^2/16\pi)\delta\varepsilon \quad (3.16)$$

or the electrical part with the $(+)$ sign at the fixed induction D:

$$\delta U = \delta(|D|^2/16\pi\varepsilon)_{D=\text{const}} = -(|D|^2/16\pi\varepsilon^2)\delta\varepsilon$$
$$= -(|E|^2/16\pi)\delta\varepsilon \quad (3.17)$$

L. P. Pitaevski showed also[6] that the expressions (3.16), (3.17) remain correct if the frequency dependence of the dielectric permittivity is taken into account. The expressions are easily generalized also to the case of tensoral dielectric permittivity.

The essence of the mistakes made in a number of papers on the orientational action of light on LC we shall illustrate by the example of oblique incidence of a broad light beam on a planar layer of the medium, in which the electrostriction effect is developed. The considerations in those papers are approximately the following. Let us take the total energy density U and (neglecting dispersion $\varepsilon(\omega)$) express it in terms of the Ponting vector z-component S_z

$$S_z = \frac{c|E|^2}{8\pi}\sqrt{\varepsilon - \frac{\mathbf{q}^2c^2}{\omega^2}}; \qquad U = 2\,\frac{\varepsilon|E|^2}{16\pi} = \frac{\varepsilon S_z}{c\sqrt{\varepsilon - \mathbf{q}^2c^2/\omega^2}} \quad (3.18)$$

The z-axis is directed here perpendicular to the layer boundaries, and $\mathbf{q}$ is the wave vector transverse component; as it is known, the magnitude of $\mathbf{q}$ is conserved at propagation in the medium with z-dependent dielectric permittivity. Varying the expression (3.18) for U by ε and considering S_z = const (in contradiction with the conservation of the adiabatic invariant for virtual changes of ε), we get

$$\delta U \stackrel{?}{=} \left(\frac{\delta U}{\delta \varepsilon}\right)_{S_z = \mathrm{const}} \delta\varepsilon = \frac{1}{2}\frac{U}{\varepsilon}\delta\varepsilon\,\frac{\varepsilon - 2\mathbf{q}^2c^2/\omega^2}{\varepsilon - \mathbf{q}^2c^2/\omega^2} \qquad (3.19)$$

It is obvious that (3.19) is wrong. First of all it has the wrong sign, even at the normal incidence ($\mathbf{q} = 0$)! Actually, it would follow from (3.19) that the energy goes up along with the density increase at $\partial\varepsilon/\partial\rho > 0$, i.e. that the medium would be pushed out of the region occupied by the field and would not be attracted into it as takes place in reality. Moreover, $|\delta U|$ from (3.19) also turns out to be wrong at $\mathbf{q} \neq 0$.

Returning to the problem of LC in a light field let us repeat once more: the correct equations for the director are obtained by variation of the sum of the expressions (3.3) and (3.10) at fixed $\mathbf{E}(\mathbf{r})$.

4 GON INVESTIGATION IN CELLS WITH LC

We have already noted in § 1.2 that the energy density, with which LC resists the light field orienting force, decreases proportionally to a^{-2} when the space scale a of the inhomogeneity of the director disturbance increases. Therefore if a is of the order of the cell thickness, $a \sim 10^{-2}$cm, the nonlinearity has giant value—9 orders greater than the nonlinearity of liquid carbon bisulphide. Later we shall use the term "giant optical nonlinearity" (GON) for the dielectric permittivity distortion which is proportional to the 1-st power of the incident light intensity and having space scale a maximal possible for the given sample geometry. a will play the role of the cell thickness L or the beam cross section size $a_\perp$ if $a_\perp \lesssim L$.

4.1 Planar cell. Theory of GON

Consider a cell with a NLC in planar orientation, Figure 6. The normal to the cell walls we shall take coincident with the z-axis, and the undisturbed direction of the director $\mathbf{n}^0$ coincident with the x-axis, i.e. $\mathbf{n}^0 = \mathbf{e}_x$. Consider also that the orientation is strongly anchored

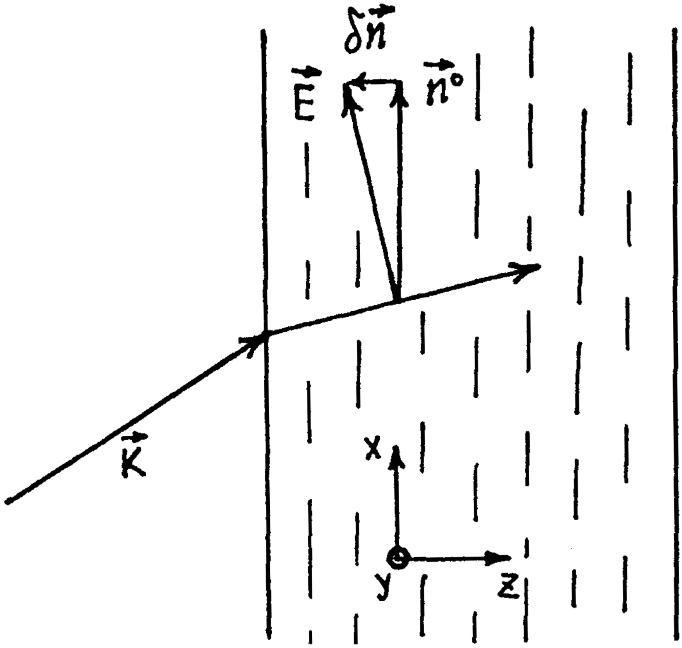


FIGURE 6 Oblique incidence of an extraordinary wave to the planar *NLC*-cell.

on the cell walls: $\mathbf{n}(z = 0,x,y) = \mathbf{n}(z = L,x,y) = \mathbf{e}_x$. Let a plane monochromatic wave of the extraordinary type with wave vector $\mathbf{k}$ and complex amplitude $\mathbf{E} = \mathbf{e}E$, where $\mathbf{e} = \mathbf{e}^*$ is the unit vector, propagate through the NLC. The disturbed state of the system we will seek in the form

$$\mathbf{n}(\mathbf{r},t) \approx \mathbf{e}_x + \mathbf{e}_z n_z(z,t) + \mathbf{e}_y n_y(z,t) \tag{4.1}$$

i.e. we consider the solution homogeneous in the (x,y)-plane. Then we get from (3.11)

$$\gamma \frac{\partial n_y}{\partial t} - K_2 \frac{\partial^2 n_y}{\partial z^2} = \frac{\varepsilon_a}{16\pi}(E_x E_y^* + E_x^* E_y)$$

$$\gamma \frac{\partial n_z}{\partial t} - K_1 \frac{\partial^2 n_z}{\partial z^2} = \frac{\varepsilon_a}{16\pi}(E_x E_z^* + E_x^* E_z) \tag{4.2}$$

where only terms of the 1-st order in $|E|^2$ were taken into account.

Let the light field intensity be turned on abruptly at the moment

$t = 0$. Since the right-hand side of Eq. (4.2) does not depend on z, one can easily obtain:

$$n_{y,z}(z,t) = \sum_{m=1}^{\infty} A_{y,z}^{m}(t) \sin \frac{m\pi z}{L}, \tag{4.3}$$

$$A_{y,z}^{m} = f_{y,z}^{m}(t) \frac{\varepsilon_a(\mathbf{e})_x(\mathbf{e})_{y,z}|E|^2}{4\pi^2\gamma\Gamma_{y,z}^{m}} \frac{1}{m}[1 - (-1)^m], \tag{4.4}$$

$$\Gamma_y^m = \frac{K_2}{\gamma}\left(\frac{m\pi}{L}\right)^2, \qquad \Gamma_z^m = \frac{K_1}{\gamma}\left(\frac{m\pi}{L}\right)^2,$$

$$f_{y,z}^{m}(t) = 1 - \exp(-\Gamma_{y,z}^{m} t) \tag{4.5}$$

The multiplier $m^{-1}[1 - (-1)^m]$ is nonzero for odd values of m only and corresponds to the constant function Fourier series expansion in the interval $(0, L)$. The constant $\Gamma^m(s^{-1})$ characterizes the establishment rate for the corresponding sinusoidal mode. Having the director disturbances (4.1), (4.3) it is not difficult to calculate, with the same accuracy, the disturbance of the dielectric permittivity $\delta\varepsilon_{ik} = \varepsilon_a(n_i^0\delta n_k + n_k^0\delta n_i)$ and the phase disturbance of the transmitted wave.

Let us discuss the structure of the director disturbance given by the expressions (4.3)–(4.5). Even in the transient regime ($\Gamma t \ll 1$), the mode with the lowest index $m = 1$ is excited the most effectively; the next mode with $m = 3$ has 3 times smaller amplitude when $\Gamma^3 t \ll 1$. When $\Gamma^1 t \gg 1$, a stationary distribution is established, for which the amplitude of the mode with $m = 3$ is 27 times smaller relatively to the mode with $m = 1$; therefore we shall confine further discussion to the contribution of this lowest mode.

If the light wave vector $\mathbf{k}$ is strictly perpendicular to the director $\mathbf{n}^0$, i.e. if $k_x = 0$, the polarization unit vector of the extraordinary wave coincides with $\mathbf{e}_x$ and the director disturbance is absent. The director disturbance is identically zero also for the ordinary wave of any direction of propagation because its polarization unit vector $\mathbf{e}$ is strictly perpendicular to the optical axis. Consider now an extraordinary wave with $k_x \neq 0$, $k_y \neq 0$. It generates deformations of both types: twist (δn_y) and splay (δn_z). Usually the constant K_2 for twist deformation is 2–3 times smaller than the constant K_1; therefore the corresponding contribution is established slower ($\Gamma \sim K^{-1}$), but attains, for other equal conditions, a greater stationary value.

Most of the experiments have been carried out in a geometry with $k_y = 0$, i.e. for the case, when the wave vector lies in the plane of the normal $\mathbf{e}_z$ to the walls and the undisturbed director $\mathbf{n}^0 = \mathbf{e}_x$. Therefore, we shall confine ourselves, for simplicity, to a discussion of this particular case, when the twist deformation is not excited. We shall carry out first the calculations for a crystal with a small optical anisotropy, $\varepsilon_a << \varepsilon_\perp$, in order to avoid cumbersome calculations. Then we can write $\mathbf{k} = k(\mathbf{e}_z\cos\alpha + \mathbf{e}_x\sin\alpha)$, where α is the refraction angle, $\mathbf{e} \approx \mathbf{e}_x\cos\alpha - \mathbf{e}_z\sin\alpha$, and the beam path length through the medium dl is related to the change of the z-coordinate by $dl \approx dz/\cos\alpha$, in other words, here we do not take into account the small (of the order of $\varepsilon_a/\varepsilon_\perp$) difference in the directions of the group and phase velocities. In the same approximation it is not difficult to obtain the field phase change due to the disturbance of $\delta\varepsilon$

$$\frac{d\varphi}{dz} = \frac{1}{\cos\alpha}\frac{d\varphi}{dl} = \frac{1}{\cos\alpha}\frac{\omega}{2cn}e_i\delta\varepsilon_{ik}(z)e_k \tag{4.6}$$

where $n = kc/\omega$ is the refractive index. As a result, we obtain for the mode with $m = 1$ in the stationary regime

$$\delta\varphi = \frac{\omega}{cn}\frac{\varepsilon_a^2\sin^2\alpha\cos\alpha L^3|E|^2}{\pi^5 K_1} \tag{4.7}$$

The z-component of the Ponting vector in the light wave with the same accuracy is equal to $S_z \approx S\cos\alpha \approx cn|E|^2\cos\alpha/8\pi$. If the expression (4.7) is written in the form $\delta\varphi = \omega L\varepsilon_2|E|^2/4cn\cos\alpha$ (compare with § 2.3) then we obtain the effective constant of nonlinearity ε_2:

$$\varepsilon_2 = \frac{4\varepsilon_a^2\sin^2\alpha\cos^2\alpha L^2}{\pi^5 K_1} \tag{4.8}$$

Thus, the constant of the giant optical nonlinearity (GON) turns out to be proportional to ε_a^2 (for moderate ε_a). The quantity ε_2^{-1}(erg/cm^3), after extraction of the multiplier ε_a^{-2} and of the angular dependence $(\sin\alpha\cos\alpha)^{-2}$, coincides with the energy density K_1/L^2 which is necessary for strong (~100%) splay deformation in a cell of thickness L.

As it is known, the order of magnitude of the Frank constants can be estimated by the relation $K/a_m^2 \sim Nk_BT$, where a_m is the molecular size, $N \sim a_m^{-3}$ is the density of molecules. In other words, the disturbed energy density for 100% deformation with the scale a_m, coincides with Nk_BT within an order of magnitude. Comparing the estimate (2.9) for the isotropic liquid (IL) orientational nonlinearity constant with ε_2 from (4.8) we conclude that the advantage factor for the GON is

$$\frac{\varepsilon_2(\mathrm{GON})}{\varepsilon_2(\mathrm{IL})} \approx \left(\frac{L}{a_m}\right)^2 \tag{4.9}$$

Actually, this factor is about 10^9 in agreement with experimental results for $L \sim 5.10^{-3}$ cm, $a_m \sim 10^{-7}$ cm. Practically the same factor governs the increase of the establishment time of the nonlinearity $\tau \sim \Gamma^{-1}$. That means that to obtain the given value of $\delta\varepsilon$ one needs approximately the same value of the product $|E|^2 t_p$ both for NLC and for IL; here t_p is the light pulse duration.

We considered orientational effects in the first order in the light field intensity. Let us estimate the power density, for which the reorientation angle becomes of the order of 1. The corresponding value of $|E|^2$ for $\sin\alpha\cos\alpha \sim 0.5$ is defined by the condition

$$\frac{\varepsilon_a|E|^2}{32\pi} \sim K\left(\frac{\pi}{L}\right)^2$$

Numerically, when ($L \sim 100$ μm, $K \sim 10^{-6}$ dyne, $\varepsilon_a \sim 0.7$, the required power density is about $2{,}5.10^3$ W/cm^2. Such values are accessible to argon lasers. Qualitatively new effects do not arise in the planar cell for greater power, but only nonlinearity saturation occurs.

It is interesting to note that an obliquely incident *o*-wave can also produce the reorientation without threshold in intensity. The mechanism is the narrow-angle strong thermal scattering of the incident beam and the subsequent *e*-wave-induced reorientation. To observe that effect one should use a separate probe light beam.

4.2 Narrow beams

Up to now we considered the case when an infinite plane wave is incident on a NLC layer. That model is valid only if the beam transverse size $a_\perp$ is larger than the layer thickness L. The situation is changed, if the beam is narrow, $a_\perp \lesssim L$.

The calculation of the self-focusing effect in the latter case we shall perform for a model of an infinite single-constant NLC, it's orientation along the direction $\mathbf{n}^0$ being held by a static magnetic field $\mathcal{H} = \mathcal{H}\mathbf{n}^0$. Here it is convenient to choose a coordinate frame with the z-axis along the beam axis so that $\mathbf{E} = \mathbf{e}_x E(xy)$, $\mathbf{n}^0 = \mathbf{e}_x \cos\alpha + \mathbf{e}_z \sin\alpha$, see Figure 7. The equation for the director perturbation $\mathbf{n}(x,y) - \mathbf{n}^0 \approx \theta(\mathbf{e}_x \sin\alpha - \mathbf{e}_z \cos\alpha)$ can be obtained from the variational principle:

$$\frac{\partial^2\theta}{\partial x^2} + \frac{\partial^2\theta}{\partial y^2} - \rho^{-2}\theta = -B|E(x,y)|^2 \tag{4.10}$$

Here $\rho = (K/\chi_a\mathcal{H}^2)^{1/2}$ is the magnetic coherence length, $B = \varepsilon_a \sin\alpha \cos\alpha/8\pi K$. Equation (4.10) can be considered as approximately correct also for the case when the initial orientation is held by the cell walls and not by a magnetic field, if one makes the substitution $\rho^2 \rightarrow L^2/\pi^2$, where L is the NLC layer thickness.

Solution of Eq. (4.10) can be obtained with the help of the Green function:

$$\delta\theta(x,y) =$$

$$-\frac{B}{4}\int |E(x',y')|^2 iH_0^{(1)}\left(i\rho^{-1}\sqrt{(x-x')^2+(y-y')^2}\right)dx'dy' \tag{4.11}$$

where $iH_0^{(1)}(iz) = 2K_0(z)/\pi$ is the zero-order Hankel function of the purely imaginary argument. However, this expression (4.11) is rather complicated and, therefore, we shall analyze separately the structure of the solution in various regions of space for various limiting cases.

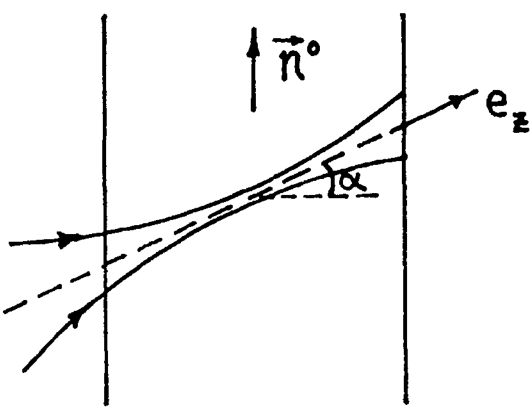


FIGURE 7 On the choice of the coordinate frame in the GON problem for a narrow light beam; $\mathbf{e}_z$ coincides with the beam axis, undisturbed director is $\mathbf{n}^0 = \mathbf{e}_x \cos\alpha + \mathbf{e}_z \sin\alpha$.

The space derivative in (4.10) can be neglected when the beam transverse size $a_\perp$ is large, $a_\perp >> \rho$; then the solution (4.10) has the form $\theta(x,y) = \rho^2 B|E(x,y)|^2$, and we reobtain the result of the previous section. In the other limit, when $a_\perp << \rho$, it is necessary to consider separately the director behaviour inside and outside the beam. Further, we shall consider separately the axial symmetric cylindrical beam $E = E(r)$, $r = \sqrt{x^2 + y^2}$, and the one-dimensional ribbon-shaped beam $E = E(x)$. For formulae simplification we also consider $E(r) = E_0$ inside the beam of diameter $2a_\perp$ (or of width $\Delta x = 2a_\perp$) and $E(r) = 0$ outside the beam. Inside the beam, and for $a_\perp << \rho$, the solutions of Eq. (4.10) are

$$\theta(r) = \frac{r^2}{4} B|E_0|^2 + \theta_2, \qquad \theta(x) = \frac{x^2}{2} B|E_0|^2 + \theta_1 \tag{4.12}$$

the first—for cylindrical and the second—for ribbon beams. The reciprocal focal length of the corresponding nonlinear lens in those cases is equal to (compare with § 2.4)

$$f_2^{-1} = \frac{\varepsilon_a^2 \sin^2\alpha \cos^2\alpha\, L|E_0|^2}{16\pi K}, \qquad f_1^{-1} = 2f_2^{-1} \tag{4.13}$$

The real distribution of intensity in the beam has a bell-shaped form, and then the fringes of aberrational self-focusing arise; their number equals

$$N \approx \frac{\varphi(r = 0) - \varphi(r = a_\perp)}{2\pi} = \frac{L}{\lambda} \frac{\varepsilon_a^2 a_\perp^2 |E_0|^2 \sin^2\alpha \cos\alpha}{32\pi K} \tag{4.14}$$

Here we related the number of fringes to the phase difference in the beam centre and at its edge, but no simply with the phase in the beam centre. The point is that, at $a_\perp << \rho$, the director perturbation differs considerably from zero far outside of the beam, where the intensity is equal to zero. The director deflection in the beam centre (θ_1 or θ_2) is defined not only by the local intensity $|E_0|^2$. For example, in the one-dimensional case at $a_\perp << \rho$ we have

$$\theta_1 \approx -\frac{1}{2} B\rho \int |E(x')|^2 dx' \tag{4.15}$$

i.e. $|\theta_1| \sim |\theta(0) - \theta(a_\perp)|\rho/a_\perp$, see Figure 8. On both sides of the ribbon shaped beam peculiar nonlinear prisms are formed in this case. How-

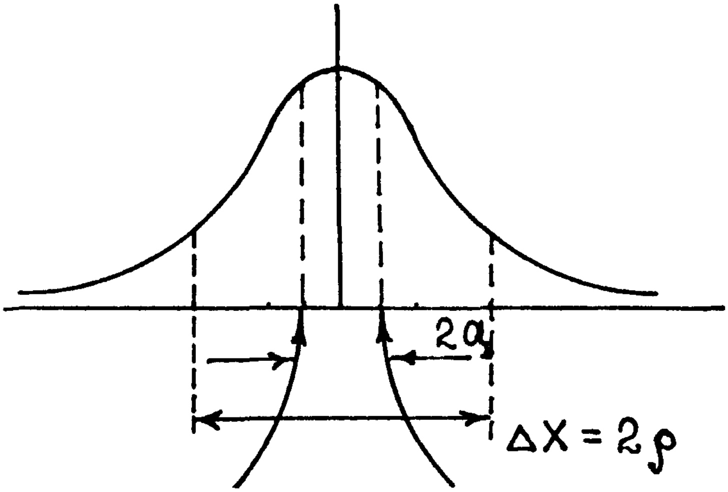


FIGURE 8 A narrow ($2a_\perp << \rho$) ribbon-shaped beam generates the director perturbations in the region $\Delta x \sim 2\rho$.

ever it is possible to register them with the aid of a probe beam only. Such tails of $\theta(r)$ distribution outside of the beam when $a_\perp << r \leqslant \rho$, for cylindrical initial beam, also must be present, but the quantity $\theta_2/|\theta(0) - \theta(a_\perp)|$ is not so large in that case; it is of the order of $\ln(\rho/a_\perp)$. This conclusion can be obtained from an analysis of the expression (4.11). For $\delta\mathbf{n}(\mathbf{r},t)$ establishment time estimate it is necessary to add the term $\gamma K^{-1}(\partial\theta/\partial t)$ in the left hand side of Eq. (4.10). As a result, one obtains that when $a_\perp << \rho$ the self-focusing lens must be established during the time $\tau \sim \gamma a_\perp^2/K$, and the magnitudes θ_1 and θ_2 together with the tails of the function $\theta(r)$—during the time $\tau^1 \sim \gamma\rho^2/K$.

4.3 Homeotropic cell. Theory of GON

The effects to first order in the light intensity for the homeotropic cell ($\mathbf{n}^0 = \mathbf{e}_z$) are described by practically the same equations. An obliquely incident ordinary wave does not cause GON, but an extraordinary wave deflects the director at an angle $\delta\theta \approx n_y(z,t)$ in the plane of n^0 and the wave vector $\mathbf{K}$, where the x-axis is chosen so that $\mathbf{K} = K(\mathbf{e}_y\sin\alpha + \mathbf{e}_z\cos\alpha)$, α is the refraction angle. For $\delta\theta(z,t) \sim |E|^2$ we have the equation

$$\gamma\frac{\partial\delta\theta}{\partial t} - K_3\frac{\partial^2\delta\theta}{\partial z^2} = \frac{\varepsilon_a}{16\pi}(E_yE_z^* + E_y^*E_z) \qquad (4.16)$$

The solution of this equation will have a form analogous to Eqs. (4.3)–(4.5) for n_y with the simple substitution $K_2 \to K_3$. The phase shift in the stationary case is equal to

$$\delta\varphi = \frac{\omega}{cn} \frac{\varepsilon_a^2 \sin^2\alpha \cos\alpha L^3 |E|^2}{\pi^5 K_3} \tag{4.17}$$

This expression has also been written within a relative error of the order of $\varepsilon_a/\varepsilon_\perp$. The stationary GON in a homeotropic cell is somewhat weaker than in a planar cell for the same conditions, since usually the constant of the bend effect is greater than the splay effect constant, $K_3 > K_1$. For the same reason, however, the hometropic cell is more transparent than the planar one.

Here, in the meanwhile, we do not touch upon the light induced Friedericksz transition, which can develop in homeotropic cells even at normal incidence; it requires much higher power density for observation and will be considered in chapter 5.

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

Consider still one problem of interest for comparison with experiment. Let two waves be incident on a cell (e.g. a homeotropic one), both having the refraction angle α to the director and a very small angle β relative to each other: $\mathbf{k}_{1,2} = k(\mathbf{e}_z \cos\alpha + \mathbf{e}_y \sin\alpha \pm \mathbf{e}_x \beta/2)$; then the intensity $|E|^2$ will be modulated by the law

$$|E(x)|^2 = |E_1|^2 + |E_2|^2 + E_1 E_2^* e^{iqx} + E_1^* E_2 e^{-iqx} \tag{4.18}$$

where $q = 2\pi/\Lambda$; $\Lambda = \lambda/n\beta = \lambda/\beta$; β_{air} is the angle between the waves in air. In that case Eq. (3.11) takes the form

$$\gamma \frac{\partial \delta\theta}{\partial t} - K_2 \frac{\partial^2 \delta\theta}{\partial x^2} - K_3 \frac{\partial^2 \delta\theta}{\partial z^2} = \frac{\varepsilon_a}{8\pi} |E(x)|^2 \sin\alpha \cos\alpha \tag{4.19}$$

The behaviour of x-independent terms was considered above; the interference grating is also recorded:

$$\delta\theta(x,z,t) = (1 - e^{-\Gamma t}) \frac{\varepsilon_a |E_1 E_2|}{\pi^5 [K_3(\pi/L)^2 + K_2 q^2]} \cdot \cos(qx + \Delta\varphi) \cdot \sin(\pi z/L) \tag{4.20}$$

Here $\Delta\varphi = \arg(E_1^* E_2)$, $\Gamma = [K_3(\pi/L)^2 + K_2 q^2]/\gamma$; we confine ourselves, as before, only to the lowest harmonic $\sin(m\pi z/L)$ with $m = 1$. As a result, the LC layer may be considered as a phase modulating film with the phase shift $\delta\varphi(x) = \varphi_0 + \eta\cos(qx + \Delta\varphi)$, where φ_0 is given by expression (4.17) with the substitution $|E|^2 \to |E_1|^2 + |E_2|^2$, and the parameter η in the established regime is equal to

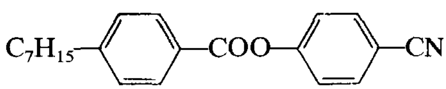
$$\eta = \frac{\omega}{cn}\frac{2\varepsilon_a^2 \sin^2\alpha\cos\alpha L|E_1E_2|}{\pi^3[K_3(\pi/L)^2 + K_2q^2]}$$

$$= \varphi_0 \frac{2|E_1E_2|}{|E_1|^2 + |E_2|^2}\left(1 + \frac{K_2q^2L^2}{\pi^2K_3}\right)^{-1} \qquad (4.21)$$

The expression (4.21) shows that the transverse inhomogeneity of $|E(x)|^2$ leads to the additional term K_2q^2 in the deformation energy; therefore the interference term $|E_1E_2|\cos(qx + \Delta\varphi)$ gives a weaker response. The electrodynamic problem of the light diffraction on such a phase film has been discussed in § 2.4.

4.4 Planar cell with a nematic: experiment

Qualitative discussion of the results obtained in [8] was presented in § 1.1. Here we discuss them in more detail. A cell in planar orientation was used with a NLC, comprised of 60% of mixture "A" and 40% of the substance



The liquid crystal had a nematic phase at room temperature and initially was chosen in order to obtain good homeotropic alignment in a large volume by applying a radio-frequency electric field (it has $\varepsilon_a > 0$ at radio-frequency). For a number of reasons, however, electrical orientation in a cell ~3 mm thick turned out to be unsatisfactory and the first experiment was carried out using the same NLC with planar orientation supported by the walls, preliminarily rubbed with diamond paste.

The parameters of the medium were $\eta_\perp \approx 1.51$, $\eta_\parallel \approx 1.71$, $K_1 \approx 8.5.10^{-7}$ dyne, the cell thickness was $L = 60$ μm. The quantitative measurements of the self-focusing effect of a He – Ne laser radiation ($\lambda = 0.628$ μm) with ideal Gaussian transverse profile were carried out in two ways. In the first method different lenses were placed in place of the cell. Their focal length was chosen to give approximately the same change of the beam angular divergence, as the self-focusing effect gave. That method was convenient in the case where the number of fringes in the self-focusing picture was small, $N \leqslant 1$, i.e. the divergence change was of the order of the initial diffraction divergence of the Gaussian beam. The next method consisted of the measurement of the number of fringes in the far zone of the transmitted beam, if $N \geqslant 1$.[9] The results of both methods gave practically the same value for the nonlinearity constant. Figure 9 represents the dependence of the nonlinear lens equivalent force f^{-1} on the beam power (Figure 9a) and on the angular factor $\sin^2\alpha\ \cos\alpha$ (Figure 9b). Both plots demonstrate good linear dependence in accordance with the theory.

The absolute comparison of the expression (4.8) with the experiment requires knowledge of the power density $cn|E(0)|^2/8\pi$ in the beam waist, which is one of the most difficult questions of experiment. The following expression describing the intensity profile change of an ideally focused Gaussian beam during its diffraction in the air can be used for this purpose

$$|E(x,y,z)|^2 = |E_0|^2(1 + z^2/z_0^2)^{-1} \exp[-2r^2/a_\perp^2(1 + z^2/z_0^2)] \quad (4.22)$$

Here $z_0 = ka_\perp^2/2$, $2a_\perp$ is the waist diameter (the full width at the level equal to e^{-2} of maximum the intensity, $2a_\perp = FWe^{-2}M$), z_0 is

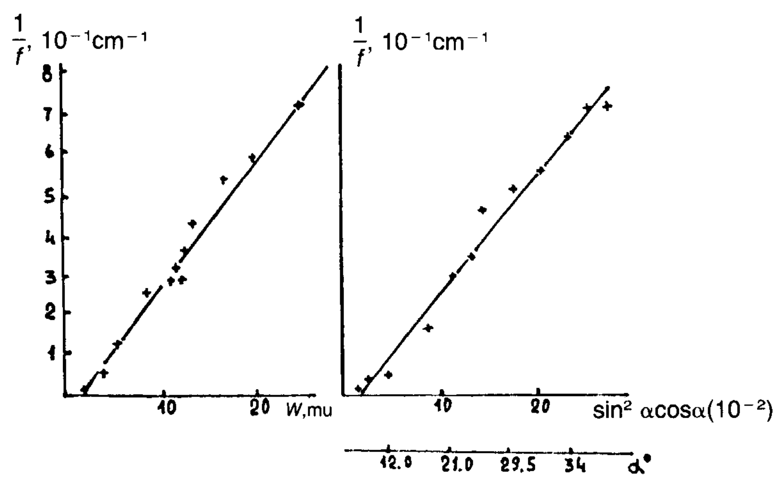


FIGURE 9 The dependence of the inverse focal length on the beam power (a) and on the geometrical factor $\sin^2\alpha\cos\alpha$ (b) [8].

the length of the waist, $\lambda = 2\pi/k$ is the wavelength in the air. This beam in the far zone has a Gaussian angular distribution, $j(\theta) \sim \exp(-2\theta^2/\theta_0^2)$ with the angular width $\Delta\theta(FWe^{-2}M) = 2\theta_0 = 2/ka_\perp$. The magnitude $2/Ka_\perp = \lambda/\pi a_\perp$ is the diffraction divergence of a beam of size $a_\perp$ which has a plane wavefront in the waist. If the focused beam is obtained by transmitting a parallel Gaussian beam of a diameter $D = FWe^{-2}M$ through a lens with a focal length f, then $\theta_0 \approx D/2f$ and consequently $a_\perp \approx 2f/kD$; it is assumed that $a_\perp << D$. The total power carried by the beam equals $S = ca_\perp^2|E_0|^2/16$.

In the experiments[8,9] the focusing of a He—Ne laser beam by a lens with $f = 25$ cm into the cell gave $2a_\perp \approx 240$ μm, $z_0 = 7$ cm. The magnitude $2a_\perp$ was much larger than the thickness of the LC layer ($L = 60$ μm); that justifies the use of the theory developed for an infinite plane wave. The magnitude $z_0 = 7$ cm exceeded considerably the whole thickness of the cell, 8 mm (LC + glass plates).

Experiment gave $\varepsilon_2 \approx 0{,}07$ cm³/erg for the refraction angle $\alpha = 32°$. Theoretical estimate by formula (4.8) gave exactly the same value. Such an coincidence is, obviously, the result of accidental compensation of a number of errors; for example, the values of the constants K_1, $n_\parallel - n_\perp$ and the beam power density were known with an accuracy of about 20% each.

Experimentally the establishment time is most conveniently defined as the interval τ during which the number of self-focusing fringes is changed by an amount of $1 - e^{-1} \approx 0.63$ of its stationary value. It turns out to be identical for intensity switching on and for its abrupt decrease to a very low value (if the light is completely switched off, there is nothing to be focused). For the experimental conditions [8,9] it was $\Gamma^{-1} \sim 10$ s. Hence by formula (4.5) the orientational viscosity constant can be estimated as $\gamma \sim 2$ poise—a quite reasonable value for such media (the value of γ for that particular substance was not known).

Experimental investigation of argon laser ($\lambda = 0.49$ μm) radiation self-focusing in a planar cell with MBBA of 50 μm thick has been carried out in the paper.[10] The method of nonlinear lens force measurement was somewhat different from that used in.[8,9] Quite satisfactory correspondence with the simple theory for a plane wave was obtained, although the beam width in the waist ~40 μm was already comparable to the cell thickness. The measured value of ε_2 for 50 μm thick MBBA and for the refraction angle $\alpha = 30°$ was 0.009 cm³/erg. The relaxation time was 4.3 s. Orientational self-focusing gradually was changed to thermal self-defocusing after exposure of ~4 min to a power of about 100 mW.

In the paper[11] the aberrational fringes number (N) dependence on the cell thickness (L) has been investigated. For this purpose two planarly oriented wedge-shaped cells with MBBA were used, allowing the choice of the local thickness in the range from 17 to 100 μm (to an accuracy of ~1 μm) and from 150 to 500 μm (to an accuracy of ~3.5 μm). The waist diameter of the focused radiation of the He—Ne laser, $\lambda = 0.628$ μm, with Gaussian profile was $2a = FWe^{-2}M \approx 46$ μm. Experimentally the linear dependence of the fringes number (N) on the parameter $\sin^2\alpha \cos\alpha$ and on the intensity was confirmed. The dependences of $\ln N$ on $\ln L$ and $\tau(L)$ are represented in Figure 10 for the incidence angle $\alpha_{air} = 55°$. The dependence $N(L)$ for the cell

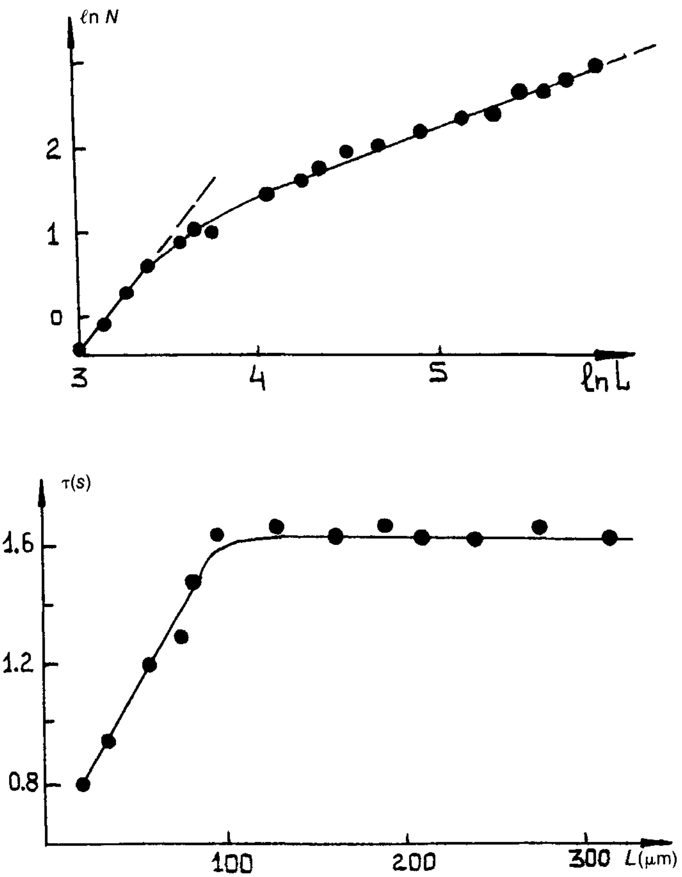


FIGURE 10 The dependence of the fringe number and the establishment time on the cell thickness [11].

of small thickness ($L \leqslant a\sqrt{2}$) is cubic according to the theory for broad beams; for large L, the experimental values of N increase somewhat slower than $N \sim L^1$; the latter would follow from the theory for narrow beams. The last effect was probably related to light scattering effects in a thick cell. The establishment time, measured by the appearance of the first several fringes, increases with L if $L \leqslant a\sqrt{2}$ and becomes constant $\tau \sim 1.7$ s at $L >> a$. The value $\tau = 0.8$ s for $L = 37$ μm coincides to an accuracy of 10% with the theoretical expression $\tau \sim \gamma L^2/\pi^2 K_1$ if $\gamma = 0.7$ poise, $K = 10^{-6}$ dyne. The constant τ is in good agreement with the same expression where $L >> a$, if one takes $a\sqrt{2}$ instead of L. Detection of the nonlinear phase shift caused by the GON is possible not only by self-focusing or self-diffraction, but also by the effect of interference of the wave under consideration with a reference one not subjected to nonlinear changes. If the two waves have identical polarization, the interference leads to intensity modulation. But if the waves have orthogonal polarizations, the result of interference appears as the change of the polarization state. In the experiment[12] on the GON investigation an ordinary wave played the role of the reference wave; it was obtained from the initial beam having oblique polarization; the extraordinary wave under consideration was obtained from the same beam and gave rise to the GON. It is essential that the phase of the *o*-type wave was not changed by the smooth GON-reorientation of the director.

The intensity dependent change of the transmitted wave polarization was observed in;[12] it corresponded to the *e*-wave self-focusing. Let us emphasize that the *o*-type wave did not take part in the nonlinear interaction and played the role of a reference wave.

4.5 Homeotropic cell with a nematic: experiment

The GON in a hometropic cell was observed in the paper[13] by the focusing of a powerful enough (~ 0.12 W) radiation of the argon laser ($\lambda = 0.5145$ μm) to a cell with a NLC OCBP. In that paper the fringe structure of the light self-focusing for oblique incidence of the extraordinary wave was registered. The waist size was about 100 μm; two cells were used with thickness 50 μm and 150 μm respectively. The divergence of the self-focusing type was gradually increased with the incidence angle increase for relatively low power level ($W \leqslant 40$ mW). The number of fringes and the divergence angle $\Delta\theta$ strongly depended on the power (from 60 to 100 mW) and the saturation level $\Delta\theta_{max} \sim 30°$ was achieved at the higher power.

Detailed experimental investigations of self-focusing and self-diffraction in hometropic cells with MBBA were carried out in the paper.[14] Argon laser radiation, $\lambda = 0.5145$ μm, was focused on the sample; the waist size $2a$ was about 40 μm; the thicknesses of the cells used were 25, 50, and 75 μm. The observed number of self-focusing fringes agreed well with the theoretical predictions both in absolute value, and in the functional dependence on the incidence angle. Thus a nonlinear lens with the focal length $f \approx 20$ cm was produced in a 50 μm cell for incidence angle 25° and power density 20 W/cm^2.

A homeotropic cell of 75 μm thick MBBA has been used in the paper[14] for self-diffraction effects investigations. The incidence of two coherent waves separated by an angle $\beta_{air} \sim 0.8°$ gave an interference picture $\sim\cos qx$ with a period $\Lambda = 2\pi/q = \lambda/\beta_{air} = 36.7$ μm for $\lambda = 0.5145$ μm. The interference grating should be recorded $[1 + (K_2/K_3)(2L/\Lambda)^2] \approx 10$ times weaker, than the homogeneous part of the disturbance, here we took $K_2 = 4.10^{-7}$ dyne, $K_3 = 7.5.10^{-7}$ dyne, $K_2/K_3 = 0.53$, and $L = 75$ μm for MBBA.

The parameter η of the periodic phase modulation (see formulae (2.34) and (4.16)) is very small for power levels $S_1 \sim 5$ W/cm^2 and $S_2 \sim 3$ W/cm^2. The intensity registration in the plus first and the minus second orders of the diffraction ($n = +1$ and $n = -2$ in the formula (2.35)) allowed one to measure the dependence of the parameter η on the incidence angle.

The dependence turned out to be in good agreement with the theory up to an angle of 25°. Strong self-focusing was observed for a larger angle; it distorted the experimental results on self-diffraction. The dependence of the diffracted beam intensities on the laser radiation intensity was $S_{dit} \sim S_{in}^3$ in accordance with the theory up to $S_{in} \sim 30$ W/cm^2. Self-focusing again distorted the results for higher intensity. Absolute measurements of the diffracted wave intensities also gave good agreement with the theory; numerically a power $\sim 10^{-2}$ W/cm^2 was radiated to the $+1$st order of diffraction.

In the paper[15] the phase grating was recorded due to the GON mechanism in a homeotropic NLC-cell when two waves interfered: one plane (E_1) and the second—with a complicated wave-front ($E_3(\mathbf{r})$). These waves were directed to the cell with an incidence angle 15°; their central directions made an angle 0.7° with each other. The reconstruction of the thin hologram obtained $\delta\varphi(\mathbf{r}) \sim E_1E_3^*(\mathbf{r}) + E_1^*E_3(\mathbf{r})$ was carried out by a wave counter-propagating to the E_1-wave. As a result of reconstruction, besides other waves, the phase-conjugate wave $E_4 \sim E_1E_2E_3^*(\mathbf{r})$ was generated propagating opposite to the wave $E_3(\mathbf{r})$. The ratio $|E_4/E_3|^2$ was $\sim 10^{-2}$ for a power density 10 W/cm^2 in each of the three waves. Strong effects of the GON-self-focusing led to considerable distortion of the wave $E_4(\mathbf{r})$ in comparison with the ideally phase-conjugate configuration $E_4 \sim E_3^*$.

4.6 NLC-cell with hybrid orientation

Besides planar and homeotropic cells, it is possible in principle to make a cell with a tilted orientation, homogeneous over the whole volume: $\mathbf{n} = \mathbf{e}_z \sin\theta_0 + \mathbf{e}_x \cos\theta_0$, $\theta_0 = \text{const}$. Then even for a normally incident wave, the angle θ_0 between the wave vector of the e-type wave and the director lies in the interval between 0° and 90°; therefore GON must arise. If $\varepsilon_a/\varepsilon_\perp << 1$ we can take $\mathbf{E} = \mathbf{e}_x E$ for the vector $\mathbf{E}$ of the normally incident extraordinary wave. The nonlinear phase shift in this approximation equals

$$\delta\varphi = \frac{\omega}{cn} \frac{\varepsilon_a^2 \sin^2\theta_0 \cos^2\theta_0 \, L^3 |E|^2}{\pi^5 K_{ef}} \tag{4.23}$$

where $K_{ef} = K_1 \cos^2\theta_0 + K_3 \sin^2\theta_0$; the director is supposed to be fixed at the boundaries of the medium.

Here we shall discuss the case of hybrid orientation, where planar orientation is fixed on the wall at $z = 0$, $\mathbf{n}(z = 0) = \mathbf{e}_x$, and homeotropic orientation on the wall at $z = L$, $\mathbf{n}(z = L) = \mathbf{e}_z$. Then

$$\mathbf{n}(z) = \mathbf{e}_x \cos\theta(z) + \mathbf{e}_z \sin\theta(z) \tag{4.24}$$

and in the approximation $K_1 = K_3 = K$ the equilibrium profile $\theta(z)$ is linear in the absence of external fields: $\theta(z) = \pi z/2L$. Then the equation for $\delta\theta(z,t) = \theta(z,t) - \pi z/2L$ takes the form

$$\gamma \frac{\partial\theta}{\partial t} + K \frac{\partial^2\theta}{\partial z^2}$$

$$= -\frac{\varepsilon_a}{16\pi} \left[(|E_x|^2 - |E_z|^2) \sin\frac{\pi z}{L} + (E_x E_z^* + E_x^* E_z) \cos\frac{\pi z}{L} \right] \tag{4.25}$$

The case of normal incidence is again of most interest. In the approximation of moderate anisotropy $|E_z| \approx 0$, $|E_x| \approx \text{const}$, and then

$$\delta\theta(z,t) = (1 - e^{-\Gamma t}) \frac{\varepsilon_a |E_x|^2}{16\pi K (\pi/L)^2} \sin\frac{\pi z}{L} \tag{4.26}$$

The correction to the e-wave effective dielectric permittivity is equal to $\delta\varepsilon_{ef} \approx \varepsilon_a \delta\theta(z,t)\sin(\pi z/L)$ and then integration gives the nonlinear phase shift

$$\delta\varphi = \frac{\omega}{cn} \frac{\varepsilon_a^2 L^3 |E|^2}{64\pi^3 K} \tag{4.27}$$

We recall that to the same accuracy $S(\text{erg/cm}^2 s) = cn|E|^2 8\pi$. Tilted cell was not yet explored experimentally for GON. Very recently GON was observed in a hybrid cell [103]; an important feature was the presence of GON for any direction of incidence of e − wave.

4.7 The influence of weak anchoring

So far we were considering a cell with the director strongly anchored on a boundary. Director reorientation effects and the GON become even stronger, if the anchoring is not strong. Thus, the possibility appears for studying the orienting influence of the interface between NLC and various media by nonlinear optics methods. Consider, for example, a planar cell, in which the director is strongly anchored on the boundary $z = 0$; $\mathbf{n}(z = 0) = \mathbf{e}_x$, and on the boundary $z = L$ the condition is given by

$$\frac{dn_z}{dz} + \frac{1}{R_a} n_z = 0 \tag{4.28}$$

Here R_a is the anchoring length. Boundary condition of the form (4.28) can be obtained, if a surface term $\int F_{sur} d^2S$ is added to the free energy, where $F_{sur}(\text{erg/cm}^2) = -0.5\,\sigma_a n_z^2$. The parameter σ_a can be called the surface tension coefficient anisotropy, and $R_a = K_1/\sigma_a$. The Euler-Lagrange linearized equation for the steady-state

$$K_1 d^2 n_z/dz^2 = -\varepsilon_a |E|^2 \sin\alpha\cos\alpha/8\pi \tag{4.29}$$

with the boundary condition (4.28) and with $n_z(z = 0) = 0$ has the solution

$$n_z(z) = -\frac{\varepsilon_a |E|^2 \sin\alpha\cos\alpha}{16\pi K_1}\left(z^2 - zL\,\frac{2 + \xi}{1 + \xi}\right) \tag{4.30}$$

where $\xi = L/R$. When $\xi \to \infty$ (strong anchoring on both boundaries) the solution

$$n_z(z) = -\frac{\varepsilon_a |E|^2 \sin\alpha \cos\alpha}{16\pi K_1} z(z - L) \tag{4.31}$$

identically coincides with the sum of the series (4.3) for $t \to \infty$. Note that the difference of the expression (4.31) from the first term of the series (4.3) is very small, ~3% in the cell centre. That was the reason why we neglected practically everywhere the higher harmonics $\sim\sin(m\pi z/L)$ with $m > 1$.

The director disturbance becomes greater for weak anchoring under the same conditions, see Figure 11. Correspondingly the phase shift increases:

$$\delta\varphi = \frac{\pi^4}{96} \frac{\omega}{cn} \frac{\varepsilon_a |E|^2 L^3 \sin^2\alpha \cos^2\alpha}{\pi^5 K_1} \left(1 + \frac{3}{1 + \xi}\right) \tag{4.32}$$

Expression (4.32) differs at $\xi \to \infty$ from expression (4.7) by the factor $\pi^4/96 \approx 1.01$ since in (4.7) we consciously omitted the higher har-

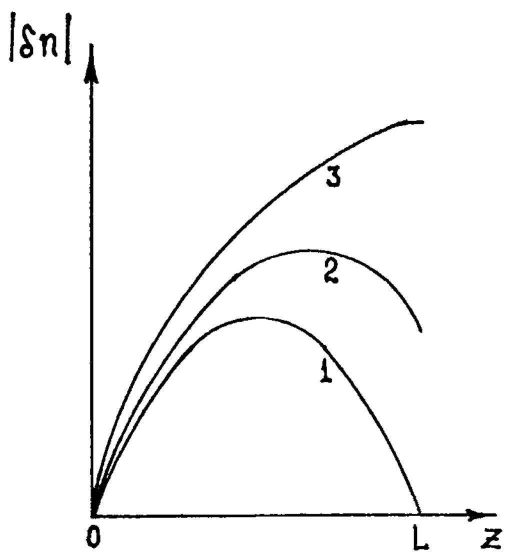


FIGURE 11 The director perturbations in the cell of thickness L for various values of the parameter ξ (in arbitrary units): 1) $\xi = 0$, 2) $\xi = 1$, 3) $\xi \to \infty$.

monics contributions. The phase shift (4.32) at $\xi = 0$ is 4 times larger than the phase shift for strong anchoring on both sides.

If the wave vector of light is out of the plane (x,z), i.e. if $\mathbf{k} = k(\mathbf{e}_z\cos\alpha + \mathbf{e}_x\sin\alpha\cos\beta + \mathbf{e}_y\sin\alpha\sin\beta)$, the extraordinary wave electric field has y-component and, according to equations (4.2), must also cause twist-deformation $\delta n_y \neq 0$.

Qualitatively new effect must arise due to the twist deformation in a planar cell with one free surface. Namely, here at $z = L$ the director turns in the plane (x,y) at an angle $\delta\psi = n_y(z = L)$, where

$$n_y(z) = \frac{\varepsilon_a|E|^2}{16\pi K_2} \cdot \sqrt{1 - \sin^2\alpha\cos^2\beta}\,(\sin^2\alpha\sin\beta\cos\beta)\, z(z - 2L) \qquad (4.33)$$

The transmitted wave polarization vector will be adiabatically turned together with the director. The turning angle itself is small in comparison with the nonlinear phase shift in the same conditions:

$$\frac{\delta\psi}{\delta\varphi} \approx 0.24\,\frac{n}{\varepsilon_a}\,\frac{K_1}{K_2}\,\frac{\lambda}{L}\,\frac{\sin\beta\cos\beta\sqrt{1 - \sin^2\alpha\cos^2\beta}}{\cos^2\alpha} \qquad (4.34)$$

It is important, however, that it is easy to register the rotation of polarization plane even when $|\delta\psi| \sim 10^{-3}$ rad. Besides, $\delta\psi$ measurement can have some advantages for a NLC with relatively small value of ε_a.

The above considered effect, $\delta\psi \sim |E|^2$, can be called the "nonlinear optical activity" (NOA). Right-left asymmetry here is introduced by the geometry of the vectors $\mathbf{e}_z$,$\mathbf{n}$ and $\mathbf{k}$ (vector $\mathbf{e}_z$ is directed from the "hard" to the "soft" wall). Let us write $\delta\psi$ in the form $\delta\psi = \text{ALP}$; then at $\alpha \sim 30°$, $\beta = 45°$, $\varepsilon_a = 0.5$, $L = 10^{-2}$ cm, for the constant A we get $A = 6.10^{-2}$ rad cm/W. As is known, the order of magnitude of the constant A for electron nonlinearity is $A = 10^{-13}$ rad cm/W.

If the surface tension anisotropy σ_a has another sign, then $\xi < 0$, i.e. the surface $z = L$ tries to orient the director not planarly, but homeotropically. However, up to $-1 < \xi$, the influence of the hard surface $z = 0$ supports the planar orientation along the whole thickness. Homogeneous planar orientation becomes unstable only when $\xi < -1$. This, by the way, one can also see from expression (4.29), which diverges when $\xi \to -1$.

The case of a hometropic cell with one soft boundary may be considered analogously. Boundary conditions then take the form

$$\left[\frac{dn_x}{dz} + \frac{1}{R_a} n_x\right]_{z=L} = 0, \qquad \left[\frac{dn_y}{dZ} + \frac{1}{R_a} n_y\right]_{z=L} \tag{4.35}$$

Introducing a parameter $\xi = L/R_a$, it is easy to obtain the same result (4.32) for the nonlinear phase shift by a simple substitution $K_1 \rightarrow K_3$.

There are a number of theoretical and experimental indications that the surface energy dependence on the director orientation is much more complicated than described by the Rapini potential

$$F_{\text{sur}} = -\frac{1}{2}\sigma_a(\mathbf{nn}^0)^2 = -\frac{1}{2}\sigma_a \sin^2(\theta - \theta_0) \tag{4.36}$$

which we used. Therefore, there is a special interest in surface orienting influence investigations by nonlinear optical methods, in particular, in the regime of large director deflection. There are reasons to expect for relatively high experimental accuracy due to the possibility of calibrating geometrical and power characteristics of a light beam on a cell with the same NLC, but with strong anchoring surfaces.

4.8 Twist cell (cholesteric with large pitch)

Consider the orientational nonlinearity for a planar twist cell. Let the director undisturbed equilibrium orientation have the form

$$\mathbf{n}^0(z) = \mathbf{e}_x \cos\theta_0(z) + \mathbf{e}_y \sin\theta_0(z), \qquad \theta_0(z) = qz \tag{4.37}$$

We can deal with a cholesteric liquid crystal (CLC) with a large pitch $h = 2\pi/q$ or (when $q \leqslant \pi/2L$) with an artificially twisted nematic. In the case of a large helix pitch, the polarization adiabatically follows the optical axis rotation. Therefore the torque applied to the director, turns out to be modulated in space as $\cos qz$. As a result, the deformation energy $\delta\mathbf{n}^2 K q^2$ turns out to be considerably larger than $\delta\mathbf{n}^2 K(\pi/L)^2$ for the GON in homogeneous cells; hence the nonlinearity becomes weaker. Let us give the final result for the nonlinear phase shift $\delta\varphi$ in our usual approximation $\varepsilon_a/\varepsilon_\perp << 1$.[16]

$$\delta\varphi = \frac{\omega}{cn}\frac{\varepsilon_a^2|E|^2}{8\pi q^2}\left(\frac{\sin^4\alpha}{16K_2} + \frac{\sin^2\alpha\cos^2\alpha}{K_1 + K_3}\right)\frac{L}{\cos\alpha} \tag{4.38}$$

The experiment[16] was carried out with a nematic 5CB mixed with 0.1 to 1 weight % of cholesteryl chloride. The helix pitch satisfied the condition $qL = m\pi$, where m is an integer. Argon laser radiation was used, $\lambda = 0.51\ \mu m$, with power up to 200 mW. It is convenient to introduce a parameter ζ characterizing the ratio of the nonlinear phase shift in a CLC-cell to the shift in a NLC-cell of identical thickness and for the same values of the power density, $K_i, n_\parallel, n_\perp$ and incidence angle α_{air}. Experimental dependence of the parameter ζ on the helix reciprocal pitch h^{-1} is given on Figure 12; the solid line is drawn according to the formulae (4.38), (4.7); very good agreement is observed.

However, for very small pitch, $h \leqslant \lambda/\varepsilon_a$, the regime of the Mauguin adiabatic following fails, and the polarization unit vector of the light field stays almost constant throughout the whole volume. At first sight one might suppose that the giant change of the orientation of the axis of CLC spiral is possible in that antiadiabatic regime, and that the corresponding constant $\varepsilon_2 \sim L^2\varepsilon_a^2/K$. However it was shown

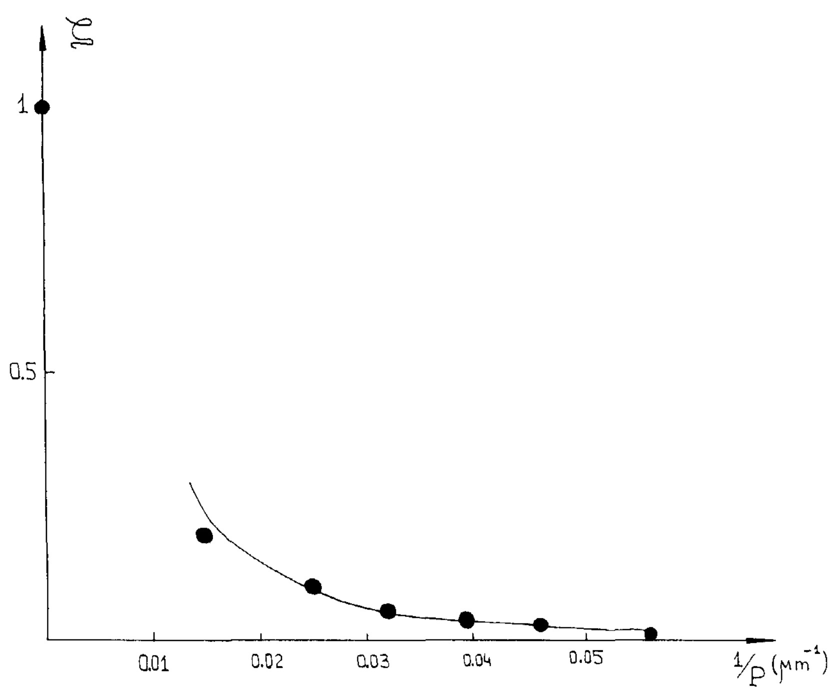


FIGURE 12 The plot of the parameter ζ versus the inverse pitch of the helix h^{-1} [16].

that for the Grandjean CLC cell with two or even with one hard boundary the optical nonlinearity is considerably smaller. The reason is that bending the axis of CLC results in change of the pitch and hence produces high elastic energy.

CLC orientational nonlinearity connected with the structure change within the period has been discussed above, see also Chapter 6. Such a nonlinearity is weaker than the GON for NLC by the factor h^2/L^2, where h is the helix pitch. An interesting problem—the GON in narrow beams ($a_\perp \lesssim L$) for CLC—requires separate consideration.

4.9 Smectics-C

Smectic phase of a liquid crystal (SLC) is similar to the nematic one with respect to the local orientation of the molecules. Therefore smectic may be considered approximately as an optically uniaxial medium. The free energy of interaction of SLC with the light field is identical to that of nematics. However, the existence of a periodical layered structure changes strongly the elastic properties of SLC in comparison with nematics. Namely, the change of interlayer period requires very high energy density. That is the reason why we shall discuss only such deformations of SLC which keep that period constant. We shall calculate how the light field excites such deformations and how the latter modify the propagation of light.

A possibility of such deformation is most obvious for smectics-C. If one takes the z-axis perpendicular to the flat smectic layers, then the director $\mathbf{n}$ can be represented as

$$\mathbf{n}(\mathbf{r}) = \mathbf{e}_z \cos\phi + (\mathbf{e}_x \cos\zeta(\mathbf{r}) + \mathbf{e}_y \sin\zeta(\mathbf{r}))\sin\phi \tag{4.39}$$

Here $\phi = \text{const} \neq 0$ is the angle between the director $\mathbf{n}$ and the layer's normal $\mathbf{e}_z$. The deformations (4.39) are characterized by the distribution of the azimuth $\zeta(\mathbf{r})$ of the director projection on the (x,y)-plane. SLC-C deformation free energy can be written in the form

$$F = \frac{1}{2}(B_1\cos^2\zeta + B_2\sin^2\zeta)\left(\frac{\partial\zeta}{\partial x}\right)^2 + \frac{1}{2}(B_1\sin^2\zeta + B_2\cos^2\zeta)\left(\frac{\partial\zeta}{\partial y}\right)^2 + \frac{1}{2}B_3\left(\frac{\partial\zeta}{\partial z}\right)^2 + B_{13}\frac{\partial\zeta}{\partial z}\left(\cos\zeta\frac{\partial\zeta}{\partial x} + \sin\zeta\frac{\partial\zeta}{\partial y}\right) \tag{4.40}$$

It is possible to verify that (4.40) is the most general expression invariant over rotations and reflections in the plane (x,y). Non-negativity of the deformation energy implies the condition $|B_{13}| \leq (B_1 B_3)^{1/2}$. For optically uniaxial SLC-C we have $\varepsilon_{ik}(r) = \varepsilon_\perp \delta_{ik} + \varepsilon_a n_i(\mathbf{r}) n_k(\mathbf{r})$ and the free energy of SLC interaction with a light field takes the form

$$-16\pi F_E = \varepsilon_\perp (EE^*) + \varepsilon_a \cos^2\phi |E_z|^2 + \varepsilon_a \sin\phi\cos\phi$$

$$\cdot [E_z^*(E_x \cos\zeta + E_y \sin\zeta) + E_z(E_x^* \cos\zeta + E_y^* \sin\zeta)]$$

$$+ \varepsilon_a \sin^2\phi [|E_x|^2 \cos^2\zeta + |E_y|^2 \sin^2\zeta$$

$$+ (E_x E_y^* + E_x^* E_y) \sin\zeta\cos\zeta] \qquad (4.41)$$

The dissipative function can be taken in the form

$$R = \frac{1}{2} \gamma_S (\partial\zeta/\partial t)^2 \qquad (4.42)$$

where γ_S is a constant of the same dimensions as the orientational viscosity of nematics γ; the order of magnitude is $\gamma_S \sim \gamma \sin^2\phi$.

For the GON discussion we shall consider the cell walls in the planes $z = 0$ and $z = L$, respectively. For a broad beam $a_\perp >> L$ and a definite type of polarization, the product $E_i E_k^*$ can be assumed to be constant throughout the cell volume, and then $\zeta(\mathbf{r},t)$ depends only on z and, during the establishment process, on time t.

We shall choose $\zeta_0(\mathbf{r}) \equiv 0$, i.e. the c-director along the x-axis as the undisturbed state. Linearized variational equations take the form

$$\gamma \frac{\partial\delta\zeta}{\partial t} - B_3 \frac{\partial^2\delta\zeta}{\partial z^2} = \frac{\varepsilon_a \sin\phi\cos\phi}{16\pi} (E_z E_y^* + E_z^* E_y)$$

$$+ \frac{\varepsilon_a \sin^2\phi}{16\pi} (E_x E_y^* + E_x^* E_y) \qquad (4.43)$$

An ordinary wave does not cause the c-director reorientation and to obtain GON it is necessary to use an extraordinary wave.

For moderate anisotropy, $\varepsilon_a/\varepsilon_\perp << 1$, the electric field vector of the e-type wave can be written in the form

$$\mathbf{E} = |\mathbf{E}| [\mathbf{k}[\mathbf{kn}]] / |[\mathbf{k}[\mathbf{kn}]]| \qquad (4.44)$$

where the wave vector **k** is $\mathbf{k} = k(\mathbf{e}_z \cos\alpha + \mathbf{e}_x \sin\alpha\cos\beta + \mathbf{e}_y \sin\alpha \sin\beta)$. The change of the angle ζ under the action of the light field in steady state, and for director strong anchoring on the boundaries $z = 0$ and $z = L$, equals

$$\zeta(z) = \frac{\varepsilon_a|E|^2}{16\pi B_3} z(z - L)\sin\alpha\sin\beta\sin\phi \cdot (\sin\alpha \sin\phi\cos\beta + \cos\alpha\cos\phi) \quad (4.45)$$

The relaxation constant $\Gamma(s^{-1})$ is $\Gamma = B_3\pi^2/\gamma_s L^2$. The nonlinear phase shift for $\zeta(\mathbf{r})$ from (4.45) and for $\varepsilon_a << \varepsilon_\perp$ is

$$\delta\varphi = \frac{\omega}{cn}\frac{\varepsilon_a^2|E|^2}{96\pi B_3} L^3\sin^2\alpha\sin^2\beta\sin^2\phi \cdot (\sin\alpha\sin\phi\cos\beta + \cos\alpha\cos\phi)^2 \quad (4.46)$$

To an accuracy of geometrical factors, the GON for SLC-C proves to be analogous to the GON of nematics. Note a characteristic property of the GON for a SLC-C cell with layers parallel to the walls. Namely, the GON vanishes not only for normal incidence, but also in the case where the wave vector **k** is in the plane (x,z) or (y,z). Note that at $\phi \rightarrow 0$, when the **n**-director approaches the direction $\mathbf{e}_z$ of the normal to the smectic layers, the nonlinear phase shift decreases as ϕ^2.

Numerical estimates give here the value of nonlinearity about an order of magnitude smaller than that for NLC, since the constants B_i have approximately the same values $B_i \sim 10^{-6} - 10^{-7}$ dyne, as K_i for NLC, and $\sin^2\phi \sim 0.1$. By analogy to the case of NLC, if an SLC-C cell with one free surface is used, the nonlinear optical activity i.e. light induced polarization plane rotation should take place.

Only one experiment on the orientational action of light on SLC-C has been carried out [17]; however no numerical results were obtained experimentally. Argon laser radiation was used with power density ~100 W/cm^2 and the freely suspended layer of smectic had unknown thickness.

The orientation of molecules in smectics-A (**n**-director) is parallel to the smectic layers ($\phi = 0$) and the degree of freedom corresponding to the *c*-director is absent for SLC-A. The director reorientation in SLC-A generally must be accompanied by layers deformation, and therefore the GON effect in SLC-A must be absent (compare with analogous statement for cholesterics in § 4.8).

5 LIGHT INDUCED FRIEDERICKSZ TRANSITION (LIFT)

The Friedericksz effect in static or radio-frequency fields (electric $\mathscr{E}$, or magnetic $\mathscr{H}$) is well known. Generally any reorientation of the director in the cell by external fields is called the Friedericksz effect, see e.g. [15], § 4.2. If the unperturbed director and the field are neither perpendicular nor parallel, then such a generalized Friedericksz effect arises even for weak field: $\delta\theta \sim \mathscr{E}^2$ or $\delta\theta \sim \mathscr{H}^2$. However, in a very important particular case, when the director of the unperturbed state corresponds to the maximum of the interaction energy with the field (for example, $\mathbf{n}^0 \parallel \mathscr{H}$ and $\chi_a < 0$), the reorientation arises beginning from some threshold value of the field.

As it has been shown in Chapters 1 and 4, the light field also reorients the LC director. This reorientation occurs already for weak fields $\delta\theta \sim |E|^2$, as in the static case, at oblique incidence of the light field; the effect is easily observed for an intensity ~50 W/cm^2.

A threshold effect of the influence of light on NLC has been observed experimentally in [13] by the use of much more powerful radiation (~10^3 W/cm^2). The effect is observed if the light beam is normally incident to an homeotropic oriented NLC cell, Figure 13. For such a great power density S, even moderate relative excess over the threshold, $S - S_{th} \sim 0.3\, S_{th} \sim 300$ W/cm^2 leads to a very strong

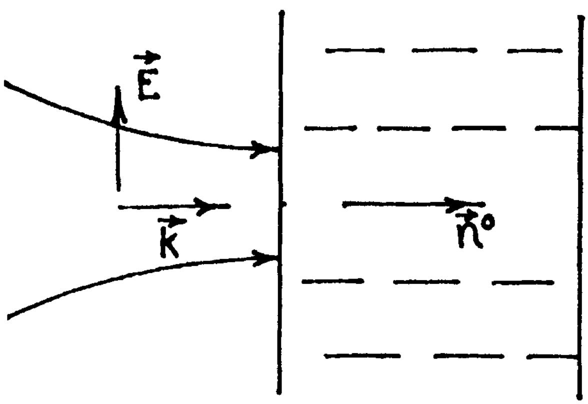


FIGURE 13 The geometry where the director reorientation has a threshold.

reorientation and, therefore, to a very large increase of the transmitted beam divergence. The effect observed in [13] was named "light induced Friedericksz transition" (LIFT). By tradition this name is used for designating just the threshold effect; the director reorientation linear in the weak field intensity is named "giant orientational nonlinearity" (GON). In the present Chapter we shall present at first the theoretical results with the purpose to use them further for the discussion of experiments.

5.1 Broad beams: threshold behaviour

The derivation of the threshold condition for the Friedericksz transition in a static electric field can be found in any textbook on liquid crystals. For homeotropic aligned cells, $\mathbf{n}^0 = \mathbf{e}_z$ and $\mathscr{E} = \mathbf{e}_z\mathscr{E}$ at $\varepsilon_a^0 < 0$, the threshold is determined by the relation $|\varepsilon_a^0|\mathscr{E}^2/4\pi = K_3(\pi/L)^2$. At first sight, for determination of the LIFT threshold in broad beams it would be enough to replace $|\varepsilon_a^0|\xi^2$ by 0.5 $\varepsilon_a EE^*$, where E is the complex amplitude of the light field, and ε_a is the optical anisotropy:

$$\frac{\varepsilon_a|E|^2_{\text{th}}}{8\pi} \stackrel{?}{=} K_3\left(\frac{\pi}{L}\right)^2 \tag{5.1}$$

This is correct to an order of magnitude, and corresponding numerical estimates give $|E|^2 \approx 2.5$ erg/cm^3 for $\varepsilon_a \approx 1$, $K_3 \sim 10^{-6}$ dyne, $L = 100$ μm, the light power density equals $cn|E|^2/8\pi \approx 500$ W/cm^2.

A more accurate theory must be based on the Euler-Lagrange-Rayleigh variational equations and on the Maxwell equation. In this section we shall consider the case of strictly normal incidence of the light of the polarization e_x to an homeotropic cell, Figure 13. For a broad beam we can confine ourselves to a discussion of the problem homogeneous in the plane (x,y). Then the field vector throughout the cell will remain, from symmetry considerations, in the plane (x,z) : $\mathbf{E}(z) = \mathbf{e}_xE_x(z) + \mathbf{e}_zE_z(z)$; z-component appeared because of the influence of nondiagonal components of the tensor $\varepsilon_{xz} = \varepsilon_{zx}$, arising when the director is deflected. The director distribution may be taken as

$$\mathbf{n}(z,t) = \mathbf{e}_z\cos\theta + \mathbf{e}_x\sin\theta; \qquad \theta = \theta(z,t) \tag{5.2}$$

Equation for θ has the form

$$\gamma \frac{\partial\theta}{\partial t} = (K_1 \sin^2\theta + K_3 \cos^2\theta) \frac{\partial^2\theta}{\partial z^2} - (K_3 - K_1) \sin\theta \cos\theta \left(\frac{\partial\theta}{\partial z}\right)^2$$

$$+ \frac{\varepsilon_a}{16\pi} [\sin 2\theta(|E_x|^2 - |E_z|^2) + \cos 2\theta(E_x E_z^* + E_x^* E_z)] \quad (5.3)$$

Eq. (5.3) and the corresponding Maxwell equations have an exact solution $\theta \equiv 0$, $\mathbf{E} = \mathbf{e}_x E \exp(i\omega n_\perp z/c)$. For the threshold determination we should linearize equation (5.3) in terms of the small deflection θ. It turns out to be necessary to take into account the field modification by the director disturbance even for the threshold calculation. Within the required accuracy this modification can be found from the equation $\mathrm{div}\mathbf{D} = \partial \mathbf{D}_z/\partial z = \partial(\varepsilon_{zx} E_x + \varepsilon_{zz} E_z)/\partial z = 0$. This equation gives $E_z = -\varepsilon_{zx} E_x/\varepsilon_{zz}$ so that in the linear in θ approximation $E_z \approx -\varepsilon_a \theta E_x/\varepsilon_\parallel$. The linearized problem is described by the equation

$$\gamma \frac{\partial\theta}{\partial t} = K_3 \frac{\partial^2\theta}{\partial z^2} + \frac{\varepsilon_a}{8\pi}\left(1 - \frac{\varepsilon_a}{\varepsilon_\parallel}\right)|E_x|^2\theta \quad (5.4)$$

The correction $\varepsilon_a/\varepsilon_\parallel$ in (5.4) corresponds to the light field z-component, arisen from the director disturbance; this correction is ~0.3 when $\varepsilon_a \approx 0.9$, $\varepsilon_\parallel = 3$, namely this correction changes the exact calculation of the LIFT threshold from the simple replacement (5.1). The solution of Eq. (5.4) is, with the boundary conditions $\theta(z = 0,t) = \theta(z = L,t) = 0$,

$$\theta(z,t) = \sum_{m=1}^{\infty} c_m \exp(-\Gamma_m t) \sin(m\pi z/L)$$

$$\Gamma_m = \gamma^{-1} K_3 \left(\frac{m\pi}{L}\right)^2 - \gamma^{-1}\frac{\varepsilon_a}{8\pi}\left(1 - \frac{\varepsilon_a}{\varepsilon_\parallel}\right)|E_x|^2 \quad (5.5)$$

The lowest value of the threshold intensity has the mode with $m = 1$, the threshold condition $\Gamma_1 = 0$ corresponds to

$$S_{\mathrm{th}}\left(\frac{\mathrm{erg}}{\mathrm{cm}^2 s}\right) = \frac{c n_\perp}{8\pi} |E|^2_{\mathrm{th}} = \frac{c\,\varepsilon_\parallel K_3}{\varepsilon_a \sqrt{\varepsilon_\perp}} \left(\frac{\pi}{L}\right)^2 \quad (5.6)$$

The disturbances' characteristic growth (or attenuation) time slows down near the threshold,

$$\exp(-\Gamma t) = \exp\left[-\frac{\pi^2}{\gamma L^2}\left(1 - \frac{S}{S_{th}}\right)t\right] \tag{5.7}$$

similar to FT in quasistatic fields.

5.2 The effects of polarization of the incident light

To determine the LIFT threshold in a beam with arbitrary polarization state we shall write the director disturbance in the form

$$\mathbf{n}(z,t) \approx \mathbf{e}_z + \mathbf{e}_x\theta_x(z,t) + \mathbf{e}_y\theta_y(z,t) \tag{5.8}$$

Besides, from the equation div**D** = 0 we have $E_z \approx -(\varepsilon_a/\varepsilon_\parallel)(\theta_x E_x + \theta_y E_y)$. As a result the equation for the director disturbance will take the form

$$\gamma\frac{\partial\theta_i}{\partial t} = K_3\frac{\partial^2\theta_i}{\partial z^2} + \frac{\varepsilon_a}{8\pi}\left(1 - \frac{\varepsilon_a}{\varepsilon_\parallel}\right)\frac{E_i E_K^* + E_i^* E_K}{2}\theta_K \tag{5.9}$$

where the indices i, k take two values (x,y). Therefore, the LIFT threshold is defined by the symmetrized matrix composed from x,y components of the incident field. It is convenient to represent the polarization matrix in the form (see, for example [19] or [20])

$$\begin{pmatrix}\langle E_x^* E_x\rangle & \langle E_x^* E_y\rangle \\ \langle E_y^* E_x\rangle & \langle E_y^* E_y\rangle\end{pmatrix} = \frac{1}{2}\langle(\mathbf{EE}^*)\rangle\begin{pmatrix}1+\xi_3 & \xi_1 - i\xi_2 \\ \xi_1 + i\xi_2 & 1-\xi_3\end{pmatrix} \tag{5.10}$$

where the angular brackets mean averaging (for partially-polarized light). The quantities ξ_1, ξ_2, ξ_3, are the components of the so called normalized Stokes vector. Let us introduce the eigenvalues $I_{1,2} = 0.5\,\langle(\mathbf{EE}^*)\rangle\,[1 \pm (\xi_1^2 + \xi_3^2)^{1/2}]$ of the symmetric part of the matrix (5.10) i.e. values satisfying the equation

$$\frac{1}{2}\langle E_i^* E_k + E_i E_k^*\rangle\theta_k = I\theta_i \tag{5.11}$$

and let e_1 and e_2 be eigenvectors of the matrix (5.11). Then the solution of Eq. (5.9) has the form

$$\theta(z,t) = \sum_{m=1}^{\infty} \sin\frac{m\pi z}{L}\,[e_1 c_{1m}\exp(-\Gamma_{1m}t) + e_2 c_{2m}\exp(-\Gamma_{2m}t)],$$

$$\Gamma_{(1,2)m} = \gamma^{-1}\left(\frac{K_3 m^2\pi^2}{L^2} - \frac{\varepsilon_a}{8\pi}\frac{\varepsilon_\perp}{\varepsilon_\parallel} I_{1,2}\right) \tag{5.12}$$

The quantity I_1 corresponds to the intensity of the strongest component of the linear polarized field. The LIFT threshold corresponds to the condition $\Gamma = 0$ so that

$$S_{\text{th}}\left(\frac{\text{erg}}{\text{cm}^2\text{s}}\right) = \frac{c\,\varepsilon_\perp^{1/2}\langle(\mathbf{E}\mathbf{E}^*)\rangle_{\text{th}}}{8\pi} = \frac{c\pi^2\varepsilon_\parallel K_3}{\varepsilon_a\varepsilon_\perp^{1/2}L^2}\,\frac{2}{1 + (\xi_1^2 + \xi_3^2)^{1/2}} \tag{5.13}$$

If the radiation is completely depolarized ($\xi_1 = \xi_2 = \xi_3 = 0$), or if it is purely circularly polarized ($\xi_1 = \xi_3 = 0$, $|\xi_2| = 1$), the threshold turns out to be twice that for linear polarization. In the case $\xi_1 = \xi_3 = 0$ the eigenvectors e_1 and e_2 are arbitrary and, therefore, the disturbance direction in the established state above threshold depends on some subtle effects (for example, on transverse distribution of intensity in the beam).

5.3 Geometrical optics of an inhomogeneous nematic

The determination of the steady-state structure above threshold requires the knowledge of the exact solution of the Maxwell equation (3.9) in an inhomogeneous medium. The field $\mathbf{E}(\mathbf{r})$ we shall represent in the form

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}^{(0)}(\mathbf{r})\exp\left(i\frac{\omega}{c}\psi\right) \tag{5.14}$$

It is convenient to introduce the vector

$$\mathbf{P} = \nabla\psi \tag{5.15}$$

which coincides with wave vector **k** divided by ω/c for a homogeneous medium, i.e. the length of the vector **P** is equal to the "phase refractive index." In the zeroth approximation in c/ω it follows then from (3.9) that

$$(P_i P_k - P^2\delta_{ik} + \xi_{ik}(r))E_k^{(0)}(\mathbf{r}) = 0 \tag{5.16}$$

Equating to zero the determinant of the linear homogeneous system (5.16) we get the known Fresnel equation for $\mathbf{P}(\mathbf{r})$ for the local value of the tensor $\varepsilon_{ik}(\mathbf{r})$. We are interested in the case of a nematic in which the director lies in the (x,z) plane,

$$n_x = \sin\theta(z), \qquad n_y = 0, \qquad n_z = \cos\theta(z) \tag{5.17}$$

and the inclination angle $\theta(z)$ does not depend on the coordinates x and y; the z axis is chosen here normal to the planes of the cell. The Fresnel equation has the form

$$\left(\frac{P^2}{\varepsilon_\perp} - 1\right)\left[\frac{(P_x\sin\theta + P_z\cos\theta)^2}{\varepsilon_\perp} + \frac{(P_x\cos\theta - P_z\sin\theta)^2}{\varepsilon_\parallel} - 1\right] = 0 \tag{5.18}$$

We are interested in the second root of this equation, namely the one corresponding to the extraordinary wave. Assume that an e-polarized wave is incident on the cell from the air at an angle α_{air} to the normal in the (x,z) plane. By virtue of the translational invariance of the problem to displacements along the x axis we can seek a solution inside the medium of the form

$$\psi(x,z) = sx + \psi_1(z), \qquad s = \sin\alpha_{\text{air}} \tag{5.19}$$

It follows then from the Fresnel equation (5.18) that

$$\psi_1(z) = \int_0^z P_z(z')dz',$$

$$P_z(z) = \frac{(\varepsilon_\parallel\varepsilon_\perp)^{1/2}(\varepsilon_\perp - s^2 + \varepsilon_a\cos^2\theta)^{1/2} - s\varepsilon_a\sin\theta\cos\theta}{\varepsilon_\perp + \varepsilon_a\cos^2\theta} \tag{5.20a}$$

where $\theta = \theta(z)$. The direction of the field vector $\mathbf{E}(\mathbf{r})$ is determined from equation (5.16):

$$\frac{E_z}{E_x} = \frac{SP_z(\theta) + \varepsilon_a \sin\theta\cos\theta}{S^2 - \varepsilon_\perp - \varepsilon_a\cos^2\theta} \tag{5.20b}$$

The Poynting vector $\mathbf{S} = c[\mathbf{EH}^*]/8\pi$ can be expressed in terms of the vector $\mathbf{P}$ with the aid of the equation $\mathbf{H} = [\mathbf{PE}]$, this yields

$$\mathbf{S} = \frac{c}{8\pi}[\mathbf{P}(\mathbf{EE}^*) - \mathbf{E}^*(\mathbf{PE})] \tag{5.21}$$

In the problem with $\hat{\varepsilon} = \hat{\varepsilon}(z)$ for a propagating plane wave, the Poynting vector $\mathbf{S}(z)$ is itself far from constant. This statement can be verified in a trivial fashion with a scalar medium having $\varepsilon = \varepsilon(z)$ as the example. In the absence of absorption, however $div\mathbf{S} = 0$, and for the problem homogeneous in x,y it follows therefore that $S_z(z) = \text{const}$. It is easy to verify that a field in the form

$$E_x(x,z) = A(\varepsilon_\perp - s^2 + \varepsilon_a\cos^2\theta)^{1/4}\exp\left[i\frac{\omega}{c}(sx + \psi_1(z))\right] \tag{5.22}$$

$$E_z(x,z) = -A\,\frac{s(\varepsilon_\|\varepsilon_\perp)^{1/2} + \varepsilon_a\sin\theta\cos\theta(\varepsilon_\perp - s^2 + \varepsilon_a\cos^2\theta)^{1.2}}{\exp\left[i\frac{\omega}{c}(sx + \psi_1(z))\right]} \tag{5.23}$$

is the solution, of interest to us, of Maxwell's equations in the geometrical-optics approximation. In this case A is constant and

$$S_z\left(\frac{\text{erg}}{\text{cm}^2 s}\right) = \frac{c(\varepsilon_\|\varepsilon_\perp)^{1/2}}{8\pi}|A|^2 \tag{5.24}$$

5.4 Broad beams: above-threshold structure

The equation for the above-threshold steady state structure can be obtained by substitution of E_x and E_z from (5.22), (5.23) into equation (5.3):

$$(K_1\sin^2\theta + K_3\cos^2\theta)\frac{d^2\theta}{dz^2} - (K_3 - K_1)\sin\theta\cos\theta\left(\frac{d\theta}{dz}\right)^2 + \frac{\varepsilon_a\varepsilon_\parallel\varepsilon_\perp |A|^2\sin\theta\cos\theta}{8\pi(\varepsilon_\perp + \varepsilon_a\cos^2\theta)^{3/2}} = 0 \quad (5.25)$$

This equation has an exact integral, which is easy to obtain by multiplying (5.25) by $2d\theta/dz$ and integrating over dz. The integration constant can be expressed in terms of the maximum declination angle θ_m, so that

$$\left(\frac{d\theta}{dz}\right)^2 = \frac{\varepsilon_\perp^{1/2}S[1 - (\varepsilon_a/\varepsilon_\parallel)\sin^2\theta]^{1/2} - [1 - (\varepsilon_a/\varepsilon_\parallel)\sin^2\theta_m]^{1/2}}{cK_3(1 - \tilde{K}\sin^2\theta)[1 - (\varepsilon_a/\varepsilon_\parallel)\sin^2\theta]^{1/2}[1 - (\varepsilon_a/\varepsilon_\parallel)\sin^2\theta_m]^{1/2}} \quad (5.26)$$

where $\tilde{K} = (K_3 - K_1)/K_3$.

The existence of such an integral is not surprising; it follows from E. Noether's theorem. The Frank equations for the director are not invariant with respect to displacement of the z-coordinate, since the force $\sim EE^*$ is z-dependent. The Maxwell equations are, in their turn, not invariant with respect to the z-displacement, since $\hat{\varepsilon} = \hat{\varepsilon}(z)$. However, the combined system "nematic + electromagnetic field" has the free energy (3.3), (3.10) which does not depend on z explicitly. Therefore according to E. Noether, the zz component of the momentum flow is conserved. An equation for $\theta(z)$ can be obtained from (5.26):

$$\int_0^{\theta(z)} d\varphi \left\{\frac{(1 - K\sin^2\varphi)[1 - (\varepsilon_a/\varepsilon_\parallel)\sin^2\varphi]^{1/2}[1 - (\varepsilon_a/\varepsilon_\parallel)\sin^2\theta_m]^{1/2}}{[1 - (\varepsilon_a/\varepsilon_\parallel)\sin^2\varphi]^{1/2} - [1 - (\varepsilon_a/\varepsilon_\parallel)\sin^2\theta_m]^{1/2}}\right\}^{1/2} = \frac{z}{2}\left(\frac{\varepsilon_\perp^{1/2}S}{cK_3}\right)^{1/2} \quad (5.27)$$

Taking into account that the maximum angle θ_m is achieved in the cell center, we should put $\theta(z) = \theta_m$ and $z = L/2$ in (5.27), as a result of which an implicit equation for θ_m is obtained. At $z = L/2$ Eq. (5.27) and its analog can be solved generally only numerically. A qualitative analysis is more conveniently performed with the aid of the original equation (5.25).

Considering a small excess over the threshold the problem can be limited to terms not higher than the third degree in θ. Writing down then the solution satisfying the boundary conditions in the form

$$\theta(z) = \theta_1 \sin(\pi z/L) + \theta_3 \sin(3\pi z/L) + \cdots$$

we get from (5.25)

$$\theta_1 = \pm\left(2\,\frac{S - S_{th}}{S_{th}}\right)^{1/2} V^{-1/2}, \tag{5.28}$$

$$V = 1 - \frac{9}{4}\frac{\varepsilon_a}{\varepsilon_\parallel} - \frac{K_3 - K_1}{K_3}, \tag{5.29}$$

$$\theta_3 = \frac{1}{48}\left(1 - \frac{9}{4}\frac{\varepsilon_a}{\varepsilon_\parallel} - 3\,\frac{K_3 - K_1}{K_3}\right) \tag{5.30}$$

An important conclusion follows from (5.28): the magnitude of $\theta_1 = \theta(z = L/2)$ increases very fast ($\sim\sqrt{S - S_{th}}$) when the power exceeds the threshold value. For MBBA, for example, we have $V = 0.28$ so that even for $(S - S_{th})/S_{th} \approx 4\%$ from (5.28) we get $\theta_1 \approx 0.5$. rad $\approx 30°$; for OCBP $V = 0.24$. In the same approximation the nonlinear phase shift is equal to

$$\delta\varphi = \frac{\omega}{2c} L \frac{\sqrt{\varepsilon_\perp}\,\varepsilon_a}{\varepsilon_\parallel V} \frac{S - S_{th}}{S_{th}} \tag{5.31}$$

Numerically small value of V characterizes the system compliance to above-threshold influences $\theta_1 \sim [(S - S_{th})/V]^{1/2}$. This parameter is unity in the LIFT simplification, i.e. in the single-constant approximation and without taking into account the influence of the director reorientation on the wave ($\varepsilon_a \to 0$).

It is very important that the value of the parameter V turns out to be negative for some NLC. For example, $V = -0.12$ for PAA at $T = 125°C$. In this case the relation (5.28) has no field of application at all. On the basis of the more exact solution of equation (5.27) it is possible to show that the LIFT in broad beam for NLC with $V < 0$ must have a hysteresis. The transition will occur for a power density S determined by (5.6) if the intensity increases from zero, Figure 14; θ_m increases abruptly. The transition to the state $\theta = 0$ will also occur abruptly, but at some lower power value, when the power is decreased.

5.5 Effects of finite beam sizes

The Frank energy for a beam with finite transverse sizes has an additional term $\delta F \sim K/a^2$, where a is the disturbance transverse size. Therefore, a rough estimate of the LIFT threshold has the form

$$|E|^2_{\text{th}} \geqslant \frac{8\pi K}{\varepsilon_a}\left(\frac{\pi^2}{L^2} + \frac{1}{a^2}\right) \tag{5.32}$$

As for the GON, the form of the exact solution turns out to be essentially dependent on whether the problem is one- or two-dimensional in the transverse coordinates. Namely, if the beam size is small, $a_\perp << L$, the disturbance size a in a one-dimensional problem turns out to be $a \sim \sqrt{a_\perp L}$, so that $|E|^2_{\text{th}} \sim K/a_\perp L$. Unlike this, the estimate

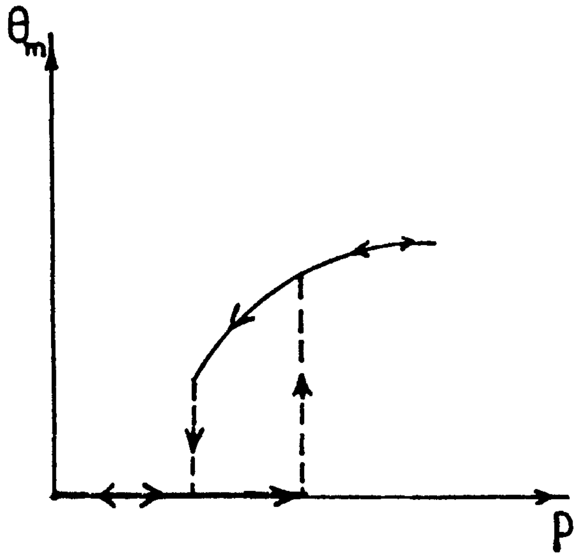


FIGURE 14 The hysteresis of LIFT. The arrows indicate the direction of the light power change.

of the form (5.32) in the two dimensional problem is valid for $a = a_\perp$ to a logarithmic accuracy.

For the LIFT threshold determination the field components can be taken as in the undisturbed wave, E_z can be expressed in terms of E_x, E_y and θ_x, θ_y, so that in linear approximation in θ_x, θ_y the Euler-Lagrange-Rayleigh equations yield

$$\gamma \frac{\partial \theta_i}{\partial t} = K_3 \frac{\partial^2 \theta_i}{\partial z^2} + K_2 \frac{\partial^2 \theta_i}{\partial x_k \partial x_k} + (K_1 - K_2) \frac{\partial^2 \theta_K}{\partial x_i \partial x_k} + \frac{\varepsilon_a \varepsilon_\perp}{8\pi \varepsilon_\parallel} I_{ik}(\mathbf{r}) \theta_k(\mathbf{r}, z, t) \quad (5.33)$$

Here $I_{iK} = 0.5(E_i E_k^* + E_i^* E_k)$, and we have separated explicitly the transverse ($\mathbf{r}$) and longitudinal (z) coordinates; $\partial/\partial x_i$ denotes differentiation with respect to the transverse coordinates.

Eq. (5.33) is analogous to the Schrödinger equation with the difference that the wave function is a two-component vector, and the operators of the kinetic and potential energy have a more complicated tensor form, and is very complicated in the general case. Therefore, the cases for which it is possible to find the exact solution of the problem of the LIFT threshold and unstable mode profile, are of special interest. For all these cases the perturbations remain purely planar by virtue of the symmetry properties. Let us enumerate them in an arbitrary order.

Let the intensity of the light beam depend only on one transverse coordinate, which we denote by y (ribbon beam). In the case $\mathbf{E}(y) = \mathbf{e}_x \sqrt{I(y)}$ the nonzero perturbation will be

$$\delta \mathbf{n} \sim \mathbf{e}_x \, \theta(y) \sin(\pi z/L)$$

and in the case $\mathbf{E}(y) = \mathbf{e}_y \sqrt{I(y)}$

$$\delta \mathbf{n} \sim \mathbf{e}_y \, \theta(y) \sin(\pi z/L)$$

For both cases, Eq. (5.33) takes the form

$$\left[\gamma\Gamma - K_3 \left(\frac{\pi}{L}\right)^2 + \tilde{K} \frac{\partial^2}{\partial y^2} + \frac{\varepsilon_a}{8\pi} \frac{\varepsilon_\perp}{\varepsilon_\parallel} I(y)\right] \theta(y) = 0 \quad (5.34)$$

where $\tilde{K} = K_2$ for $\mathbf{E} \sim \mathbf{e}_x$ and $\tilde{K} = K_1$ for $\mathbf{E} \sim \mathbf{e}_y$. We now obtain the threshold intensity $I(y = 0)$ and the perturbation profile $\theta(y)$ for a number of specific $I(y)$ distributions.

1) Let $I(y) = I_0$ at $-a \leqslant y \leqslant a$ and $I(y) = 0$ at $|y| > a$ (rectangular profile with total width $2a$). The threshold condition can then be written in the form

$$\kappa = (p^2 - \kappa^2)^{1/2}\tan(p^2 - \kappa^2)^{1/2}a \ ,$$

$$\kappa = \frac{\pi}{L}\left(\frac{K_3}{\tilde{K}}\right)^{1/2}, \qquad p^2 = \varepsilon_a\varepsilon_\perp I_0/8\pi\varepsilon_\| \tilde{K} \tag{5.35}$$

we recall that $S(\mathrm{erg/cm^2 s}) = c\sqrt{\varepsilon_\perp}I_0/8\pi$.

Since $K_2 < K_1$ the threshold for polarization along the ribbon (e_x) is somewhat lower than for e_y polarization.

At $a >> L$ and $a << L$ we have the asymptotic expressions

$$I_0 = \left(\frac{\pi}{L}\right)^2 \frac{8\pi\varepsilon_\| K_3}{\varepsilon_a\varepsilon_\perp}\left[1 + \frac{\tilde{K}}{K_3}\left(\frac{L}{2a}\right)^2\right], \qquad a >> L \tag{5.36a}$$

$$I_0 = \frac{8\pi^2\varepsilon_\|(\tilde{K}K_3)^{1/2}}{\varepsilon_a\varepsilon_\perp aL}\left[1 + \left(\frac{K_3}{\tilde{K}}\right)^{1/2}\frac{\pi a}{L}\right], \qquad a << L \tag{5.36b}$$

Let us emphasize that in a narrow ribbon beam, in the limit $a << L$, the threshold value is the specific power density

$$\int_{-\infty}^{\infty} S_z(y)dy = \frac{2c\sqrt{\varepsilon_\perp}\,aI_0}{8\pi} = \frac{2\pi c\varepsilon_\|(K_3\tilde{K})^{1/2}}{\varepsilon_a\ \varepsilon_\perp^{1/2}L} \tag{5.37}$$

At $a << L$ the last statement does not depend at all on the concrete form of $I(y)$. In broad beams, the correction to I_{th} turns out to be quadratic in the small parameter L/a; this statement is specifically applicable to a flat-top type of distribution. The unstable mode is of the form

$$\theta(y) = \begin{cases} \cos(p^2 - \kappa^2)^{1/2}y \ ; & |y| \leqslant a \\ \cos[(p^2 - \kappa^2)^{1/2}a]\exp[-\kappa(|y| - a)] \ , & |y| > a \end{cases} \tag{5.38a}$$

In particular, at $a >> L$

$$\theta(y) = \begin{cases} \cos(\pi y/2a) \ , & |y| \leqslant a \\ (\pi/2\kappa a)\exp[-\kappa(|y| - a)] \ , & |y| > a \end{cases} \tag{5.38b}$$

A characteristic feature of (5.38b) is that for a rectangular intensity distribution the perturbation $\theta(y)$ has a cosine distribution and falls off practically to zero (more accurately to the small quantity $\pi/2\mathcal{H}a$) at the beam boundaries $y = \pm a$. For $a >> L$, even at a relatively small excess over threshold $(S - S_{th})/S_{th} \sim (\kappa a)^{-2}$, modes with higher transverse indices are added to the lower mode (5.38b). As a result, the perturbation inside the illuminated region tends to a constant value typical of an unbounded beam.

2) Let $I(y) = I_0 \cosh(y/b)$, with the beam half-width at half-maximum intensity a(HWHM) $\approx 0.9b$. The threshold is then determined by the equation

$$I_0 = \left(\frac{\pi}{L}\right)^2 \frac{8\pi\varepsilon_\| K_3}{\varepsilon_a \varepsilon_\perp}\left[1 + \left(\frac{\tilde{K}}{K_3}\right)^{1/2} \frac{L}{\pi b}\right] \tag{5.39}$$

and the unstable mode has the form

$$\theta(y) = \cosh^{-\beta}(y/b) ,$$

$$\beta = \frac{1}{2}\left\{-1 + \left[1 + \left(\frac{2\pi b}{L}\right)^2 \frac{K_3}{\tilde{K}}\left(1 + \left(\frac{\tilde{K}}{K_3}\right)^{1/2} \frac{L}{\pi b}\right)\right]^{1/2}\right\} \tag{5.40}$$

At $a << L$ everything coincides with the case of a square top provided the integrals $\int I(y)dy$ coincide. At $b >> L$, the correction to the threshold is linear in L/b, and the unstable mode profile has the form

$$\theta(y) \approx \exp(-y^2/a^2), \qquad a = (2b/\kappa)^{1/2} \tag{5.41}$$

Both properties, the Gaussian profile of the mode with width $a \sim (Lb)^{1/2}$ and the correction to the threshold in first order in $L/b << 1$ are typical of any broad distribution $I(y)$ with a smooth dependence near the origin, $I(y) \approx I_0(1 - y^2/b^2)$. For broad distributions that are smooth at zero it is possible to solve in the single constant approximation also the two-dimensional problem with $I(x,y)$ in the form

$$I(x,y) = I_0\left(1 - \frac{x^2}{b_x^2} - \frac{y^2}{b_y^2}\right) \tag{5.42}$$

(the higher terms in x^2, y^2 have been left out of (5.42)) for polarization $\mathbf{E} \sim \mathbf{e}_x$.

The threshold is determined here by the expression

$$I_0 = \frac{8\pi\varepsilon_{\|}K}{\varepsilon_a\varepsilon_\perp}\left(\frac{\pi}{L}\right)^2\left[1 + \frac{L}{\pi}\left(\frac{1}{b_x} + \frac{1}{b_y}\right) + 0\left(\left(\frac{L}{b}\right)^2\right)\right] \quad (5.43)$$

(we recall that by assumption b_x, $b_y >> L$), and the perturbation has the form

$$\boldsymbol{\theta} = \mathbf{e}_x \exp\left(-\frac{x^2}{a_x^2} - \frac{y^2}{a_y^2}\right),$$

$$a_x = \left(\frac{2b_xL}{\pi}\right)^{1/2}, \qquad a_y = \left(\frac{2b_yL}{\pi}\right)^{1/2} \quad (5.44)$$

Note, that at $b >> L$ the half-width of the unstable mode from (5.40) at the 1/2 level of θ in maximum is proportional to the geometric mean of the beam width b and the cell thickness L:

$$\Delta x(\mathrm{HWHM}\theta) = (2bL/\pi)^{1/2} \quad (5.45)$$

We present also the equation for the threshold in the case of a two-dimensional round flat top in the single-constant approximation:

$$I(\mathbf{r}) = I_0\begin{cases} 1, & |\mathbf{r}| \leq a \\ 0, & |\mathbf{r}| > a \end{cases} \quad (5.46)$$

The profile $\theta(r)$ is expressed in terms of the Bessel functions. At $a >> L$ we have, just for a one-dimensional flat top a correction of order

$$I_0 = \left(\frac{\pi}{L}\right)^2\frac{8\pi\varepsilon_{\|}K}{\varepsilon_a\varepsilon_\perp}\left\{1 + 0.58\left(\frac{L}{a}\right)^2\left[1 - 0.76\frac{L}{a}\frac{K_0(\pi a/L)}{K_1(\pi a/L)}\right]^2\right\} \quad (5.47a)$$

where K_ν is a modified Bessel function of order ν. At $a << L$ we have

$$I_0 = \frac{16\pi\varepsilon_{\|}K}{\varepsilon_a\varepsilon_\perp a^2}\left[1 + \frac{1}{2}\left(\frac{\pi a}{L}\right)^2\ln\left(\frac{L}{\pi a}\right)\right] \quad (5.47b)$$

Thus, accurate to logarithmic corrections, the threshold value is the total beam power $I_0 a^2$. Unfortunately, we do not know as yet whether this conclusion is valid for narrow beams ($a << L$) without assumption of the single-constant approximation.

5.6 LIFT experimental investigations

Light induced Friedericksz transition (LIFT) has been observed in a homeotropic cell filled by OCBP [13]. It was illuminated by a single transverse mode argon laser radiation, $\lambda = 0.5145$ μm. The power was up to 0.2 W, waist radius in the lens focus 5.10^{-3} cm, power density was up to 2.10^3 W/cm^2. If the light wave is incident normally to the cell, its wave vector is parallel to the unperturbed optical axis and, for small intensities, self-focusing was absent. However, beginning from the intensity of order 70–80 mW (for the cell of 150 μm thickness), after long enough time, from 10 s to 3 min, a very strong angular divergence ~10° of the transmitted beam appeared, accompanied by the characteristic picture of a large number ($N \sim 20$–50) of aberrational fringes.

In the paper [21], LIFT for a circularly-polarized beam has been registered. The threshold power turned out to be approximately twice the value for the linear-polarized radiation; in good agreement with the theory.

The threshold power density S(W/cm^2) for broad beams must decrease as L^{-2} when the cell thickness increases. This statement has not been verified quantitatively, however, the absolute value of the experimentally measured threshold power [22] agrees well with the theory.

The LIFT threshold dependence on temperature, according to the theory, is determined mainly by the factor $S_{\text{th}} \sim \varepsilon_a^{-1} K_3$. Since K_3 decreases as the square of the order parameter, $K_3 \sim Q^2$, and $\varepsilon_a \sim Q^1$ when T approaches T_c, then $S_{\text{th}} \sim Q^1$ and must also decrease with the decrease of Q when the temperature increases. In the experiment [22] on the LIFT on OCBP-cell, a decrease of the threshold was observed from $S = 51$ mW to $S = 24.5$ mW when the temperature changed from 34°C to 39.4°C ($\lambda = 0.515$ μm, the cell-thickness 150 μm, beam transverse radius 18.6 μm in the waist). In the same paper LIFT was observed by the use of a He-Ne laser radiation ($\lambda = 0.628$ μm) and two lines of argon laser ($\lambda = 0.476$ μm and $\lambda = 0.515$ μm).

The transmitted light polarization above LIFT threshold turns out to be inhomogeneous depending on the observation angle even for a linear-polarized incident beam. There are two reasons for the polarization deflection from the initial direction. Both reasons are based on the adiabatic following of the extraordinary wave polarization vector.

In the 1-st explanation one should take into account that for $K_2 \neq K_1$ the director disturbances have non-planar character, $\mathbf{n} = \mathbf{e}_z + \mathbf{e}_x\delta n_x + \mathbf{e}_y\delta n_y$ and $\delta n_y \neq 0$ even for the incident light with x-polarization. The order of magnitude of δn_y can be estimated by perturbation theory with the parameter $(K_1 - K_2)/K_3$ (not very small in fact). In this mechanism

$$\delta n_y \approx \frac{K_1 - K_2}{K_3}\left(\frac{L}{\pi}\right)^2 \frac{\partial^2 \delta n_x}{\partial x \partial y} \sim \frac{K_1 - K_2}{K_3}\,\delta n_y \frac{xy}{a^4}\left(\frac{L}{\pi}\right)^2 \tag{5.48}$$

for beams of width $a_\perp \gg L$. It is important that the polarization deflection from the initial direction $\mathbf{e}_x$ is absent both at $x = 0$ and at $y = 0$; thus polarization rotation in the transmitted beam by this mechanism must be only in the internal sides of quadrants in coordinates (x,y) but not on the "cross" $x = 0$ and $y = 0$, Figure 15.

The other mechanism is based essentially on the circumstance that inside the medium the beam suffers considerable self-focusing deflection ($\sim$10°–20°). As a result of $\mathbf{n}$ vector change, the polarization vector must be changed. In directions of the beam symmetry plane $\mathbf{k}_{\text{out}} = k(\mathbf{e}_z\cos\beta + \mathbf{e}_x\sin\beta)$ the polarization is not changed. Complicated enough geometrical constructions led to the conclusion that polarization plane rotation arises in the other directions symmetric over the plane (x,z). The effects of non-adiabatic propagation lead to ellypticity of polarization of output waves at some directions of deflection. Theoretical details are in the original papers [23,24]; the transmitted beam polarization rotation and ellypticity for LIFT experimentally have been carried out in the papers [13,14]. For a small excess over the threshold (beams small deflections) the depolarization picture corresponds to the first mechanism, see experiment in [13] and for a greater divergence (considerable excess over the threshold) there is good agreement of the experimental results of [24] with the second mechanism.

Very interesting is the problem of the behaviour of the director perturbations out of the beam (compare also with § 4.2). For its discussion it is convenient to use the linearized equation (5.33). In the region out of the beam $I_{ik} = 0$ and we seek the solution in the form θ_x, $\theta_y \sim \sin(\pi z/L)$. Then for the functions $\theta_x(x,y)$, $\theta_y(x,y)$ the following equation is obtained

$$\left[K_2 \frac{\partial^2}{\partial x_j \partial x_j}\delta_{ik} + (K_1 - K_2)\frac{\partial^2}{\partial x_i \partial x_k} + K_3\left(\frac{\pi}{L}\right)^2\right]\theta_k(x,y) = 0 \tag{5.49}$$

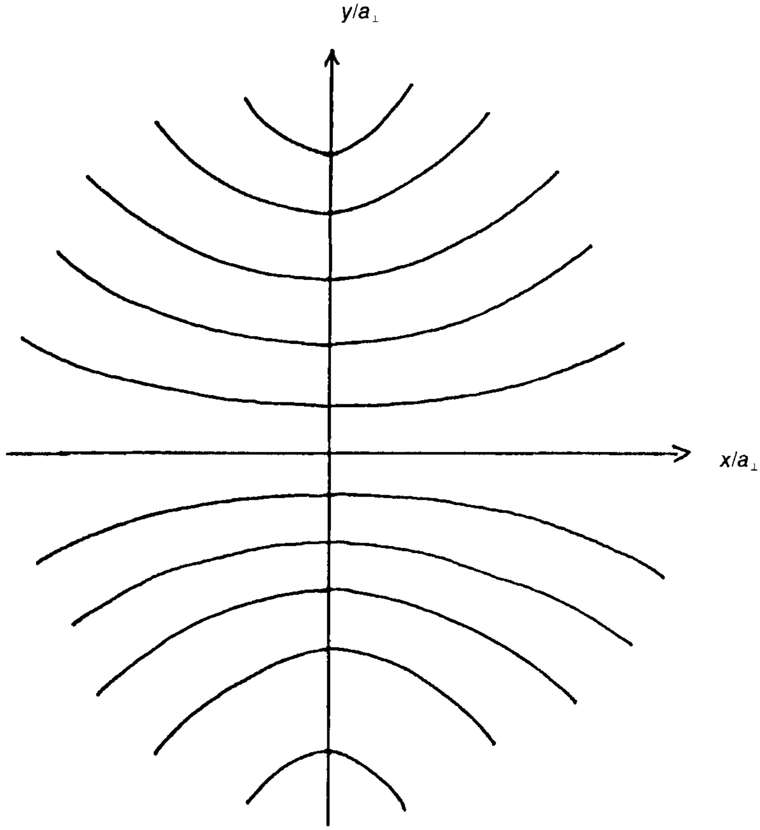


FIGURE 15 The spatial distribution of the director perturbation $\delta\mathbf{n} \sim (\mathbf{e}_x + \text{const}\,\mathbf{e}_y xy/a_\perp^4)\exp[-(x^2+y^2)/a_\perp^2]$ near the origin of the coordinate frame $x = 0$, $y = 0$. The tangents to the curves indicate the direction of δn.

Far from the disturbance source this equation has exponentially decreasing solutions

$$\delta\mathbf{n}(\mathbf{r},z) = \sin\frac{\pi z}{L}\left\{c_1\boldsymbol{\nu}\exp[-\kappa_1(\boldsymbol{\nu}\mathbf{r})] + c_2[\mathbf{e}_z\boldsymbol{\nu}]\exp[-\kappa_2(\boldsymbol{\nu}\mathbf{r})]\right\}, \tag{5.50a}$$

$$\boldsymbol{\nu}^2 = 1, \qquad \kappa_1 = (\pi/L)(K_3/K_1)^{1/2}, \qquad \kappa_2 = (\pi/L)(K_3/K_2)^{1/2} \tag{5.50b}$$

Here $\boldsymbol{\nu}$ is a unit vector along the direction of the decrease, $\boldsymbol{\nu} = (\nu_x, \nu_y)$. Since the twist constant K_2 usually is smaller than K_1, the decrease of the perturbations $\delta\mathbf{n} \sim [\mathbf{e}_z\boldsymbol{\nu}]$ transverse to the direction

$\boldsymbol{\nu}$ turns out to be faster, $\kappa_2 > \kappa_1$. Connected with the LIFT we have in view the situation when the perturbation $\delta\mathbf{n}$ itself is directed along $\mathbf{e}_x$; then the decrease of δn_x along the x-axis will be characterized by the law $\exp(-\kappa_1|x|)$, and along the y-axis—by the law $\exp(-\kappa_2|y|)$. The self-focusing lens profile dependence on y will be more abrupt than that on x, since $\kappa_2 > \kappa_1$. For the LIFT, as a result, the transmitted beam divergence in the x-direction (i.e. in the direction of polarization $\mathbf{e} = \mathbf{E}/E$) must be less than in the y-direction. In the experiment, self-focusing fringes really have the form of an oval stretched out in direction of y-axis.

Very interesting is the problem of the dynamics of the LIFT and orientational relaxation when the field is switched on or off. For broad beams the relaxation rate $\delta n \sim \exp(-\Gamma t)$ is determined by the relation $\Gamma = K_3(\pi/L)^2/\gamma$, which is valid for the GON as well as for the LIFT (and generally for the Friedericksz effect of any nature). For narrow beams ($a_\perp << L$), the main part of the perturbation relaxes with the constant $\Gamma \sim Ka_\perp^{-2}/\gamma$ if the field is switched off to zero; this statement also relates both to the GON and to the LIFT.

Near the LIFT threshold (both above and below it) all the processes become slower by approximately $S_{\text{th}}/|S - S_{\text{th}}|$ times, compare with the expressions (5.5), (5.7). This statement is valid both for broad and for narrow beams. Here the situation is similar to the 2-nd order phase transition for FT in quasistatic fields.

The concrete expressions depend on the form of the intensity transverse distribution, on the ratios of the constants K_2/K_1, K_2/K_3 and $\varepsilon_a/\varepsilon_\perp$, on the initial and final power. It is necessary to admit that a quantitative theory of the effects for narrow beams near the steady state above the LIFT threshold is missing, since it is necessary to solve together all the fully nonlinear problems for the Frank and Maxwell equations because of strong saturation.

The comparison with the linearized theory may be done most conveniently by the exponential growth of small perturbations when the power is switched on above the threshold, and attenuation of the director at small declinations from the direction $\mathbf{n}^0 = \mathbf{e}_z$ when the power is decreased to values below the threshold. Let us recall that when $\theta_m << 1$ and $\theta(z) = \theta_m \sin(\pi z/L)$, the phase shift in the beam center for the light wave at normal incidence to a homeotropic cell, according to the formula (5.20), is

$$\delta\varphi = 2\pi\frac{L}{\lambda}\frac{\varepsilon_a\sqrt{\varepsilon_\perp}}{4\varepsilon_\parallel}\theta_m^2 \equiv 2\pi\frac{L}{4\lambda}\frac{n_\perp(n_\parallel^2 - n_\perp^2)}{n_\parallel^2}\theta_m^2 \tag{5.51}$$

and therefore $\delta\varphi \sim \theta_m^2$ must be changed by the law $\exp(-2\Gamma t)$. In this connection the normal incidence characteristic for the LIFT gives a result which differs from the case of the GON, where $\delta\varphi \sim \theta \sim \exp(-\Gamma t)$. In other words, the given constant of the director relaxation or growth Γ gives for the LIFT twice as great a constant 2Γ for the fringe number $N = \delta\varphi/2\pi$.

Experimental investigation of the reorientation dynamics for the LIFT has been carried out practically in all papers where LIFT was observed. The growth of perturbations $\delta\theta(t) = \delta\theta_0 \exp(|\Gamma| t)$ takes place from a small level of initial fluctuations. The sign of the resulting perturbation is determined by the sign of $\delta\theta_0$ and randomly takes the + or − value for the particular switching on of the intensity. The establishment time is given by the expression

$$\tau \approx \frac{1}{|\Gamma|} \ln\left(\frac{1}{|\delta\theta_0|}\right) \sim \frac{1}{S - S_{\text{th}}} \ln\left(\frac{1}{|\delta\theta_0|}\right)$$

This time is long enough due to the logarithmic multiplier (which fluctuates from experiment to experiment) and is especially long near the threshold. In typical experimental conditions τ varies from seconds to tens of minutes. The experimental investigation of small perturbations growth and attenuation provided in the papers [25,26] showed the following. 1) Exponential character of fringe number dependence on time was verified to a good accuracy in the initial stage. 2) There was a near-threshold slowing down of the relaxation rate and perturbation growth with the dependence $\Gamma \sim |S - S_{\text{th}}|^{-1}$. 3) The absolute value of the relaxation constant $|\Gamma|$ for the LIFT, as before it was for the GON, was in reasonable accordance with the data on constants of viscosity γ and elasticity K_i, obtained from non-optical experiments.

An interesting experimental method from the paper [27] should be noted. The LIFT in the NLC-cell of thickness 250 μm was excited by a broad argon laser beam ($\lambda = 0.51$ μm). The induced perturbation measurement has been carried out by the phase difference of ordinary and extraordinary waves for a weak very narrow beam of a He-Ne laser ($\lambda = 0.63$ μm); in the adiabatic approximation the O-wave must not at all be sensitive to the director deformations.

5.7 On LIFT possibility in smectics-C

Consider a cell with a SLC-C oriented by its layers parallel to the walls. Then the n-director making an angle ϕ with the z-axis can be oriented in the (x,z) plane, and the undisturbed value of the angle ζ

(see § 4.9) can be considered equal to zero. Here we shall consider the case, where the incident wave direction is chosen so that it propagates exactly along the axis inside the SLS-C; in this case the GON is absent. If this wave polarization is given by the vector $\mathbf{e} = \mathbf{e}_x \cos\phi - \mathbf{e}_z \sin\phi$ then, if $\varepsilon_a > 0$, the c-director in undisturbed orientation realizes the free energy minimum. But if the incident wave polarization is $\mathbf{e} = \mathbf{e}_y$, it is convenient to turn the c-director to decrease the energy $-\varepsilon_a|\mathbf{nE}|^2/16\pi$. In the case of the director strong anchoring on the cell boundaries, the Frank energy of the order of $B_3\zeta^2(\pi/L)^2$ prevents reorientation. As a result the effect arises only for beam power higher than some threshold value:

$$\frac{\varepsilon_a \sin^2\phi |E|^2_{\text{th}}}{16\pi} = B_3 \left(\frac{\pi}{L}\right)^2 \tag{5.52}$$

Analogously, the effect of the LIFT in SLC-C must be for a normal incidence of a light field on a cell, where the layers are tilted at an angle ϕ relative to the walls, and the n-director is placed along the normal to the walls, Figure 16. In this case

$$\frac{\varepsilon_a \sin^2\phi |E|^2_{\text{th}}}{16\pi} \approx \left(\frac{\pi}{L}\right)^2 (B_1 \sin^2\phi + B_3 \cos^2\phi + B_{13} \sin\phi\cos\phi) \tag{5.53}$$

From our point of view it would be very interesting to observe and investigate GON and LIFT specifics for smectics-C.

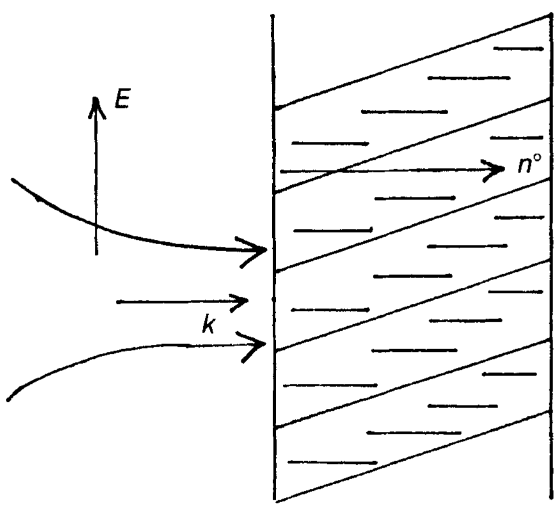


FIGURE 16 LIFT in smectics-C.

5.8 LIFT due to the ordinary wave.

Theory

Absolute direction of polarization is not essential for strictly normal incidence of a broad beam on a homeotropic cell, since the problem has rotation symmetry around the z-axis. For strongly oblique incidence $\mathbf{k} = k(\mathbf{e}_z\cos\alpha + \mathbf{e}_x\sin\alpha)$, $\alpha \sim 1$, the incident wave is split into o- and e-waves and while the e-wave gives rise to the GON, the o-wave passes practically without phase change, adiabatically following the director. A special case occurs for small angle α, when the deflection of the direction of $\mathbf{k}$ from the optical axis is small. Let us take

$$\mathbf{E}(\mathbf{r},z) = \exp\left(i\frac{\omega}{c}n_\perp\boldsymbol{\alpha}\mathbf{r} + ik_1z\right)\left\{\mathbf{B}(z) - \mathbf{e}_z\frac{\varepsilon_\perp}{\varepsilon_\parallel}(\mathbf{B}\boldsymbol{\alpha})\right\} \quad (5.54)$$

Here $\mathbf{B}(z)$ is the transverse part of the electric field of the wave, $\mathbf{B} = (B_x,B_y)$; $\boldsymbol{\alpha}$ is the two-dimensional refraction angle, $\boldsymbol{\alpha} = (\alpha_x,\alpha_y)$; $|\boldsymbol{\alpha}| << 1$ and $\boldsymbol{\alpha} \approx \boldsymbol{\alpha}_{\text{air}}/n_\perp$, $k_1 = (\omega/c)(\varepsilon_\perp - \alpha^2_{\text{air}})^{1/2}$. The vector (5.54) satisfies the equation $\text{div}\mathbf{D} = \text{div}(\hat{\varepsilon}\mathbf{E}) = 0$. We take the perturbed director as $\mathbf{n}(z,t) \approx \mathbf{e}_z + \boldsymbol{\theta}(z,t)$; $\boldsymbol{\theta} = (\theta_x,\theta_y)$, $|\boldsymbol{\theta}| << 1$. The equations describing the effects in question are

$$\frac{d\mathbf{B}}{dz} = i\xi(\boldsymbol{\alpha} - \boldsymbol{\theta})\big(\mathbf{B}(\boldsymbol{\alpha} - \boldsymbol{\theta})\big), \quad (5.55)$$

$$\gamma\frac{\partial\boldsymbol{\theta}}{\partial t} - K_3\frac{\partial^2\boldsymbol{\theta}}{\partial z^2} = \frac{\varepsilon_a\varepsilon_\perp}{8\pi\varepsilon_\parallel}\frac{1}{2}\left\{\mathbf{B}\big(\mathbf{B}^*(\boldsymbol{\theta} - \boldsymbol{\alpha})\big) + \mathbf{B}^*\big(\mathbf{B}(\boldsymbol{\theta} - \boldsymbol{\alpha})\big)\right\} \quad (5.56)$$

where $\xi = \omega\varepsilon_a\varepsilon_\perp^{3/2}/2c\varepsilon_\parallel$. We shall not present the derivation of the system (5.55–57) from the basic equations. Instead we shall explain its meaning with some simple examples.

Consider first $\boldsymbol{\theta} \equiv 0$, and $\boldsymbol{\alpha} = \mathbf{e}_x\alpha$. Then the solution of (5.55) is

$$\mathbf{B}(z) = \mathbf{e}_xB_x(0)e^{i\mu z} + \mathbf{e}_yB_y(0),$$
$$\mu = \xi\alpha^2 \equiv \frac{\omega}{c}\frac{\varepsilon_a\varepsilon_\perp^{3/2}}{2\varepsilon_\parallel}\alpha^2 \quad (5.57)$$

That means that the phase difference between the extraordinary wave ($\mathbf{e}_x$) and the ordinary one ($\mathbf{e}_y$) equals μz in accord with Fresnel theory of uniaxial crystals.

If $\boldsymbol{\theta} \neq 0$, but $\boldsymbol{\theta}$ does not depend on z then with the same accuracy one has

$$\mathbf{B}(z) = \mathbf{e}_1 B_1(0) \exp\{iz\xi(\boldsymbol{\theta} - \boldsymbol{\alpha})^2\} + \mathbf{e}_2 B_2(0) \qquad (5.58)$$

where $\mathbf{e}_1 = (\boldsymbol{\alpha} - \boldsymbol{\theta})/|\boldsymbol{\alpha} - \boldsymbol{\theta}|$, $\mathbf{e}_2 = [\mathbf{e}_z \mathbf{e}_1]$. Equation (5.58) also describes the propagation of the extraordinary wave ($\mathbf{e}_1$) and the ordinary one; now their phase difference is $\xi(\boldsymbol{\alpha} - \boldsymbol{\theta})^2 z$. We are going to use Eq. (5.55) for the z-dependent perturbations, $\boldsymbol{\theta} = \boldsymbol{\theta}(z)$. This equation is the only one which gives the correct results at $\boldsymbol{\theta}$ = const and which does not contain the spatial derivatives $d\boldsymbol{\theta}/dz$.

Equation (5.56) correctly describes GON, if one takes $\boldsymbol{\theta} = 0$ in the right-hand-side, and describes the linearized LIFT theory at $\boldsymbol{\alpha} = 0$.

Now let us use Eqs. (5.55–56) for the calculation of the LIFT threshold for oblique incidence of the ordinary wave $\boldsymbol{\alpha} = \mathbf{e}_x \alpha$, $\mathbf{B}(z = 0) = \mathbf{e}_y B_0$. We may linearize Eqs. (5.55–56) assuming $\theta_y(z,t)$ and $B_x(z,t)$ as infinitesimals of the 1-st order, and then

$$\frac{\partial \theta_y}{\partial t} = \frac{K_3}{\gamma} \frac{\partial^2 \theta_y}{\partial z^2} + \frac{\varepsilon_a \varepsilon_\perp}{8\pi \varepsilon_\parallel \gamma} \left\{ |B_0|^2 \theta_y - \frac{1}{2} \alpha (B_0^* B_x + B_0 B_x^*) \right\}, \qquad (5.59a)$$

$$\frac{\partial B_x}{\partial z} = i\mu B_x - i\xi B_0 \alpha \theta_y \qquad (5.59b)$$

and an equation, complex conjugate to (5.59b). Taking into account boundary conditions, we obtain for $\theta_y \equiv \theta(z,t)$

$$\frac{\partial \theta}{\partial t} = \hat{M}\theta = \left(\frac{\pi}{L}\right)^2 \frac{K_3}{\gamma} \left\{ \left(\frac{L}{\pi}\right)^2 \frac{\partial^2 \theta}{\partial z^2} + \rho \left[\theta(z,t) - \mu \int_0^z \theta(z',t) \sin\mu(z - z') dz' \right] \right\} \qquad (5.60)$$

Here ρ is the ratio of the incident intensity $|B_0|^2$ of the o-wave to the threshold value for LIFT at normal incidence; the latter case may easily be obtained from Eq. (5.60) by the substitution $\alpha \to 0$, $\mu \sim \alpha^2 \to 0$.

Considering the influence of the integral term by the perturbation theory, it is possible to obtain the correction to the LIFT threshold

$$S_{th} = S_{th}(\mu = 0)\left[1 + \left(\frac{\mu L}{\pi}\right)^2 + 9\left(\frac{\mu L}{\pi}\right)^4 + 0((\mu L)^6)\right] \quad (5.61a)$$

Thus, the dependence of the o-wave LIFT threshold on the refraction angle α is

$$S_{th}(\alpha) = S_{th}(\alpha = 0)\left[1 + \alpha^4\left(\frac{L}{\lambda}\frac{\varepsilon_a\varepsilon_\perp^{3/2}}{\varepsilon_\parallel}\right)^2 + \cdots\right] \quad (5.61b)$$

Let us discuss the reason for the threshold increase at $\mu L >> 1$. Suppose there is some smooth infinitesimal variation of $\theta = \theta_y$. Then the propagation of the wave in such a deformed medium leads to the adiabatic change of the polarization direction. It can easily be seen from (5.59b): if we omit $\partial B_x/\partial z$ for smooth variations, then $B_x \approx B_0\theta_y/\alpha$, and the adiabatically turned vector $\mathbf{B} = B_0(\mathbf{e}_y + \mathbf{e}_x\theta_y/\alpha)$ still belongs to the o-type wave, with E being orthogonal to the director. Therefore such smooth variation of the director does not lead to a decrease of the free energy. The polarization of the wave is unchanged, $\mathbf{E} \sim \mathbf{e}_y$ for a fast z-dependence of $\theta_y(z)$, and the electrical part of free energy goes down. However, such deformation gives rise to a high positive elastic contribution to the free energy.

The solution of eq. (5.60) allows one to find the compromise between these two tendencies, and at $\mu L \gtrsim 1$ the LIFT threshold increases considerably as a result.

Very interesting peculiarities of the director near-threshold behaviour, at $\mu L \gtrsim 1$, can arise due to the fact that the operator $\hat{M}$ in (5.60) is a real non-Hermitian. Consequently, the small perturbation behaviour can have more complicated character than simple exponential growth, namely the behaviour of the unstable solution can have the form $\exp(\Gamma t)$ with $\Gamma = \Gamma' + i\Gamma''$. The solution of Eq. (5.60) can be obtained by the method of separation of variables:

$$\theta(z,t) = \text{const}\, e^{(\Gamma' + i\Gamma'')t}(\sinh\mu_1 L \sinh\mu_2 z - \sinh\mu_1 z \sinh\mu_2 L) \quad (5.62)$$

where

$$\mu_1 = \left\{\frac{1}{2}\left[\left(\rho + \mu^2 - \frac{\gamma\Gamma}{K_3}\right)^2 + 4\gamma\Gamma\mu^2\right]^{1/2} - \left(\rho + \mu^2 - \frac{\gamma\Gamma}{K_3}\right)\right\}^{1/2} \quad (5.63a)$$

$$\mu_2 = \left\{\frac{1}{2}\left[\left(\rho + \mu^2 - \frac{\gamma\Gamma}{K_3}\right)^2 - 4\gamma\Gamma\mu^2\right]^{1/2} + \left(\rho + \mu^2 - \frac{\gamma\Gamma}{K_3}\right)\right\}^{1/2} \quad (5.63b)$$

and the parameters Γ', Γ'' are determined by the solution of the complex transcendental equation

$$(\mu_1^2 + \mu^2)\mu_2 \sinh\mu_1 L = (\mu_2^2 + \mu^2)\mu_1 \sinh\mu_2 L \quad (5.64)$$

For a given light intensity (parameter ρ) and angle dependent detuning μ, the quantities Γ' and Γ'' satisfying Eq. (5.64) form a discrete set of eigenvalues, and besides, the couple $+\Gamma''$ and $-\Gamma''$ corresponds to the given Γ'. For $\mu = 0$, the corresponding equations give the formula (5.5) with $\Gamma'' \equiv 0$, since the operator in this case becomes self-conjugated. The LIFT threshold for a given value of μ, is determined by the intensity minimal value, for which some of the complex increment $\Gamma' \pm i\Gamma''$ eigenvalues change the sign of the real part, $\Gamma' = 0$. Thus, at the threshold it is not necessary to have $\Gamma'' = 0$. Preliminary theoretical investigation of this question shows that the appearance of the threshold with $\Gamma'' \neq 0$ arises when $\mu L \gtrsim 2$.

An analogous consideration can be carried out for the LIFT threshold in a thin planar cell for an *o*-wave normal incidence. The role of the parameter μ, in this case, is played by the quantity $k_e - k_0 = (\omega/c)(n_\parallel - n_\perp)$. Eq. (5.60) with the substitution $K_3 \rightarrow K_2$ and all of its consequences remain valid also for this case. The LIFT threshold for a thin layer turns out to be high $S_{th} \sim L^{-2}$. A propitious circumstance here is the proportional shortening of the establishment time.

Experiment

Some preliminary experimental data on LIFT for ordinary type incident wave were presented in the paper [13] where the LIFT was observed for the first time. The authors of [13] pointed to some increase of the threshold and observed some oscillations of the resulting picture. The first purposeful experiment on the *o*-wave LIFT was carried out in [28]. A homeotropic cell of 135 μm thickness was filled by NLC 5CB. Argon laser radiation ($\lambda = 0{,}49$ μm) with power from zero up to 0,6 W was focused onto the cell; the spot diameter was $FWe^{-2}M \approx 180\mu$m. A strongly tilted probe beam served as a very sensitive tool for registration of the director distortions.

We shall discuss here only those results of the experiment [28] which can be compared quantitatively with the theory. The points in Figure 17 show the experimentally registered dependence of the LIFT threshold power on α_1^4 i.e. on the fourth degree of incidence angle. The solid line is the best-fit straight line, and the dashed line corresponds to the theoretical dependence $S_0[1 + (\mu L/\pi)^2]$ with μ taken from the experiment. So the agreement between the theory and experiment is very good for $\mu L \leqslant 1$. A nonmonotonic and even oscillating time behavior of the number of aberration fringes was observed in [28] for *o*-wave LIFT. We have as yet no clear theoretical explanation for that phenomenon; it may be in some way connected with the stimulated scattering of light, see chapter 7. Up to now we assumed tacitly that the threshold effects of light interaction with NLC take place only if the initial director $\mathbf{n}_0$ is perpendicular to the un-

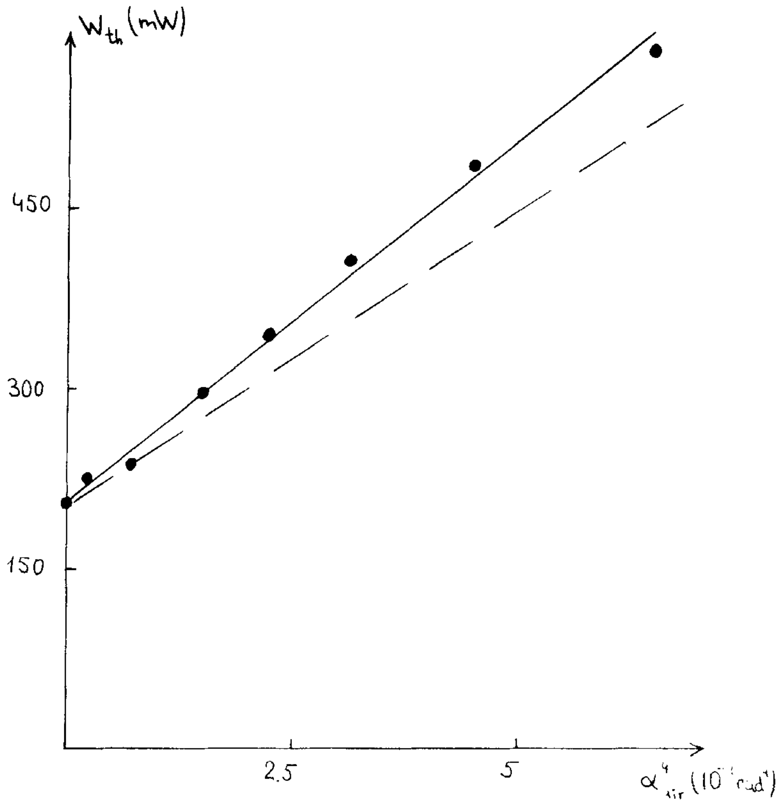


FIGURE 17 The plot of the LIFT threshold power versus the refraction angle [28].

disturbed vector $\mathbf{E}_0$ of the light field everywhere inside the NLC volume and only thresholdless effects likewise GON are possible at oblique disposition of $\mathbf{E}_0$ and $\mathbf{n}_0$.

In the recent paper [104] some schemes were suggested and theoretically investigated where threshold effects appear at extraordinary waves oblique incidence (LIFT-II). Since threshold effects usually occur beginning with the power sufficient to give about a 100 per cent GON-reorientation, one should specially take care to make weaker the GON. The following possibilities were discussed: 1) compensation of the GON in field of two waves; 2) application of additional quasi-static field (electric or magnetic); 3) *e*-wave propagation almost along the director in a planar cell. The director reorientation is in the waves' incidence plane in the first case. Besides, in all of the three cases a LIFT-II is possible, where the director comes out of incidence plane; this effect is specific for the light field and is due to the adiabatic following of the polarization vector $\mathbf{E}$ after the plane $(\mathbf{k}, \mathbf{n})$.

6 GRATING ORIENTATIONAL NONLINEARITY (GRON)

In § 2 we have considered the process of the interaction of two waves $E_1\exp(i\mathbf{k}_1\mathbf{r})$ and $E_2\exp(i\mathbf{k}_a\mathbf{r})$ of equal frequency and of considerably different wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ in a medium with nonlinearity of the form $\delta\varepsilon = 0.5\ \varepsilon_2|E|^2$. A wonderful phenomenon arises in this case: the interference of the waves E_1 and E_2 records in the medium a grating of dielectric permittivity and the same waves are diffracted on this grating with satisfaction of the Bragg condition (condition of phase matching). This phenomenon is surprising since such scattering does not lead to energy transfer from one wave to the other but only changes the waves effective phase velocities. The correction to the given wave (for example E_1) refractive index due to such a "grating" process is proportional to the intensity of another wave ($|E_2|^2$ in this example).

In the present Chapter we show that the "grating" orientational nonlinearity (GRON) must occur also for the interaction of light waves in liquid crystals.

GRON should not be mixed with the self-diffraction process considered in § 2.4. In the latter case a grating of the dielectric permittivity is also recorded: $\delta\varepsilon \sim E_1E_2^*\exp(i\mathbf{qr}) + E_1^*E_2\exp(-i\mathbf{qr})$, $\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2$. However, it is so smooth in the self-diffraction geometry that both the terms scatter the incident radiation with the same efficiency, the Bragg condition is not essential, and new waves with the wave vectors $\mathbf{k}_1 + \mathbf{q}$, $\mathbf{k}_2 - q$, $\mathbf{k}_1 + 2\mathbf{q}$, $\mathbf{k}_2 - 2\mathbf{q}$ etc. appear. There are no new waves in the GRON-case, where the Bragg condition works. One can show that the Bragg condition effectively works if the angle θ_{12} between the waves, the difference $n_1 - n_2$ of refractive indices and the NLC-layer thickness L fulfil the relation $L(\theta_{12}^2 + |n_1 - n_2|) >> \lambda$.

6.1 Mutual-focusing due to GRON in nematics

Consider the propagation of a plane wave incident normally on a planarly oriented cell with a NLC, Figure 18. Let

$$\mathbf{E}(z) = \mathbf{e}_x E_x(z) + \mathbf{e}_y E_y(z) \tag{6.1}$$

then for weak beams the light propagation through the undisturbed NLC is described by the following solution of the Maxwell equation

$$\mathbf{E} = \mathbf{e}_x E_1 \exp(ik_e z) + \mathbf{e}_y E_2 \exp(ik_0 z) \tag{6.2}$$

where $k_0 = \omega n_\perp | c$, $k_e = \omega n_\| | c$. The $\mathbf{E}$ vector local direction exhibits periodic changes with a period $\Lambda = \lambda/(n_\| - n_\perp)$. The torque reorienting the director turns out to be modulated by the same law. In this problem the director can be represented in the form $\mathbf{n}(z,t) \approx \mathbf{e}_x + \theta|z,t|\mathbf{e}_y$. Then, to a linear approximation in θ or, which is the same,

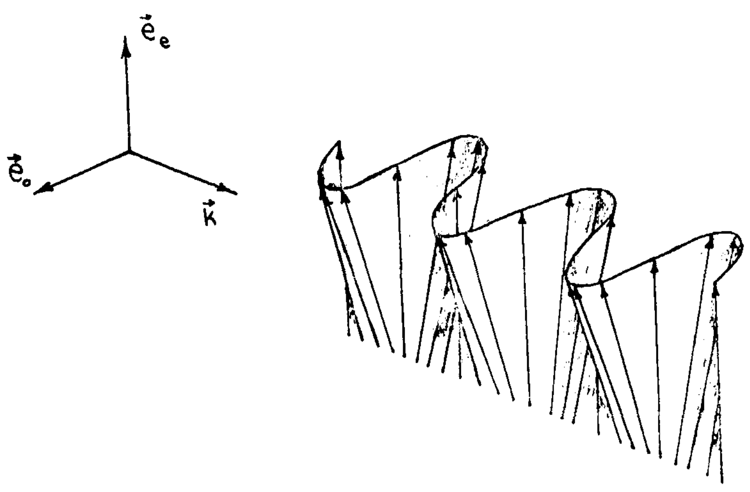


FIGURE 18 The periodic perturbations of the director (grating of $\hat{\varepsilon}$ in the volume of the *NLC*), generated by the interference of *o*- and *e*-waves.

in $|E|^2$, we obtain the variational equation

$$\gamma \frac{\partial\theta}{\partial t} - K_2 \frac{\partial^2\theta}{\partial z^2} = \frac{\varepsilon_a}{16\pi} [E_x(z)E_y^*(z) + E_x^*(z)E_y(z)] \tag{6.3}$$

Taking account of the perturbation $\delta\varepsilon_{ik} = \varepsilon_a(n_i^0\delta n_k + n_k^0\delta n_i)$ the Maxwell equations take the form

$$\frac{d^2E_x}{dz^2} + k_e^2E_x = -\frac{\omega^2}{c^2} \varepsilon_a\theta(z,t)E_y, \tag{6.4a}$$

$$\frac{d^2E_y}{dz^2} + k_0^2E_y = -\frac{\omega^2}{c^2} \varepsilon_a\theta(z,t)E_x \tag{6.4b}$$

Assume that the stationary solution of the system (6.3), (6.4) has the form

$$E = e_xE_1e^{ik_1z} + e_yE_2e^{ik_2z} + \cdots \tag{6.5}$$

where $\cdots$ means small corrections. Then the interference of the waves E_x and E_y in (6.3) generates a space-periodic grating

$$\delta\theta = \frac{\varepsilon_a}{16\pi K_2q^2} (E_1E_2^*e^{iqz} + E_1^*E_2e^{-iqz}) \tag{6.6}$$

with establishment time $\tau = \Gamma^{-1} = \gamma/K_2q^2$, where $q = k_1 - k_2$. If on the right hand side of Eq. (6.4) we keep only the terms satisfying the phase matching condition (the Bragg condition), then, for example, (6.4a) takes the form

$$d^2E_x/dz^2 + (k_e^2 + \omega^2\varepsilon_a|E_y|^2/16\pi K_2q^2)E_x = 0$$

This mean that the wave vector k_1 is changed due to the "grating" process: $k_1 = (k_e^2 + \omega^2\varepsilon_a|E_y|^2/16\pi K_2q^2)^{1/2}$. Since we keep to considerations in the first order of perturbation theory in $|E|^2$, it is possible to substitute $q = k_e - k_0$ and to write

$$k_1 = k_e + \frac{\omega^2\varepsilon_a|E_y|^2}{32\pi c^2k_eK_2q^2}, \qquad k_2 = k_0 + \frac{\omega^2\varepsilon_a|E_x|^2}{32\pi c^2k_0K_2q^2} \tag{6.7}$$

Relations (6.7) express the given wave (for example E_x) phase velocity dependence on another's intensity ($|E_y|^2$) typical for the "grating" optical nonlinearity. If the propagating waves have a bell-shaped intensity cross distribution, the beam $|E_y(\mathbf{r})|^2$ induces a focusing lens for the wave E_x and vice versa—the beam $|E_x(\mathbf{r})|^2$ induces the lens for the wave E_y. It can be said that GRON leads to the mutual-focusing of the waves E_x and E_y.

Let us point out once more the principal difference between the GRON and the self-diffraction considered in § 2.4. In the process of the GRON the terms in induction $\delta\mathbf{D}(\mathbf{r})$ of the form $E_1^2E_2^* \exp[i(2\mathbf{k}_1 - \mathbf{k}_2)\mathbf{r}]$ do not satisfy the phase matching condition and therefore do not play any role. On the contrary, the interfering waves in the self-diffraction process have approximately the same wave vector and propagate at a very small angle to each other. In this case just the terms of the form $E_1^2E_2^*$ give the diffraction higher orders, and the remaining small discrepancy in the wave vector is compensated by the uncertainty relation.

Let us make some numerical estimates. For OCBP at wavelength $\lambda = 0.5$ μm, $n_4 = 1.68$, $n_\perp = 1.53$. If we take $K_2 = 4.5.10^7$ dyne, $\Lambda = 2\pi/q \approx 3{,}3$ μm, the phase shift $\delta\varphi_y = GS_x$, where $G = 1.2.10^{-2}$ cm/W, and $S_x = cn_\parallel|E_x|^2/8\pi$ is measured in Watts per cm^2. The same magnitude G can be recalculated to the GRON nonlinearity constant

$$\varepsilon_2(\text{GRON}) = \frac{\varepsilon_a^2}{8\pi K_2 q^2}, \qquad \delta\varphi = \frac{\omega\varepsilon_2|E|^2L}{4\sqrt{\varepsilon}\,c} \tag{6.8}$$

The GRON-nonlinearity (6.8) is different from the GON-nonlinearity (4.7) mainly by the substitution $\pi/L \to q = 2\pi(n_\parallel - n_\perp)/\lambda$. The constant ε_2 (GRON) turns out to be, therefore, approximately $[(n_\parallel - n_\perp)L/2\lambda]^2$ times smaller than for the GON; for $L = 100$ μm, $n_\parallel - n_\perp \sim 0.15$, this weakening factor is about 250. By the same factor also the establishment time $\tau^{-1} = \Gamma = K_2q^2/\gamma$ is shortened.

One more essential advantage of the GRON is the possibility to take considerably more narrow beams: the decrease of the beam radius $a_\perp$ to the value $a_\perp << L$ does not influence to the constant ε_2(GRON) and, for a given total power of the beam, allows to increase considerably the power density S and, thus, the nonlinear phase shift.

We considered above the mechanism of the GRON in a NLC on an example of the simplest geometry. It is also good that here the GON is absent and therefore observation of the GRON can be carried out in the pure form. Using equations (3.11) there is no difficulty in treating the GRON also for arbitrary directed waves E_1 and E_2 of arbitrary polarizations.

Not writing down explicitly the expressions which are simple in principle (but cumbersome enough in form), let us point out their main properties. 1) The constant ε_2(GRON) is identically zero when two ordinary waves interact. 2) The constant ε_2(GRON) decreases proportionally to q^{-2}, where $\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2$. In particular, if the angle α_{12} between the directions of k_1 and k_2 is not small, then $q = 2k \sin(\alpha_{12}/2)$, and $\varepsilon_2 \sim \sin^2(\alpha_{12}/2)$, i.e. decreases fast with increase of the angle α_{12}; and shortens also the GRON establishment time. 3) If the wave vectors are collinear, the nonlinearity constant weakly depends on $\varepsilon_a = n_\parallel^2 - n_\perp^2$. Let us explain this on the above considered example. If the waves propagate perpendicular to the optical axis, then $q = (\omega/c)(n_\parallel - n_\perp)$ and

$$\varepsilon_2(\text{GRON}) = \frac{(n_\parallel + n_\perp)^2\lambda^2}{32\pi^3 K_2} \tag{6.9}$$

In particular, recording and reconstructing processes grow weak as ε_a^{+1} each at $\varepsilon_a \to 0$; however "rigidity" of the NLC relative to the grating decreases by the same degree; $Kq^2 \sim \varepsilon_a^2 \to 0$. The ε_2(GRON) such a weak dependence on q and ε_a for collinear $\mathbf{k}_1$ and $\mathbf{k}_2$ takes place also for an arbitrary angle between $\mathbf{k}$ and the optical axis $\mathbf{n}^0$. However, when $q \to 0$ the GRON establishment time increases as q^{-2}.

In our point of view, the GRON can be used to produce phase-conjugated waves by the fourwave scheme discussed in § 2.3. It is convenient to choose extraordinary waves, propagating exactly in opposite directions and exactly perpendicular to the director $\mathbf{n}^0$, as reference (E_1 and E_2), and as a signal—an ordinary wave approximately along the propagation direction of the wave $E_1(\mathbf{r})$. The interference of the waves E_1 and E_3^* then will record a grating and the wave E_2 will reconstruct it giving an o-type phase-conjugated signal $E_y(\mathbf{r}) \sim E_1E_2E_3^*(\mathbf{r})$. The important advantage of the scheme, where $(\mathbf{K}_1\mathbf{n}^0) = (\mathbf{K}_2\mathbf{n}^0) = 0$, is in the complete absence of reference waves self-focusing. The latter, as it is known, is the main reason of quality deterioration of the phase-conjugated wave in the four-wave method (compare also with § 4.5).

6.2 GRON in cholesterics: mutual-focusing and nonlinear optical activity

The fast spatial modulation of the torque with which the light tends to reorient the director is characteristic of the GRON. This modulation for nematics is connected with the relatively large value of the wave vector difference $|\mathbf{q}| = |\mathbf{k}_1 - \mathbf{k}_2|$ of interfering waves. Such a modulation of the torque in CLC appears even in the case of a smoothly varying field vector **E**—due to the director's fast periodic change.

Consider a planar helical structure in a cell with a cholesteric liquid crystal (CLC):

$$\mathbf{n}(z) = \mathbf{e}_x \cos\theta(z) + \mathbf{e}_y \sin\theta(z) \qquad (6.10)$$

In the equilibrium state when there are no external influences

$$\theta(z) = qz \qquad (6.11)$$

Assume that the light field propagates exactly along the z-axis. In this case the Maxwell equations in the circular polarization components

$$\mathbf{E}(z) = \frac{\mathbf{e}_x + i\mathbf{e}_y}{\sqrt{2}} E_+(z) + \frac{\mathbf{e}_x - i\mathbf{e}_y}{\sqrt{2}} E_-(z) \qquad (6.12)$$

take the form

$$\begin{aligned} (d^2/dz^2 + k^2)E_+ &= -(\omega^2\varepsilon_a/2c^2)\exp(-2i\theta(z))E_-(z) \\ (d^2/dz^2 + k^2)\,E_- &= -(\omega^2\varepsilon_a/2c^2)\exp(2i\theta(z))E_+(z) \end{aligned} \qquad (6.13)$$

where $k^2 = (\varepsilon_\parallel + \varepsilon_\perp)\omega^2/2c^2$. We shall consider the fastest process of the helix profile change $\delta\theta(z, t) = \theta(z,t) - qz$ inside the period at a fixed pitch.

The Euler-Lagrange-Rayleigh equations determining the director reorientation by light fields within a linear in $\delta\theta$ accuracy then have the form

$$\gamma \frac{\partial \delta\theta}{\partial t} - K_2 \frac{\partial^2 \delta\theta}{\partial z^2} = \frac{\varepsilon_a}{16\pi} [(|E_y|^2 - |E_x|^2)\sin 2qz$$

$$+ (E_x E_y^* + E_x^* E_y)\cos 2qz] \qquad (6.14a)$$

$$\equiv \frac{\varepsilon_a}{16\pi} (iE_+ E_-^* e^{2iqz} - iE_+^* E_- e^{-2iqz}) \qquad (6.14b)$$

Bilinear combinations from the light field amplitudes propagating along the $+z$ direction can be considered as smooth functions of z relative to the fast multipliers $\exp(\pm 2iqz)$, $\sin 2qz$, $\cos 2qz$ and then the stationary perturbation

$$\delta\theta(z) \approx \frac{\varepsilon_a}{16\pi K_2(2q)^2}\,[iE_+(z)E_-^*(z)e^{2iqz} - iE_+^*(z)E_-(z)e^{-2iqz}] \tag{6.15}$$

is established by the law $1 - \exp(-\Gamma t)$, where $\Gamma = K_2(2q)^2/\gamma$.

As a result of the director reorientation in the tensor of the medium's dielectric permittivity a perturbation of the form $\delta\varepsilon_{ik}(z) = \varepsilon_a(n_i^0\delta n_k + n_k^0\delta n_i)$ appears, where there are fast varying terms $\cos 4qz$ and $\sin 4qz$ and also terms constant in space, $\delta\bar{\varepsilon}$. Retaining only the latter we get

$$\delta\bar{\varepsilon}_{ik} = \frac{1}{2}\varepsilon_2(E_i^*E_k + E_iE_k^* - E_lE_l\delta_{ik}^{(2)}),\ \varepsilon_2 = \varepsilon_a^2/16\pi K_2(2q)^2 \tag{6.16}$$

and $\delta_{ik}^{(2)} = \delta_{ik} - (\mathbf{e}_z)_i(\mathbf{e}_z)_k$ is the two dimensional Kronecker's symbol. Thus, the strong linear-polarized wave $\mathbf{E} = \mathbf{e}_x E$ induces birefringence in the medium: $\delta\bar{\varepsilon}_{xx} = 0.5\ \varepsilon_2|E|^2$, $\delta\varepsilon_{yy} = -0.5\ \varepsilon_2|E|^2$. The presence of such a birefringence can be registered with the aid of a weak auxiliary wave, in general of another frequency and also maybe of the opposite direction of propagation. But if we are interested in self-focusing effects of the strong wave, then

$$(\delta\mathbf{D})_i = \delta\bar{\varepsilon}_{ik}\mathrm{E}_k = \frac{1}{2}\varepsilon_2(\mathbf{E}^*)_i(\mathbf{EE}) \tag{6.17}$$

Thus, "grating" perturbations of the director give nonzero contributions in the spatially averaged tensor $\delta\hat{\varepsilon}$ due to the CLC's periodic inhomogeneity.

In order to understand what kind of observable effects are generated by the addition to the induction of the form (6.17), it is necessary to take into account the effect of linear optics of CLC, namely, the rotation of polarization vector.

Taking into account Eq. (6.13), the corrections of the first order in $\delta\theta$, we get the set of nonlinear equations for the field amplitudes

E_+ and E_-

$$\left(\frac{d^2}{dz^2} + k^2 + \frac{\omega^2\varepsilon_a^2|E_-|^2}{16\pi c^2 K_2(2q)^2}\right) E_+ = -\frac{\omega^2\varepsilon_a}{2c^2} e^{-2iqz}E_-(z),$$
$$\left(\frac{d^2}{dz^2} + k^2 + \frac{\omega^2\varepsilon_a^2|E_+|^2}{16\pi c^2 K_2(2q)^2}\right) E_- = -\frac{\omega^2\varepsilon_a}{2c^2} e^{2iqz}E_+(z) \quad (6.18)$$

Equations (6.18) have been obtained by omitting the terms of the form $E_-^2 E_+^* \exp(-4iqz)$ strongly out of phase matching.

The action of the right hand side of (6.18) for small field intensity and out of the selective reflection band describes the CLC optical activity and can be taken into account by perturbation theory. For a right-polarized wave propagating along the $+z$ direction

$$E_+(z) \approx A_+e^{ik_+z}; \qquad E_-(z) = \frac{\omega^2\varepsilon_a}{8c^2q(K+q)} A_+e^{i(k_+ + 2q)z}$$

$$k_+ \approx k + \frac{\varepsilon_a^2\omega^4}{32c^4kq(k+q)} \quad (6.19)$$

and for the left-polarized wave

$$E_-(z) \approx A_-e^{ik_-z}, \quad E_+(z) = -\frac{\omega^2\varepsilon_a}{8c^2q(k-q)} A_-e^{i(k_- - 2q)z},$$

$$k_- \approx k - \frac{\varepsilon_a^2\omega^4}{32c^4kq(k-q)} \quad (6.20)$$

The specific optical activity of CLC ϕ(rad/cm) in this approximation is equal to

$$\phi = (k_+ - k_-)/2 \quad (6.21)$$

The field influence on the helix profile inside the period in addition to the corrections $k_+ - k$ and $k_- - k$ from (6.19), (6.20) leads also to nonlinear corrections of the form

$$\delta k_+(\text{GRON}) = \frac{\omega^2\varepsilon_a^2|E_-|^2}{32\pi c^2 kK_2(2q)^2}, \quad \delta k_-(\text{GRON}) = \frac{\omega^2\varepsilon_a^2|E_+|^2}{32\pi c^2 kK_2(2q)^2} \quad (6.22)$$

Nonlinear corrections (6.22) give, as for the GRON in NLC, the effect of mutual-focusing of the wave E_+ and E_-. Besides, if $|E_+|^2 \neq |E_-|^2$ (i.e. the polarization is not exactly linear but is elliptical), Eq. (6.22) describes the additional rotation of the major axis of polarization ellipse:

$$\delta\phi = \frac{\delta k_+ - \delta k_-}{2} = \frac{\omega^2 \varepsilon_a^2}{64 k c^2 K_2 (2q)^2} (|E_-|^2 - |E_+|^2) \quad (6.23)$$

It is interesting to note that in the approximation used (i.e. far from the selective reflection band and far from the Mauguin limit) both effects: GRON-mutual focusing and GRON-rotation—do not depend on the CLC chirality sign $q/|q|$.

The relations (6.15), (6.16) show that a linear-polarized light wave changes the helix profile along the period approximately so as does a magnetic field transverse to the helix axis. As it is known, the deformation induced by the magnetic field can bring out the higher order Bragg reflection for normal incidence. Using this analogy, it can be assumed that linear-polarized strong wave influence on the CLC can lead to the appearance of the Bragg higher order reflection or in reflection of some probe wave. The concrete calculations here, however, are cumbersome.

6.3 GRON of smectics-C

The GRON in smectics-C generally is analogous to the GRON in NLC. The difference is mainly in the circumstance that the condition of almost ideal incompressibility of SLC-C layers restricts the possible changes in the director direction. Consider, for example, the geometry where the **n**-director of a SLC-C is oriented parallel to the cell walls, $\mathbf{n}^0 = \mathbf{e}_x$; therefore the normal $\mathbf{e}_{z'}$ to the smectic layers makes an angle $\pi/2 - \phi$ with the z-axis $\mathbf{e}_{z'} = \mathbf{e}_x \cos\phi + \mathbf{e}_z \sin\phi$. Let the light wave be incident exactly normal to the cell walls; then

$$\mathbf{E}(z) = \mathbf{e}_x E_x e^{ik_1 z} + \mathbf{e}_y E_y e^{ik_2 z} \quad (6.24)$$

and besides in weak fields, when the SLC structure is not deformed $k_1 = \omega n_\parallel | c$, $k_2 = \omega n_\perp | c$. The equation for the c-director rotation angle ζ can be obtained, if in (4.45) the coordinate axes x and z are rotated at the angle $\pi/2 - \phi$. As a result one has

$$\gamma \frac{\partial \zeta}{\partial t} - \tilde{B} \frac{\partial^2 \zeta}{\partial z^2} = \frac{\varepsilon_a \sin\phi}{16\pi} (E_x E_y^* + E_x^* E_y),$$

$$\tilde{B} = B_1\cos^2\phi + B_3\sin^2\phi - 2B_{13}\sin\phi\cos\phi \qquad (6.25)$$

Stationary solution of this equation taking into account (6.24) has the form

$$\zeta(z) = \frac{\varepsilon_a \sin\phi}{16\pi\tilde{B}q^2}\left(E_\gamma E_\zeta^* e^{iqz} + E_x^* E_y e^{-iqz}\right) \qquad (6.26)$$

where $q = k_1 - k_2$. Let us substitute $\zeta(z)$ into the equation for the dielectric permittivity tensor. Let us omit the non-Bragg terms as it is usually done for the GRON consideration. Then for the wave vector nonlinear shift we obtain

$$\delta k_1 = \frac{\omega^2\varepsilon_a^2\sin^2\phi}{32\pi c^2 k_1 \tilde{B}q^2}|E_y|^2 \qquad (6.27)$$

and analogous expression with substitution $\delta k_1 \to \delta k_2$, $k_1 \to k_2$, $|E_y|^2 \to |E_x|^2$. If we take that the quantity $\tilde{B}$ for the SLC-C is of the order of the constant K_2 for NLC, the GRON-nonlinearity for SLC turns out to be smaller by approximately the multiplier $\sin^2\phi$. The GRON establishment by the law $1 - \exp(-\Gamma t)$ is characterized by the quantity $\Gamma = \tilde{B}q^2/\gamma$. It is not difficult to obtain the GRON coefficient also for an arbitrary geometry of the interacting waves.

We considered the c-director reorientation assuming strict preservation of the SLC layer structure. On the face of it it is possible to create such a deformation of the layers where the interlayer distance is not changed and, therefore, the large term in the elastic energy does not arise. For example, if $\mathbf{e}_z$ again coincides with the direction of the normal to the plane layers, and $u(\mathbf{r})$ (of dimensions of cm) is the layer displacement relative to the undisturbed planes, then the distortion $u(\mathbf{r}) = u_0\cos qx$ satisfies to relation $\partial u/\partial z \equiv 0$, i.e. does not lead to layer compression, but changes the director $\delta n_x \sim u_0 q$. If, however, the orienting influence proportional to $\cos qx$ is localized in a spatially restricted volume, then the distortion itself $u \sim \cos qx$ spreads without fading out of this region's boundaries to some distance L_1. In this external region the condition $\partial u/\partial z = 0$ is not already fulfilled and the quantity L_1 is determined by the minimization of the sum of the energy $L_1 B_0(\partial u/\partial z)^2 \sim B_0 u^2/L_1$ and the Frank energy $LK(uq^2)^2$; so that $L_1 \sim (B_0/Kq^4)^{1/2} \sim (a_{\text{mol}}q^2)^{-1}$. Here we took into account that the change of interlayer distance leads to the additional contribution in free energy density $\delta F = 0{,}5\, B_0(\partial U/\partial z)^2$, where $B_0 \sim k_B T/a_m^3$. For example, for a typical value $q \sim 3.10^4$

cm^{-1}, the value of L_1 is about 10^{-2} cm. The establishment time of the grating $\sim U_0 \cos qx$, therefore, will not be $\Gamma^{-1} \sim \gamma/Kq^2$, but $\tau \sim \gamma L_1^2/K$. At the required GRON power density $S \sim 5.10^4$ W/cm^2 and even at a very small absorption $\sigma = 10^{-1}$cm^{-1}, the medium is heated to 10^4 degree (???). This estimate leads to the conclusion that the GRON in a SLC accompanied by layer deformation is unlikely to be observed.

The stationary gratings of director orientation were experimentally studied in [29]. Those gratings were excited by the interference pattern of two collinear coherent plane waves of different (*o* and *e*) polarizations falling at a small angle (about 0,15 rad) of incidence at a homeotropic 100 μm cell with NLC 5CB. Argon laser radiation with $\lambda = 0{,}49$ μm and a power density about 500 W/cm^2 was used. In a standard GRON scheme one should observe the mutual-focusing effect. In experiment [29] however, a slightly different scheme was used to observe the induced grating. A counterpropagating *o*-wave was directed from the opposite side of the cell. The diffraction of that wave on the grating in the forward direction produced a new *e*-wave which was registered. The coefficient of the transformation of the intensity of *o*-wave to *e*-wave was up to 70%. Both the functional dependence and the absolute values of the parameters measured (transformation coefficient, relaxation time etc) on the power density of radiation and on the incident angle were in a good agreement with the theory based on Eq. (5.55).

7 ORIENTATIONAL STIMULATED SCATTERING (SS) OF LIGHT

7.1 General theory

Let the electromagnetic field in a medium consist of two waves of different frequency:

$$\mathbf{E}_{\text{real}}(\mathbf{r},t) = \frac{1}{2}\left(\mathbf{E}_1 e^{i\mathbf{k}_1\mathbf{r} - i\omega_1 t} + \mathbf{E}_2 e^{i\mathbf{k}_2\mathbf{r} - i\omega_2 t}\right) \tag{7.1}$$

If $\Omega = \omega_1 - \omega_2 \neq 0$, the grating $\delta\varepsilon(\mathbf{r},t)$ generated by the interference pattern of these waves turns out to be shifted in phase relative to the torque $\sim E_1 E_2^*$, because of the finite relaxation time. For example, if the wave $\mathbf{E}_1 = \mathbf{e}_x \cdot E_1 \exp(ik_1 z - i\omega_1 t)$ is extraordinary and the wave $\mathbf{E}_2 = \mathbf{e}_y E_2 \exp(ik_2 z - i\omega_2 t)$ is of the ordinary type, and the undisturbed director is along the $\mathbf{e}_x$ axis, (normal incidence of waves on the planar cell), then it is not difficult to obtain from (6.3) that

$$\delta\theta(z,t) = \frac{\varepsilon_a}{16\pi K_2 q^2}\left(\frac{E_1E_2^* e^{iqz - i\Omega t}}{1 - i\Omega/\Gamma} + \frac{E_1^* E_2 e^{-iqz + i\Omega t}}{1 + i\Omega/\Gamma}\right) \tag{7.2}$$

where $\Gamma = K_2q^2/\gamma$, $q = \omega(n_\parallel - n_\perp)/c$; the scattering of the wave E_1 on the disturbance $\delta\varepsilon \sim E_1^*E_2$, as for the GRON process, leads to the change of the effective wave vector k_2 by an amount

$$\delta k_2 = \frac{\omega^2\varepsilon_a^2|E_1|^2}{32\pi c^2 k_e K_2 q^2}\cdot\frac{1 - i\Omega/\Gamma}{1 + \Omega^2/\Gamma^2} \tag{7.3}$$

It is very important that at $\Omega > 0$ the quantity δk_1 acquires a negative imaginary part, so that $|E_2|^2 = \text{const}\exp(gz)$,

$$g = \frac{2\Omega/T}{1 + \Omega^2/\Gamma^2}\,\frac{\omega^2\varepsilon_a^2|E_1|^2}{32\pi c^2 k_0 K_2 q^2} \tag{7.4a}$$

The exponential gain of the signal wave E_2 is the result of energy scattering on the grating $\delta\varepsilon$ from the pumping wave E_1. Such a transformation of energy appears only at $\Omega > 0$ due to the grating phase relaxational shift; namely this process is called "stimulated scattering of light" (SS). The Gain coefficient g is proportional to the pump intensity $|E_1|^2$ and achieves a maximum at $\Omega = \Gamma$. Let us make numerical estimates for the nematic SCB at wavelength $\lambda = 0.5\ \mu$m. Taking $K_2 \approx 3.10^{-7}$ dyne, $\gamma \approx 0{,}7$ poise, $\varepsilon_a = n_\parallel^2 - n_\perp^2 = 0.6$, we obtain the frequency optimal shift $\Omega/2\pi = \Gamma/2\pi = 38$ Hertz, the relaxation time of the grating $\tau = \Gamma^{-1} \approx 4.10^{-3}$ seconds. If we write $g = GS_1$ and express S_1 in units of MW/cm^2, then $G \approx 10^4$ cm/MW. The amplification $e^5 \approx 150$ times on the distance $z = 250\ \mu$m can be obtained for the pump power density $S \approx 20$ kW/cm^2. The steady state is achieved if the pump pulse duration T exceeds $2\Gamma^{-1}$. If $T \lessapprox \Gamma^{-1}$, then the transient amplification factor is given by (see, e.g., [3])

$$|E_s|^2 \sim \exp\left\{2\sqrt{2}\left(G\Gamma z\int_0^t S_1(t')dt'\right)^{1/2}\right\} \tag{7.4b}$$

It should be emphasized that the transient gain coefficient $(G\Gamma)^{1/2}$ does not depend on the grating period $\Lambda = 2\pi/|\vec{\mathbf{k}}_1 - \vec{\mathbf{k}}_2|$ and on Frank constant K; namely, $G\Gamma \sim \varepsilon_a^2/\gamma$.

Let us represent the expression for $\delta\hat{\varepsilon}(\mathbf{r},t)$, the tensor perturbation induced by the interference of the fields (7.1), in a general geometry:

$$\delta\varepsilon_{ik}(\mathbf{r},t) = 4\pi e^{i\mathbf{qr}-i\Omega t}\chi_{iklm}(\Omega,\mathbf{q})(\mathbf{E}_1)_l(\mathbf{E}_2^*)_m$$

$$+ 4\pi e^{-i\mathbf{qr}+i\Omega t}\chi^*_{iklm}(\Omega,\mathbf{q})(\mathbf{E}_1^*)_l(\mathbf{E}_2)_m;$$

$$\chi_{iklm}(\Omega,\mathbf{q}) = \frac{\varepsilon_a^2}{64\pi^2}\frac{1}{q^2-(qn)^2}\Big\{A_2[f_{il}\, n_K n_m$$

$$+ f_{mi}n_k n_l + f_{lk}n_i n_m + f_{km}n_i n_l - 4q^2 n_i n_k n_l n_m]$$

$$- A_1[P_i q_l n_k n_m + P_m q_i n_k n_l + P_l q_k n_i n_m$$

$$+ P_K q_m n_i n_l - 4(\mathbf{qn})^2 n_i n_k n_l n_m]\Big\},$$

$$A_1 = [-i\gamma\Omega + K_1 q^2 + (K_3 - K_1)(\mathbf{qn})^2 + \chi_a\mathcal{H}^2]^{-1},$$

$$A_2 = [-i\gamma\Omega + K_2 q^2 + (K_3 - K_2)(\mathbf{qn})^2 + \chi_a\mathcal{H}^2]^{-1},$$

$$\mathbf{P} = 2\mathbf{n}(\mathbf{qn}) - \mathbf{q}, \qquad \chi_{iK} = [q^2 - (\mathbf{qn})^2]\delta_{ik} + P_i q_k \neq f_{ki} \qquad (7.5)$$

The expressions (7.5) are written for the case where a magnetic field $\mathcal{H} = \mathcal{H}\mathbf{n}^0$ is also applied on the NLC; however it does not contradict anything to consider $\mathcal{H} = 0$. The gain coefficient of the wave E_2 is given by the expression

$$g_2 = -\frac{\omega^2}{c^2}\frac{4\pi}{k_2}|E_1|^2 Im\left[e_{2i}e_{2l}e_{1m}e_{1k}\chi_{iklm}(\Omega,q)\right] \qquad (7.6)$$

where $\mathbf{e}_1$ and $\mathbf{e}_2$ are polarization unit vectors (for the field $\mathbf{E}$ or induction $\mathbf{D}$ which are equivalent to the required accuracy). The expression (7.5) allows one to calculate also the GRON constant in the most general geometry if to put $\Omega = 0$.

Consider specially the case of the backward $SS(\mathbf{k}_1 = \mathbf{e}_z k_e$, $\mathbf{e}_1 = \mathbf{e}_x$; $\mathbf{k}_2 = -\mathbf{e}_z k_0$, $\mathbf{e}_2 = \mathbf{e}_y$, $\mathbf{n}^0 = \mathbf{e}_z)$. Here for the same parameters of the medium and radiation $\Gamma \approx (4\pi\sqrt{\varepsilon}/\lambda)^2 K_2/\gamma \approx 6.10^4\ \mathrm{s}^{-1}$, $G = 30$ cm/MW, and at the distance $z = 500\ \mu$m amplification of e^{10} times is achieved at the pumping power density $S_1 \approx 6$ MW/cm^2. Similar to the spontaneous scattering, SS in this geometry occurs only with polarization change.

SS in cholesterics and smectics has in principle an analogous nature and can be considered as a particular case of the GRON when the frequencies do not coincide. *SS* in NLC, CLC and SLC has, however, interesting specific angular and polarizational features. For its detailed discussions we refer to the original papers [30–32].

7.2 Observation of SS

The observation of stationary *SS* in NLC was done in [33]. The *o*-type wave was incident at a small angle to the normal on the homeotropic 110 μm cell of 5CB. Argon laser was used ($\lambda = 0.49$ μm). The use of a small angle between the director and the propagation vector leads to two points. First according to the right-hand-side of Eq. (5.56), the influence of light on the director at $\theta \approx 0$ is weakened by a small factor $\alpha << 1$; and the influence of the disturbance θ on the interaction of the waves is also weakened by the factor α (right-hand-side of Eq. (5.55) taken in the 1-st order in θ). All of the above leads to a small numerator $\sim\alpha^2$ in the nonlinearity expression. However the denominator corresponding to the Frank energy K_3q^2 is proportional to α^4, since $q \sim \alpha^2$ for small refraction angle. Therefore the gain coefficient for *SS* (going in step with GRON constant) depends as $g \sim \alpha^{-2}$ on the refraction angle α:

$$g = \frac{\lambda n_{\|}^2 |E_0|^2}{16\pi K_3 n_\perp \alpha^2}, \qquad \Omega_{\text{opt}} = \Gamma = \frac{\pi^2 n_\perp^2 (1 - n_\perp^2/n_{\|}^2) K_3 \alpha^4}{\gamma \lambda^2} \tag{7.7}$$

The relaxation time Γ^{-1} coincides with the inverse optimum frequency shift.

A powerful *o*-wave was sent to the cell. Some part of it exhibited spontaneous scattering to *e*-wave of the near-to-optimum frequency shift. That part was amplified exponentially in the volume of the cell, and the intensity of the output *e*-wave was registered experimentally, see Figure 19 for the dependence of the scattered *e*-wave intensity W_S on incident *o*-wave intensity W_L.

The measurement of the frequency shift Ω was produced in the following way. A polarizer was placed at the output of the cell at such an angle that the *o*—and *e*-waves interfered in the transmitted radiation. The temporal behaviour of the sinusoidal interference beats were directly registered on an oscilloscope.

Both the functional dependencies and the absolute values of the measured quantities (g, Ω_{opt}, W_S/W_L) on the pumping intensity and refraction angle were in a very good agreement with the theory; see the original paper for the details.

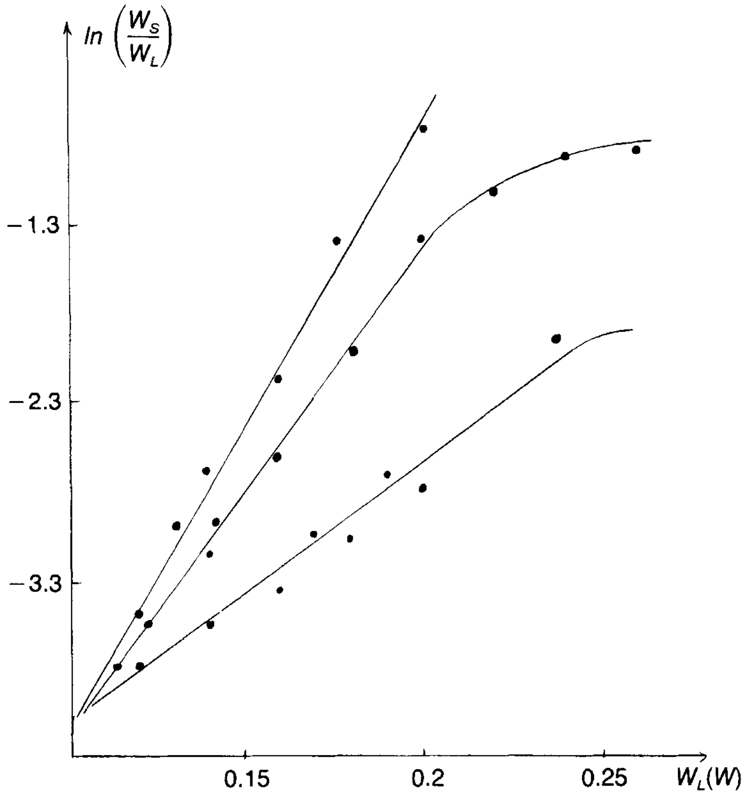


FIGURE 19 The dependence of the scattered *e*-wave intensity, W_s, on the incident *o*-wave intensity, W_L [33].

There was a feature in the observed data that has not yet been completely understood. After ca 10 to 15 seconds of the normal picture of *SS* the intensity of the resulting *e*-wave faded almost to zero. This may be due to a strong homogeneous deflection of the director by the *e*-wave via the GON mechanism; thus the angle between the optical axis and the propagation direction changed unfavorably for *SS*.

Just recently the paper† [105] was published in which the authors reported about the experimental registration of forward *SS* for the normal incidence of a *e*-wave onto a planar cell with the nematic

†B.Ya.Zel'dovich, S. K. Merslikin, N. F. Pilipetsky, A. V. Sukhov. Observation of the forward stimulated scattering of light in a planar nematic. Pisma ZhETF, **41**, No. 10, p. 418, 1985 (JETP Letters **41**, 00, 000, 1985).

5CB. The pumping radiation (λ = 0,69 μm) was generated by a ruby laser; pulse duration $\sim 8.10^{-4}$ sec was shorter than the relaxation constant $\tau = \gamma/Kq^2 \sim 2.10^{-3}$ sec for a grating with a period $\Lambda = 2\pi/q = \lambda/(n_\parallel - n_\perp) \approx 3$ μm. Therefore the transient theory of *SS* was used for the comparison with experiment. Experimental results were in a good agreement with the theory both in respect to the functional dependences and to the absolute value of the exponent of the gain factor.

8 LIGHT INFLUENCE ON THE SURFACE LAYER OF *LC*

In the present chapter a number of problems is discussed where the light wave is localized in a thin layer at a surface. On the face of it, the corresponding effects are weaker compared to the volume ones by $L/\Delta z$ times, where L is the cell thickness.

In particular problems, however, there is a number of favorable factors compensating the influence of the small parameter $\Delta z/L$. It is important that the use of surface interaction allows one to investigate *LC* in the presence of strong volume scattering and in the region of light frequencies where there is a strong absorption.

8.1 GON and LIFT in the surface light wave

Consider the orientational influence of a non-propagating light wave localized near the boundary $z = 0$; such a wave can arise when the light is incident on the NLC from a medium with a greater refractive index n_i due to the effect of total internal reflection (TIR), see Figure 20. Let the nematic be planarly oriented ($\mathbf{n}^0 = \mathbf{e}_x$) by rubbing of the surface $z = L$, and the surface $z = 0$ has no influence on the orientation.

Let us take the incident wave vector in the form

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

Consider the case where the wave is polarized perpendicular to the incidence plane $\mathbf{e}_{in} \sim [\mathbf{k}[\mathbf{k}\mathbf{e}_z]]$. In this case the field in the NLC has only the components E_x and E_y, and the equation for the director's stationary profile $\mathbf{n}(z) = \mathbf{e}_x\cos\theta(z) + \mathbf{e}_y\sin\theta(z)$ has the form

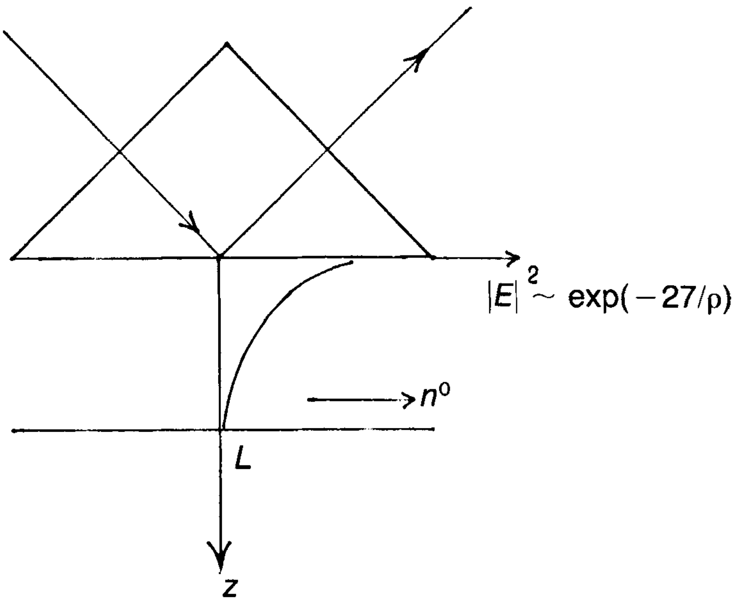


FIGURE 20 A prism serves as a plate of the *NLC*-cell in the plane $z = 0$ where there is total light reflection.

$$-\gamma \frac{\partial\theta}{\partial t} + K_2 \frac{\partial^2\theta}{\partial z^2}$$

$$+ \frac{\varepsilon_a}{16\pi}\left\{ \sin 2\theta\,(|E_y|^2 - |E_x|^2) + \cos 2\theta\,(E_x E_y^* + E_x^* E_y)\right\} = 0 \quad (8.1)$$

If the anisotropy is small, $\varepsilon_a << \varepsilon_\perp$, the field in the NLC can be taken as that for an isotropic medium; then, using the usual relations of the TIR theory, we obtain

$$\mathbf{E}(z) = \mathbf{e}_{\text{in}} E_{\text{in}} \frac{2\exp(-z/2\rho)}{1 + i\sqrt{1 - (\sin^2\alpha_{\text{TIR}}/\sin^2\alpha)}} \quad (8.2)$$

where α_{TIR} is the total reflection angle, $\sin^2\alpha_{\text{TIR}} = n^2/n_i^2$

$$\rho = \frac{\lambda}{4\pi n_i \sin\alpha}\left[1 - \left(\frac{\sin\alpha_{\text{TIR}}}{\sin\alpha}\right)^2\right]^{-1/2} \quad (8.3)$$

LIFT takes place when the unperturbed director is in the plane of incidence, i.e. $\beta = 0$, $\mathbf{e}_{\text{in}} = \mathbf{e}_y$. Linearizing the equation (8.1) yields

$$-\frac{\gamma}{K_2}\frac{\partial\theta}{\partial t} + \frac{\partial^2\theta}{\partial z^2} + b^2\theta\exp\left(-\frac{z}{\rho}\right) = 0 \quad (8.4)$$

where $b^2 = \varepsilon_a|E_{in}|^2/2\pi K_2$. If ε_a is not small, the director reorientation leads to deflection conditions modification and gives rise to the field component $E_x \neq 0$. The accurate consideration of these effects carried out in [34] does not change the qualitative conclusions and leads only to a redetermination of the quantities b^2 and ρ.

The problem of the determination of the threshold from Eq. (8.4) must be solved with the boundary conditions $\theta(z = L) = 0$, $(d\theta/dz)_{z=0} = 0$; the latter is the condition of the director free orientation at $z = 0$. Using the small parameter $\rho/L \sim 10^{-2}$ at $\rho \sim 1$ μm, $L \sim 100$ μm, the action of the term $b^2\theta\exp(-z/\rho)$ can be described by the introduction of the boundary condition

$$\left[\frac{d\theta}{dz} + A\theta\right]_{z=0} = 0; \qquad A = \int_0^\infty b^2\exp\left(-\frac{z}{\rho}\right)dz = 6^2\rho \quad (8.5)$$

and by omitting the term $\sim b^2$ in Eq. (8.4). The instability threshold is determined by the appearance of a nontrivial solution with zero time increment. Then $\theta(z) = \text{const}(z - L)$ and on the threshold $AL = 1$, therefrom we get for the threshold power density in the incident wave

$$S_{th} = \frac{cn_i|E_{in}|^2}{8\pi} = \frac{cn_iK_2}{4\pi\varepsilon_aL_\rho} = \frac{c\varepsilon_iK_2(\sin^2\alpha - \sin^2\alpha_{TIR})^{1/2}}{\varepsilon_a\lambda L} \quad (8.6)$$

Comparison with the threshold power density (5.6) for LIFT in the volume of the identical thickness homeotropic aligned cell yields

$$\frac{S_{th}(\text{SLIFT})}{S_{th}(\text{LIFT})} = \frac{\varepsilon_i}{\sqrt{\varepsilon_\perp}}\frac{K_2}{K_3}\frac{L(\sin^2\alpha - \sin^2\alpha_{TIR})^{1/2}}{\pi^2\lambda} \quad (8.7)$$

Thus, in (8.7) there is a large dimensionless factor $L/\Delta z = L(\sin^2\alpha - \sin^2\alpha_{TIR})^{1/2}/\lambda$, connected with the circumstance that the light action is realized in small part of the cell. However, the ratio K_2/K_3, which is usually about 0.3, and the numerical coefficient $\pi^2 \approx 0.1$ turn out to be propitious small parameters. All this shows that the LIFT threshold in the field of a surface light wave can be of the order of the LIFT threshold for the homeotropic cell of the same thickness.

For determination of the above-threshold structure, in the same approximation, $\rho << L$ and $\varepsilon_a << \varepsilon_\perp$, it is possible to use the boundary condition $[d\theta/dz + 0.5A\sin 2\theta]_{z=0} = 0$ therefrom $\theta(z) = \theta_1(1 - z/L)$ and the quantity θ_1 is determined from the solution of the equation

$$\frac{\sin 2\theta_1}{2\theta_1} = \frac{S_{\text{th}}}{S} \tag{8.8}$$

If the angle $\beta \neq 0$, director reorientation occurs, analogous to the GON, in the first order in the light intensity. Taking $E_x = |E|\cos\beta$, $E_y = |E|\sin\beta$, where $|E|$ is determined in (8.2) and solving Eq. (8.1) in the first order in θ, we obtain

$$\theta(z = 0) \approx \frac{1}{2}\sin 2\beta\,\frac{S}{S_{\text{th}}} \tag{8.9a}$$

The establishment time for the GON is $\tau \approx 4\gamma L^2/\pi^2 K_2$, and it is greater for the LIFT, especially near the threshold.

These effects must also take place in the case where a cholesteric is taken instead of a nematic; all the obtained formulae are valid also for a CLC, if we take $\theta(z) - q(z - L)$ instead of $\theta(z)$. Qualitatively the same effect but with slightly modified formulae must take place also for a cell with a SLC-C, where the layers are parallel to the cell walls.

If the surface $z = 0$ has a preferable orientation $\theta = 0$, then it is possible to introduce a surface energy $U_s(\text{erg/cm}^2) = 0.5\sigma\sin^2\theta(z = 0)$ which corresponds to the anchoring length R_a, compare with expression (4.22). Then the power density threshold increases by the factor

$$S_{\text{th}}(R_a) = \frac{1 + L/R_a}{1 + \rho/R_a}\,S_{\text{th}}(R_a = 0) \tag{8.9b}$$

obtained with the assumption $R_a >> \rho$. When R_a decreases to the value $R_a \lesssim L$, the threshold increases as R_a^{-1}. After R_a becomes smaller than ρ, the threshold again becomes independent of R_a and is determined by expression (8.6) with the substitution $L \rightarrow \rho$, see § 8.4.

The experiment [29] has been carried out by the scheme of Fig. 20 using argon laser radiation ($\lambda = 0.51\ \mu$m) and a nematic

MBBA. The director deflection was registered by the polarization plane rotation of a He-Ne laser probe beam transmitting through the NLC from the side of the strongly orienting surface $z = L$. In this geometry the LIFT has been registered for incident wave power density $S_{th} \approx 1.2$ kW/cm^2 for cell thickness 50 μm. Note that the value of ρ was about $2.5 \cdot 10^{-5}$ cm. The rotation angle observed with the air of the probe beam was about 25° at $S/S_{th} \sim 5$.

The absolute value of S_{th} is in good agreement with the theoretical estimate by formula (8.6). The above-threshold angle θ_1 had various signs in various experiments according to the general notions about LIFT instability developing from initial random fluctuations. The magnitude $|\theta_1|$ above the threshold strongly deviates from Eq. (8.8). This can be connected with the circumstance that the condition of TIR was not fulfilled for the extraordinary wave (the prism refractive index $n_i = 1.76$ is close to the magnitude $n_{\|} = 1.75$). The establishment time was 120 to 150 s at intensities near the threshold. The analog of GON, i.e. the effect without threshold was also observed at $\beta \neq 0$.

8.2 Surface plasmon interaction with *LC*

Localized electromagnetic waves, i.e. surface plasmons, can propagate near the interface of two media with dielectric permittivities $\varepsilon_{\perp}$ and ε_M, see, for example [30,31]. For their existence it is necessary that one of the media (usually-metal) possesses a negative ε so that $-\varepsilon_M = |\varepsilon_M| > \varepsilon_1 > 0$. Let us designate by $\mathbf{e}_z$ the normal to the interface, and let $\mathbf{m}$ be a unit vector along the surface plasmon propagation direction, $\mathbf{m} = \cos\beta \mathbf{e}_x + \sin\beta \mathbf{e}_y$; let also $\boldsymbol{\nu} = [\mathbf{e}_z \mathbf{m}] = \mathbf{e}_y \cos\beta - \mathbf{e}_x \sin\beta$. Considering then $\varepsilon = \varepsilon_1$ at $z > 0$ and $\varepsilon = -|\varepsilon_M|$ at $z < 0$, it is possible to write down for surface plasmon (SP) electric field [31]:

$$\mathbf{E}(\mathbf{r}) = Be^{ik\mathbf{mr}} \left\{ \begin{bmatrix} e^{-\kappa_1 z} \\ -\dfrac{\varepsilon_1}{|\varepsilon_M|} e^{\kappa_2 z} \end{bmatrix} \mathbf{e}_z + \begin{bmatrix} -i \dfrac{\kappa_1}{k} e^{-\kappa_1 z} \\ -i \dfrac{\varepsilon_1}{|\varepsilon_M|} \dfrac{\kappa_2}{k} e^{\kappa_2 z} \end{bmatrix} \mathbf{m} \right\} \quad (8.10)$$

In expression (8.10) the upper line refers to region $z > 0$, and the lower one to $z < 0$. The quantities $(2\kappa_1)^{-1}$ and $(2\kappa_2)^{-1}$ characterize the SP intensity localization region at $z > 0$ and $z < 0$, respectively, and further

$$\kappa_1 = \left(k^2 - \frac{\omega^2}{c^2}\varepsilon_1\right)^{1/2}, \qquad \kappa_2 = \left(k^2 + \frac{\omega^2}{c^2}|\varepsilon_M|\right)^{1/2} \tag{8.11}$$

For example, at $z > 0$ the intensity decreases as $\exp(-z/\rho)$, where $\rho = (2\kappa_1)^{-1}$. Besides, the equality $\kappa_2/\varepsilon_1 = \kappa_2/|\varepsilon_M|$ follows from field continuity at the surface; the dispersion equation (the connection between ω and $\mathbf{k}$), follows therefrom:

$$k^2 = \frac{\omega^2}{c^2}\frac{\varepsilon_1|\varepsilon_M|}{|\varepsilon_M| - \varepsilon_1} \tag{8.12}$$

The quantity $|B|^2$ can be expressed in terms of the component of the Poynting vector $\mathbf{S}$. Introduce the specific flux of energy $W(\text{erg/cm}\cdot\text{s}) = \int_{-\infty}^{\infty}(\mathbf{Sm})dz$ then

$$W = |B|^2\rho\frac{\omega\varepsilon_1}{8\pi K}\left(1 - \frac{\varepsilon_1^2}{\varepsilon_M^2}\right) \tag{8.13}$$

and the term $-\varepsilon_M^2/\varepsilon_1^2$ corresponds to the opposite flux of the energy existing in the medium with a negative dielectric permittivity.

We shall be interested in the problem, where SP propagates along the interface of a solid body with $\varepsilon = \varepsilon_M < 0$ and a LC layer. Strictly speaking it is necessary to solve the problem of SP taking into account the anisotropy of the liquid crystal tensor $\hat{\varepsilon}$. However we shall use the above formulae obtained for SP at an isotropic media interface as a simplified tool for estimations. The influence of SP on LC orientation has almost the same character as for the non-propagating surface wave obtained by TIR considered in § 8.1. In particular SP can realize the twist deformation with the threshold (if $\mathbf{m} \perp \mathbf{n} = \mathbf{e}_x$) or without it (if $0 < |\mathbf{mn}_0| < 1$) in the geometry presented in Figure 20, where a medium with $\varepsilon = \varepsilon_M < 0$ must be implied instead of the prism.

For metals $|\varepsilon_M| >> 1$ often enough even at optical frequencies. Then $k^2 \approx (\omega^2\varepsilon_1/c^2)(1 + \varepsilon_1/|\varepsilon_M|)$, the intensity localization size in LC is small, $\rho = \lambda\sqrt{|\varepsilon_M|}/4\pi\varepsilon_1$ and therefore $|\mathbf{Ee}_z|$ is greater than $|\mathbf{Em}|$ approximately by the factor $(|\varepsilon_M|/\varepsilon_1)^{1/2}$.

Then the interaction of the SP with a NLC in the geometry of Figure 20 is interesting with account of splay deformation. It is possible to consider the perturbations of the form $\mathbf{n}(z,t) = \mathbf{e}_x\cos\theta(z,t) + \mathbf{e}_z\sin\theta(z,t)$ if the director is not held on the surface $z = 0$. Let the SP propagate in the direction $\mathbf{k} = \mathbf{e}_y k$ perpendicular to the director. The linearized equation for $\theta(z,t)$ then takes the form

$$\frac{\gamma}{K_3}\frac{\partial\theta}{\partial t} - \frac{\partial^2\theta}{\partial z^2} + b^2\theta\exp\left(-\frac{z}{\rho}\right) = 0 \tag{8.14}$$

where $b^2 = \varepsilon_a|B|^2/2\pi K_3$. Treating this equation just as in § 8.1, we obtain that the threshold condition for the layer reorientation by the SP field has the form $b^2\rho L = 1$, therefrom the SP threshold specific power W_{th} (erg/cm · s) is obtained as

$$W_{th} = \frac{c\varepsilon_1 K_3}{4\varepsilon_a L}\left(\frac{|\varepsilon_M| - \varepsilon_1}{\varepsilon_1|\varepsilon_M|}\right)^{1/2}\left(1 - \frac{\varepsilon_1^2}{\varepsilon_M^2}\right) \tag{8.15}$$

For the validity of the one-dimensional approach which was used above the SP packet size along the y-coordinate must not be smaller than the quantity L, therefrom the condition on a SP whole power $W_{th}L$ (erg/s) is obtained. Assuming $\varepsilon_1/|\varepsilon_M| << 1$, $k = \omega\sqrt{\varepsilon_1}/c$, $\varepsilon_a \sim 1$, $K_3 \sim 10^{-6}$ dyne, we obtain the estimate $W_{th}L \sim 10^{-3}$ W. This quantity turns out to be independent of the thickness L. When $L = 10^{-2}$ cm the specific threshold flux is $W_{th} \sim 10^{-1}$ W/cm.

Up to now we considered the influence of surface waves on the NLC orientation. The changed orientation modifies, in its turn, the amplitude and the phase of the same waves—the reflected due to the TIR wave or propagating surface plasmon. For example, the effects of self-focusing due to the GON and LIFT are possible for SP, grating nonlinearity at two plasmons interaction, SP stimulated scattering in the presence of other fields. Very interesting are the scattering effects from SP to a volume wave and vice-versa due to the non-linear orientational interaction. This field of activity is as yet almost unexplored.

8.3 Cholesteric helix pitch change

The strongest influence of a light wave on a cholesteric helix pitch is connected with the trivial effect of the CLC heated by the light field. The temperature stationary profile establishment time is $\tau = (L/\pi)^2/\chi = 2.5.10^{-3}$ s at cell thickness $L = 50$ m and thermal diffusivity coefficient $\chi \sim 10^{-3}$ cm^2/s. For a moderately pure LC the absorption coefficient σ can be estimated as $\sigma \sim 1$ cm^{-1}. Then, for the incident wave power density $\sim 10^3$ W/cm^2, energy release during the time τ is of the order of 2.5 J/cm^3, which corresponds to a temperature increase of approximately 2°C. The detailed discussion of thermal effects will be postponed to Chapter 10. In this Section we shall neglect completely the thermal effects and discuss how the light can change the CLC helix pitch due to the direct action of the field.

The strongest effect turns out to be at Bragg reflection ($\omega n/c \approx q$) from the Grandjean planar structure of a normally incident light wave. The magnitude ε_a is usually small for CLC, $\varepsilon_a \sim 0.03$–0.3; however, this smallness is compensated for by the reflected wave penetrating to depth $\Delta z \sim \lambda/\varepsilon_a$. As a result, the torque which is transferred from the light wave to the helix, is independent on the cell thickness L, if L is large enough.

The Maxwell equations for this problem have the form (6.12), (6.13). The Euler-Lagrange-Rayleigh equations have the form

$$\gamma\frac{\partial\theta(z,t)}{\partial t} - K_2\frac{\partial^2\theta(z,t)}{\partial z^2} = \frac{\varepsilon_a}{16\pi}(iE_+E_-^*e^{2i\theta} - iE_+^*E_-e^{-2i\theta}) \quad (8.16)$$

The notations coincide with those used in § 6.2, and Eq. (6.14) is obtained by linearization of the exact equation (8.16). It follows from (6.13) and (8.16) in the stationary case that

$$dM(z)/dz = 0,$$

$$M(z) = -K_2\left(\frac{d\theta}{dz} - q\right) + \frac{c^2}{16\pi\omega^2} \cdot \left[\left(iE_+\frac{\partial E_+^*}{\partial z} - iE_-\frac{\partial E_-^*}{\partial z}\right) + C.C.\right] \quad (8.17)$$

and expresses the conservation law of angular momentum for the system CLC + field. Although (8.17) is easily verified directly from (6.13) and (8.16), the initial expression we have obtained using the Noether theorem which connects the angular momentum conservation with the invariance of the Lagrangian, with respect to rotation around the z-axis.

Outside the LC, i.e. in any other transparent dielectric or in the vacuum, the term $\sim K_2$ is absent and the expression (8.17) corresponds to the flow of a spin angular momentum $+\hbar$ or $-\hbar$ carried by each quantum with energy $\hbar\omega$ and with clockwise or counterclockwise circular polarization, respectively.

Consider the waves in the dielectric outside the CLC and denote the intensity for the incident light as $S^{(z)} = S_+^{(z)} + S_-^{(z)}$ (erg/cm^{2}s), where $S_+^{(z)}$ corresponds to clockwise circular polarization component, and $S_-^{(z)}$—to the counterclockwise one, respectively: $\mathbf{E}_+^{(z)} \sim (\mathbf{e}_x + i\mathbf{e}_y)\exp(ikz)$, $\mathbf{E}_-^{(z)} \sim (\mathbf{e}_x - i\mathbf{e}_y)\exp(ikz)$. The reflected waves may be represented in an analogous way: $S^{(-z)} = S_+^{(-z)} + S_-^{(-z)}$; $\mathbf{E}_+^{(-z)} \sim (\mathbf{e}_x + i\mathbf{e}_y)\exp(-ikz)$, $\mathbf{E}_-^{(-z)} \sim (\mathbf{e}_x - i\mathbf{e}_y)\exp(-ikz)$. Then direct calculation with (8.17) shows, that

$$M = \omega^{-1}(S_+^{(z)} - S_-^{(z)} - S_+^{(-z)} + S_-^{(-z)}) \qquad (8.18)$$

and the constancy of M is ensured here by the constancy of each of the quantities $S_k^{(i)}$. At $\varepsilon_a << \varepsilon$ the same separation can be done inside the CLC and besides $S_+^{(z)}$ and $S_-^{(-z)}$ are constant to a good accuracy for a right ($q > 0$) CLC. Due to the Bragg reflection the waves $S_-^{(z)}$ and $S_+^{(-z)}$ exchange energy so that $dS_-^{(z)}/dz = -dS_+^{(-z)}/dz$. In this case Eq. (8.17) yields

$$d\theta/dz = q + (\omega K_2)^{-1}(S_+^{(z)} + S_-^{(-z)}\omega M) - (\omega K_2)^{-1} \cdot (S_-^{(z)} + S_+^{(-z)}) \equiv q + \text{const} - (2/\omega K_2)S_-^{(z)}(z) \qquad (8.19)$$

Only the last term is variable in space.

The particular behaviour of $d\theta/dz$ is determined also by conditions at the cell boundaries. Consider the problem where a wave with a flux $S_-^{(z)}(z = 0) = S_0$ is incident to the boundary $z = 0$ of the cell. (It is not difficult to understand that in this approximation the presence of the wave $S_+^{(z)}$ changes nothing). Assume that the wave $S_-^{(z)}$ is reflected with a reflection coefficient R, so that $S_+^{(-z)}(z = 0) = RS_0$, correspondingly the transmission coefficient is determined by the equality $S_-^{(z)}(z = L) = TS_0$ and $T = 1 - R$ since $S_+^{(-z)}(z = L) = 0$. The boundary condition has the form $d\theta/dz = q$, if the director orientation is free at the boundary.

Two somewhat different statements of the problem are possible. In the first, the free boundary is at $z = 0$; then

$$\left.\frac{d\theta}{dz}\right|_{z=0} = q \Rightarrow \frac{d\theta}{dz} = q + \frac{2}{\omega K_2}\left(S_0 - S_+^{(z)}(z)\right) \qquad (8.20)$$

If the cell thickness L is much greater than the length $\rho = \lambda n/\pi\varepsilon_a$ at which the Bragg reflection is developed, $L >> \rho$, then $R \approx 1$,

$T = 0$ and in the region $z \gg \rho$ we obtain

$$\frac{d\theta}{dz} = q + \frac{2}{\omega K_2} S_0 \tag{8.21}$$

In this circumstance the helix pitch becomes shorter under the light field action, since we assume $q > 0$. The additional rotation of the director

$$\delta\theta(z = 0) = -L\delta\left(\frac{d\theta}{dz}\right) \approx -qL\frac{2S_0 n}{cK_2 q^2} \tag{8.22}$$

Here we used the Bragg condition $\omega n/c = q$ and $S_0 = cn|E_-(z = 0)|^2/8\pi$.

In the other statement of the problem the director is free at the boundary $z = L$. Then

$$\left.\frac{d\theta}{dz}\right|_{z=L} = q \Rightarrow \frac{d\theta}{dz} = q - \frac{2}{\omega K_2}\left(-S_0 T + S_-^{(z)}(z)\right) \tag{8.23}$$

i.e. the helix pitch increases in this situation, since for all z we have $S_-^{(z)}(z) \geqslant S_0 T$. At $L >> \rho$ we have $T \approx 0$, so that

$$\frac{d\theta}{dz} = -\frac{2}{\omega K_2} S_-^{(z)}(z) + q \tag{8.24}$$

i.e. in most parts of the cell the pitch remains unchanged and the change of the angle $\delta\theta(z = L)$ is equal approximately to

$$\delta\theta \sim -qL(2S_0 n/cK_2 q^2)(\rho/L) \tag{8.25}$$

All our conclusions have not been based on some specific dependence of $\theta(z)$ and $S_-^{(z)}$; we have used only the fact that when $\varepsilon_q << \varepsilon$ the waves can be separated both by propagation directions and by circular polarizations. The dependence $\theta(z)$ and $S_-^{(z)}(z)$ must be determined from the solution of the self-consistent system of equations (6.13) and (8.16).

8.4 GON and LIFT in an attenuating wave field

Let a broad light beam ($a_\perp >> L$) be incident on a cell with a NLC. If the cell is not very thin, it is necessary to take into account the field attenuation by the law

$$|E(z)|^2 = |E(0)|^2 \exp(-z/\rho) \tag{8.26}$$

caused by real absorption and scattering. The last effect, as it is known, is especially strong in LC; typically $\rho \leqslant 10^{-1}$ cm. In this case the following equation is to be solved for the GON:

$$\frac{d^2\theta}{dz^2} = A\exp\left(-\frac{z}{\rho}\right); \qquad A = \frac{\varepsilon_a \sin\alpha\cos\alpha|E|^2}{8\pi K_i} \tag{8.27a}$$

and in the problem of the LIFT threshold

$$\frac{d^2\theta}{dz^2} + b^2\theta\exp\left(-\frac{z}{\rho}\right) = 0, \qquad b^2 = \frac{\varepsilon_a\sqrt{\varepsilon_\perp}S}{c\varepsilon_\parallel K_3} \tag{8.28a}$$

In both cases we assume strong anchoring: $\theta(z = 0) = \theta(z = L) = 0$. The solution of Eq. (8.27a) has the form

$$\theta(z) = A\rho^2\left[1 - \frac{z}{L} + \frac{z}{L}\exp(-L/\rho) - \exp(-z/\rho)\right] \tag{8.27b}$$

It may be reduced to the expressions obtained earlier at $\rho \to \infty$. At $L >> \rho$ the maximum of $\theta(z)$ is achieved at the point $z \approx \rho\ln(L/\rho)$ and equals $\theta_{max} \approx b^2\rho^2$. In other words, the Frank energy is determined here by the quantity $K_i\theta^2_{max}/\rho^2$.

The solution of Eq. (8.28a) for the LIFT has the form

$$\theta(z) = c_1 J_0(2b\rho e^{-z/2\rho}) + c_2 Y_0(2b\rho e^{-z/2\rho}) \tag{8.28b}$$

where Y_0, J_0 are Bessel functions of zeroth order. The LIFT threshold is determined by the existence of a nontrivial solution of (8.28b), satisfying the boundary conditions at $z = 0$ and $z = L$.

A transcendental equation characterizing the incident beam power density follows therefrom

$$J_0(2b\rho)Y_0(2b\rho e^{-L/2\rho}) = \mathcal{J}_0(2b\rho e^{-L/2\rho})Y_0(2b\rho) \tag{8.29}$$

At $\rho \gtrsim L$, the threshold value is $b_{th} \sim L$, so that $2b\rho >> 1$ and the asymptotic expansions of $\mathcal{J}_0(z)$ and $Y_0(z)$ at $z \to \infty$ can be used; then

$$b_{th}^2 = \left(\frac{\pi}{L}\right)^2 \left(1 + \frac{L}{2\rho}\right) \quad (8.30)$$

This expression gives the former result $b_{th}^2 = (\pi/L)^2$ at $\rho >> L$. The first nonvanishing correction in (8.30) corresponds to equating the former threshold value to one averaged over the cell intensity. In the other limit $\rho << L$, the threshold is determined by the condition $2b_{th}\rho \approx j_{01} \approx 2.4$, where j_{01} is the first zero of the Bessel function, $\mathcal{J}_0(j_{01}) = 0$. Thus, when the cell thickness increases from $L << \rho$ to $L >> \rho$, the LIFT threshold decreases at first, $b_{th}^2 = (\pi/L)^2$, after which it becomes constant $b_{th}^2 \approx 1.44\ \rho^{-2}$.

9 HYSTERESIS AND OPTICAL BISTABILITY

The effects of hysteresis, bistability and multistability in nonlinear optics and quantum electronics are of great current interest [37,38]. These effects consist of the possibility for the system to be in one of several stable states depending on the pre-history for a given input parameters of the light fields. In this way one can expect the creation of optical transistors, switches, power limiters and similar devices. The great optical nonlinearity of LC allows one to model these effects at very low power levels. In the present Chapter a number of particular schemes are discussed where optical bistability is or can be observed.

9.1 Hysteresis of LIFT

It has been noted in § 5.4 that for a broad beam ($a_\perp >> L$) of normal incidence on a homeotropic cell hysteresis of the LIFT can be realized if the NLC constants satisfy a definite condition. This condition, however, is fulfilled not for all NLC.

The magnetic field $\mathcal{H} = \mathcal{H}\mathbf{e}_z$ does not induce the Friedericksz transition but, on the contrary, aids the walls to hold the homeotropic orientation, if the NLC has a positive magnetic anisotropy, $\chi_a > 0$. Therefore, the LIFT threshold increases if a $\mathcal{H} = \mathcal{H}\mathbf{e}_z \neq 0$ is switched on. It is, however, important that the magnetic field allows one to obtain the LIFT hysteresis even in the media which do not possess it at $\mathcal{H} = 0$. The quantitative consideration of this effect requires the solution of the stationary equation

$$(K_1\sin^2\theta + K_3\cos^2\theta)\frac{d^2\theta}{dz^2} - (K_3 - K_1)\sin\theta\cos\theta\left(\frac{d\theta}{dz}\right)^2 + \frac{S}{S_{th}}\left(\frac{\pi}{L}\right)^2\frac{\sin\theta\cos\theta}{(\varepsilon_\perp + \varepsilon_a\cos^2\theta)^{3/2}} - \chi_a\mathcal{H}^2\sin\theta\cos\theta = 0 \quad (9.1)$$

Here S is the incident beam power density, $S_{th} = (c\varepsilon_\parallel K_3/\varepsilon_a\sqrt{\varepsilon_\perp})(\pi/L)^2$ is the LIFT threshold in the absence of the magnetic field. Eq. (9.1) differs from (5.25) by the term proportional to $\mathcal{H}^2$. Multiplying (9.1) by $2d\theta/dz$ and integrating over z, it becomes possible to write down the solution in quadratures; compare with the analogous procedure for (5.27). Let us write down the modified value of the LIFT threshold

$$S_{th} = S_{th}(\mathcal{H} = 0)(1 + \xi); \qquad \xi = \frac{\chi_a\mathcal{H}^2}{K_3}\left(\frac{L}{\pi}\right)^2 \quad (9.2)$$

Assume that at a small excess over the threshold, the quantity $\theta_m = \theta(z = L/2)$ is small, $\theta_m << 1$. Then the calculations analogous to the ones carried out in § 5.4, yield

$$\theta_m^2 = 2\frac{1+\xi}{V}\frac{S - S_{th}}{S_{th}}; \qquad V = 1 - \frac{9\varepsilon_a}{4\varepsilon_\parallel} - \frac{K_3 - K_1}{K_3} - \frac{9\varepsilon_a}{4\varepsilon_\parallel}\xi \quad (9.3)$$

If the parameter V is positive and is not very small, $V \sim 1$, then, $|\theta_m|$ increases by the square root law, $|\theta_m| = \text{const}\sqrt{S - S_{th}}$ with the excess over the threshold. For a small value of $V > 0$, the constant in this law increases. If $V < 0$ and $S > S_{th}$ the formal expression for θ_m becomes purely imaginary; this means that in the corresponding integral expansion we should not be restricted to terms of the order of θ_m^2 even for an infinitesimal excess over the threshold. The determination of the trend of an equilibrium curve (see Figure 15) requires numerical integration. For the illustration of the situation by analytic methods one can take into account terms of the order of θ_m^4 and then obtain the solution for θ_m in the form

$$\theta_m^2 = \frac{-V \pm \sqrt{V^2 - 8c_1c_2}}{c_1} \quad (9.4)$$

where c_1 is a complicated combination of the NLC constants, $c_2 = 1 - (S/S_{th} - \xi)^{1/2}$. This expression shows, that the difference between the values of the LIFT switching on power (S_{th}) and switching off power (S_2), see Figure 15, increases approximately by the law $S_{th} - S_2 \sim V^2$.

Thus, the application of a magnetic field supporting homeotropic orientation allows one to obtain a hysteresis of the LIFT and to increase the gap between the power values for switching on and switching off. Let us make numerical estimates for MBBA which does not show hysteresis of the LIFT without a magnetic field. If we take $\varepsilon_{\parallel} = 3.06$, $\varepsilon_a = 0.7$, $\chi_a = 0.97.10^{-7}$, $K_3 = 7.5.10^{-7}$ dyne, $L = 10^{-2}$ cm, then $S_{th} = 200$ W/cm^2.

The magnetic field must be greater than the value $\mathcal{H} = 650$ G to obtain LIFT hysteresis; such a field increases the threshold of the LIFT switching on to the value 320 W/cm^2.

9.2 Hysteresis in cholesterics

The important property of CLC is the existence of the selective Bragg reflection. The change of the helix period of a CLC illuminated by light fields can lead, due to the one or other mechanism, for example, to the reflection switching on (if it was not for weak fields) or switching off (if it were). One can imagine a number of schemes where such a hysteresis must take place.

Consider, for example, a cell with planar structure of a CLC placed between two transparent glasses Π_1 and Π_2 and besides, a thin layer A of a weakly absorbing medium is placed in front of the CLC layer, Figure 21. If the absorption $1 - \exp(-\sigma_A L_A)$ is not great, $\sigma_A L_A \leqslant 1$, its influence on the transmitted intensity can be neglected. It is possible, however, to get a smaller absorption and heating in the CLC itself. In this case the established temperature of the system (absorber + CLC) will be determined by the relation $\delta T \simeq (S^{(z)} + S^{(-z)})\,\sigma_A L_A \tau/\rho C_P$. Here $S^{(z)}$ and $S^{(-z)}$ are the power densities of the light fluxes propagating in the layer A along the directions $+z$ and $-z$ respectively, $\rho C_P(J/\text{cm}^3\text{deg})$ is the heat capacity of the unit volume, $\tau = (L/\pi)^2/\chi$ is the establishment time of the stationary temperature.

Assume that the CLC does not reflect initially the incident radiation, but can reflect it after heating by ΔT_0 degree. If the incident light power density S is smaller than the quantity S_1,

$$S_1 = \rho C_P \Delta T_0/\sigma_A L_A \tau \qquad (9.5)$$

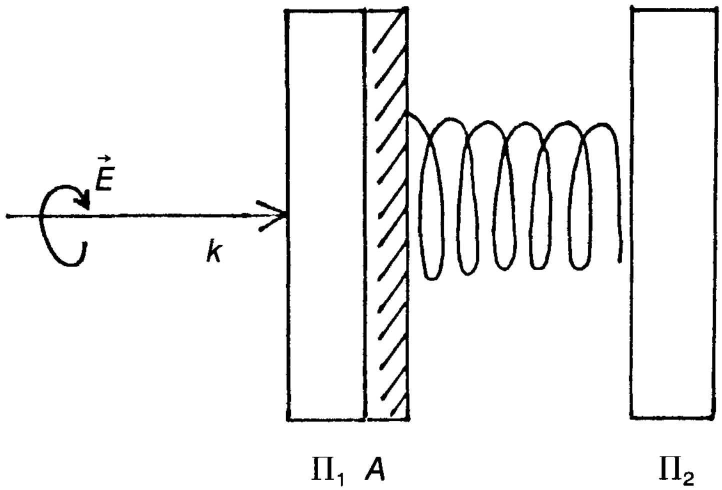


FIGURE 21 A bistable device utilizing the thermally induced change of the *CLC* helix pitch. There is a Grandjean structure of a *CLC* between the transparent plates Π_1 and Π_2, A is a thin absorbing layer. Circularly polarized light is incident normally to the plates.

the heating does not lead to the reflection. If the incident flux exceeds S_1, the reflection caused by heating switches on and two waves propagate oppositely through the absorber with total intensity almost twice as high than the incident light intensity; the absorbed power is doubled too. Subsequent decrease of the power leads to the reflection switching off when $S \approx S_1/2$. If we take $L_A \sim 500$ μm, then $\tau \sim 0.25$ s; and for the values $\sigma_A L_A = 0.25$, $\Delta T_0 = 1°C$ and $\rho C_P = 1$ $J/\text{cm}^3\text{deg}$ we obtain the switching on power density $S_1 \approx 16$ W/cm^2.

In the scheme represented on Figure 22 the hysteresis can be obtained also out of the Bragg resonance, due to the dependence on temperature of the light polarization rotation in the cell with a CLC, with the use of a polarizer, analyser and a mirror. Let the CLC layer rotate the polarization plane to an angle $\phi = \phi_0 + \phi_2(S^{(z)} + S^{(-z)})$, where $\phi_2 = \partial\phi/\partial S$ is conditioned by the light absorption thermal effects. Then $S^{(-z)} = S^{(z)} \sin^2\phi = S \sin^2\phi$, where S is the incident flux. The stationary state is determined by the solution of the transcendental equation

$$1 + \sin^2\phi = \frac{\phi - \phi_0}{\phi_2 S} \tag{9.6}$$

A simple geometrical construction shows that at $|\phi_2 S| > 1$ this equation has more than one root corresponding to multistability. The

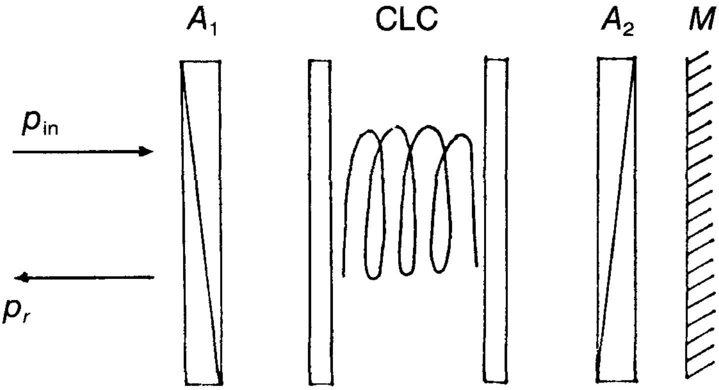


FIGURE 22 A scheme for hysteresis obtaining out of the Bragg resonance due to the temperature dependence of the light polarization plane rotation; A_1, A_2-polarizers, M-mirror.

direct orienting action of the light wave field can also lead to a CLC helix pitch change, apart from the thermal influence (compare with § 8.3). Let, for example, the boundary $z = 0$ strictly fix the director and the boundary $z = L$ be free, and let there be a weak power radiation exactly in the Bragg resonance. Then, if the power increases, the pitch near the input $z = 0$ increases, the field penetrates into the CLC deeper and finally the CLC helix goes out of the resonance almost in the whole volume. When the power decreases, the reflection switching on must occur for smaller flux than the switching off for increasing power; this corresponds to the hysteresis. Numerical estimates for a CLC with $\varepsilon_a/\varepsilon \approx 0.1$, $K_2 = 10^{-6}$ dyne, $\lambda = 1$ μm give the switching on critical power $S \sim 10^6$ W/cm^2 and the establishment time $\tau_1 \sim \gamma L^2/\pi^2 K_2 \sim 0.02$ s for $\gamma = 0.1$ poise and $L = 15$ μm. Unfortunately even for a weak absorption $\sigma \sim 10^{-1}$ cm^{-1} in CLC, about 10^5 W/cm^3 thermal energy will be released for such fluxes, so that the temperature must increase to $\Delta T = S\sigma\tau_2/\rho C_p \approx 20°$ if the thermal equilibrium establishment time is $\tau_2 \sim L^2/\pi^2\,\chi \sim 2.10^{-4}$ s. Such a large change of temperature will considerably shift the Bragg resonance or even will melt the mesophase.

The above discussed and some other schemes of optical bistability have been discussed theoretically in [106].

9.3 Fabry-Perot resonator with a nonlinear NLC-cell

One of the most popular schemes of multistable optical devices is the Fabry-Perot resonator (RFP), inside of which a medium is placed with a strong dependence of the refractive index on the intensity. If the frequency of a low power light is outside the RFP resonance transmission band, the light power in the nonlinear medium is even smaller than the incident one and the nonlinear effects are absent. The refractive index change, when the power increases leads to a shift of the resonance frequency. If the operating point is chosen conveniently, the RFP transmission increases and the light power inside the medium increases more stronger. The RFP high transmission may then stay during the decrease of the input power. This namely corresponds to the hysteresis in the plot of the transmitted power versus the incident one.

Let us write some related formulae. We assume for simplicity that all the losses are due to incomplete reflection of the mirrors, $R_1 = R_2 = R < 1$. Then the power inside the RFP is connected with the input power S_0 by the relation

$$S^{(z)} + S^{(-z)} = S_0 \frac{1 - R^2}{(1 - R)^2 + 4R\sin^2\varphi} \tag{9.7}$$

where φ is the phase shift connected with the light propagation from one mirror to the other. As in § 9.2, we can write phenomenologically $\varphi = \varphi_0 + \varphi_2(S^{(z)} + S^{(-z)})$ and then the stationary relations are determined by the solution of the equation

$$\frac{\varphi - \varphi_0}{\varphi_2 S_0} = \frac{1 - R^2}{(1 - R)^2 + 4R\sin^2\varphi} \tag{9.8}$$

Practically just after the discovery of the GON in cells with NLC at light oblique incidence, many authors expressed the natural idea to put such a cell in the RFP for an optical bistable device construction, Figure 23. The corresponding experiment [39] has been carried out with the use of a homeotropic cell with a NLC PCB of thickness $L = 50$ μm inserted at an angle 20° to the resonator axis (inside the NLC), so as to realize the GON regime. Argon laser was used, $\lambda = 0.5145$ μm, of power up to 1 W. The distance between the mirrors was 4 cm. Clearly expressed hysteresis was observed at initial phase (φ_0) conveniently chosen: high transmission (T_1) switching on for the input power ~0.36 W (input power density in the focused beam 700 W/cm^2) and its switching off to level T_2 for the power ~0.12 W with the contrast $T_1/T_2 \sim 5$. Switching on time was 2–3 s; switching off time was 4–6 s. When the inclination angle decreases to 10° the power density necessary for the GON observation increases to 2000 W/cm^2.

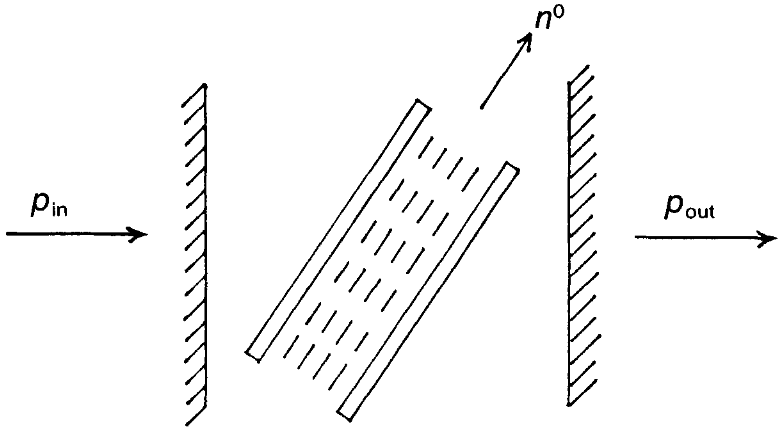


FIGURE 23 Fabry-Perot resonator with a cell of planarly oriented *NLC* operating in the GON regime.

An RFP with a homeotropic cell, where the director is exactly parallel to the RFP optical axis, Figure 24 must have very specific properties. Firstly, in such a geometry it is possible to combine the mirrors with the cell walls and, thus, to decrease considerably the device sizes up to the thickness $\lesssim$ 1 mm. Secondly, from the point of view of the physics process, a quite unusual, for nonlinear optics, phase shift dependence on the power inside the resonator must operate here—namely the threshold behaviour, which is specific for the LIFT, see, for example, the expression (5.31). If the RFP is tuned to have a maximal transmission $\varphi_0 = m\pi$ at the low intensity, the inside power is

$$S^{(z)} + S^{(-z)} = S_0 \frac{1 + R}{1 - R} \approx S_0 \frac{2}{1 - R} \tag{9.9}$$

For example, at $R \approx 0.8$, the total power inside the RFP increases approximately 10 times relative to the incident one, and at $R = 0.98$ – 100 times. Namely, by the same factor will decrease the value of the input power density S_0 at which the threshold $S_{th} = \pi^2 c\varepsilon_{\|} K_3/\varepsilon_a \sqrt{\varepsilon_\perp}\, L^2$ will be achieved inside the resonator.

If the resonator is tuned to the minimal transmission, the inside power density is equal to

$$S^{(z)} + S^{(-z)} = S_0 \frac{1 - R}{1 + R} \approx S_0 \frac{1 - R}{2} \tag{9.10}$$

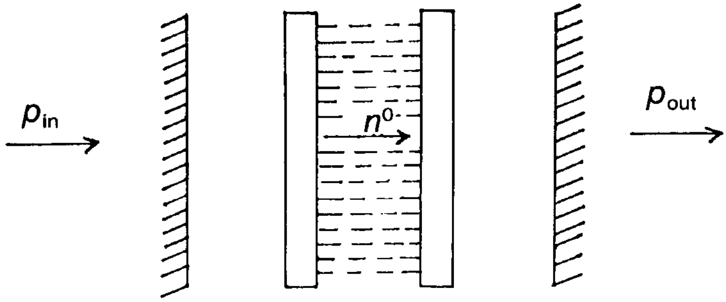


FIGURE 24 Light-induced Friedericksz transition in a resonator Fabry-Perot.

i.e. now it is smaller exactly by the same factor as the input one.

Consider now the behaviour of such a RFP at $\varphi_0 = m\pi$, if the input power is gradually increased. For the value $S_0 = S_1$, where

$$S_1 = 0.5(1 - R)S_{th} \tag{9.11}$$

the LIFT threshold is achieved. If the power S inside the RFP becomes larger than S_{th} even by a very small amount, then, according to Eq. (5.31), a considerable shift $\delta\varphi \approx (2\pi L/\lambda)\varepsilon_a(S - S_{th})/VS_{th}$ appears since in typical circumstances $2\pi L/\lambda = 10^2 - 10^3$, $\varepsilon_a \sim 1$ and $V \sim 0.3$. As a result, the RFP is shifted out of resonance and such a phase shift is established, for which the power density inside the FPR is to a great accuracy equal to the threshold value S_{th}. At this point the output power will be equal to $S_{out} \sim S_1$. This regime becomes invalid only when the power inside the RFP will be higher than the threshold even for the RFP transmission minimum. This occurs at $S_0 = S_2$, where

$$S_2 = 2S_{th}/(1 - R) \tag{9.12}$$

Thus, in a large interval of the incident power S_0

$$S_1 = \frac{1 - R}{2} S_{th} \leqslant S_0 \leqslant \frac{2}{1 - R} S_{th} = S_2 \tag{9.13}$$

the transmitted wave will be stabilized to a great accuracy on the level S_1. In the considered examples at $R = 0.8$ the output power is stabilized in the interval from S_1 to 100 S_1 and at $R = 0.98$- in the interval from S_1 to 10^4 S_1.

It is very important that a thin homeotropic cell ($L = 10$ μm, for example) possess a high transparency with respect to the normally incident wave. One can hope, therefore, that the mirrors' high reflection coefficient (for example $R \approx 0.98$) can be used as the cell walls. The increase (as L^{-2}) in the magnitude of S_{th} is here compensated to some extent by the gain factor $2/(1 - R)$. The essential advantage of thin cells is in the small value of the relaxation time.

Let us make numerical estimates. The value of S_1 for OCBP is $S_1 \approx 90$ W/cm^2 at $R = 0.8$ and $L = 100$ μm.

The discussions carried out above apply to the stationary picture and do not take into account fluctuations. Unfortunately just near the threshold of the Friedericksz transition (of any type) the relaxation time and the intensity of the thermal fluctuations increase without limit. In our point of view, the experimental investigations of such a cell behaviour in light fields are of great interest.

The multistability effects must be expressed also for a broad beam of oblique incidence to the RFP in the form of Figure 24.

The optical bistability of beams of homogeneous transverse intensity distribution was discussed above.

The beam transverse inhomogeneity leads to self-focusing—to the change of angular divergence. In particular, choosing a convenient geometry one can obtain a decrease of the central spot size in the far zone, if the power is increased. If we install a small mirror at the axis in the far zone, that will reflect the light back to the GON-cell, then such a system can demonstrate hysteresis effects. For weak power, the light spot in the far zone is large and the part of reversed light is small. The reflected wave gives additional contribution to the self-focusing effect, if the self-focusing has considerably decreased the spot size, in the mirror region. Then, if the power is decreased, the self-focusing regime is limited at a lower power than the power required for its switching on. That scheme was proposed for nonlinearity of a general type in [38]. The GON in homeotropic cell (L = 60 μm) was used to obtain bistability in that scheme in [39]; the nematic PCB was illuminated at an incidence angle 20°. The switching on was observed at a power level of about 0.3 W (500 W/cm^2), and the switching off—at 0.2 W (330 W/cm^2); the relaxation time was 1 – 2 s.

Since the nonlinearity of LC 1) is very large, 2) can have a threshold and 3) is easily saturated, there are reasons to expect multistability in many different schemes. We advise experimentalysts engaged in LC nonlinear optics, to verify the system behaviour when the power increases and decreases in order to detect multistability even in cases where its appearance is not clear beforehand.

10 ORIENTATIONAL NONLINEARITY IN THE PRESENCE OF OTHER EXTERNAL INFLUENCES

The previous considerations of theory and experiment concern the case where the LC unperturbed state is fixed by the cell walls orienting action. The superposition of external fields—static electric and magnetic, gradient of velocity, temperature and so on,—can, first of all, change the initial distribution of the director. The symmetry in several cases is such that the distribution of $\mathbf{n}(\mathbf{r})$ is either unchanged or is changed above some threshold only. In all cases, however, the external fields change the system "rigidity," i.e. its response to the external influences; in the case we are interested in—the response to the light fields orienting action. In particular, the system rigidity decreases, i.e. its susceptibility increases near the threshold of various instabilities caused by d.c. fields.

10.1 GON near the Friedericksz transition induced by static fields

Consider a cell with a NLC, the walls of which rigidly fix the homogeneous homeotropic orientation. Let a potential difference V corresponding to the static or radiofrequency field $\mathscr{E} = \mathbf{e}_z V/L$ be applied to the cell walls with the aid of electrodes. If the dielectric constant at the corresponding frequency has a negative anisotropy, $\varepsilon_\parallel^0 - \varepsilon_\perp^0 = \varepsilon_a^0$, then for the value $\mathscr{E}_{\text{th}}^2 = 4\pi^3 K_3/\varepsilon_a^0 L^2$ the Friedericksz transition takes place. Let a broad light beam ($a_\perp >> L$) be incident on such a cell at an angle α (inside the medium). In the absence of a radiofrequency field the GON is realized in this geometry. Let us obtain the expressions determining the GON behaviour near the Friedericksz transition in a radio-field. Taking $\mathbf{n}(z,t) = \mathbf{e}_z\cos\theta(z,t) + \mathbf{e}_x\sin\theta(z,t)$ and the light wave field $\mathbf{E} = E(\mathbf{e}_x\sin\alpha - \mathbf{e}_z\cos\alpha)$, the Euler-Lagrange-Rayleigh variational equations yield

$$\gamma\frac{\partial\theta}{\partial t} = (K_1\sin^2\theta + K_3\cos^2\theta)\frac{\partial^2\theta}{\partial z^2} - (K_3 - K_1)\sin\theta\cos\theta\left(\frac{\partial\theta}{\partial z}\right)^2$$

$$- \frac{\varepsilon_a^0}{8\pi}\sin 2\theta\,\mathscr{E}^2 - \frac{\varepsilon_a}{16\pi}\sin\alpha\cos\alpha\,|E|^2 \quad (10.1)$$

The main role, both for the GON and for the FT, belongs to lowest spatial harmonic perturbation of the director

$$\theta(z,t) = \theta_m(t)\sin(\pi z/L) \quad (10.2)$$

Below the FT threshold, or for not far above the FT threshold, one can obtain from (10.1) and (10.2)

$$\frac{L^2}{\pi^2 K_3}\gamma\frac{\partial\theta_m}{\partial t} = C\theta_m - \frac{1}{2}V\theta_m^3 + B, \tag{10.3}$$

$$C = \mathscr{E}^2/\mathscr{E}_{\text{th}}^2 - 1, \tag{10.4}$$

$$V = 1 + \frac{\varepsilon_a^0}{\varepsilon_\parallel^0} - \frac{K_3 - K_1}{K_3}, \tag{10.5}$$

$$B = \frac{\varepsilon_a \sin\alpha\cos\alpha\, L^2\, |E|^2}{8\pi^3 K_3} \tag{10.6}$$

The quasistatic radiofrequency field modification caused by the director deformations is taken into account for the parameter V calculation (see for more details [41]). For radio-fields $V > 1$, the cubic term more rigidly fixes the director and therefore the domain of validity of Eq. (10.3) is considerably large here. The formulae for the magnetic field induced FT threshold are obtained by the replacement $\varepsilon_a^0\mathscr{E}^2/4\pi \rightarrow \chi_a\mathscr{H}^2$, and the constant $V = K_1/K_3$ in this case.

Below the threshold $C < 0$ and in the absence of light fields there is a single solution $\theta_m = 0$. Then, to first order in the light intensity the term $\sim\theta_m^3$ can be neglected and

$$\theta_m = \frac{B}{|C|}(1 - e^{-|C|\Gamma t}), \qquad \frac{K_3}{\gamma}\left(\frac{\pi}{L}\right)^2 = \Gamma \tag{10.7}$$

At $C = -1$ (the absence of the radiofrequency field) Eq. (10.7) coincides with the previous result from § 4.3. The expression (10.7) shows that near the FT threshold (but below it) the GON constant increases by a factor $|C|^{-1}$; the establishment time becomes longer by the same factor.

There are an unstable ($\theta_m = 0$) and two stable ($\theta_m = \pm\sqrt{2C/V}$) equilibrium positions above the FT threshold at $C > 0$. Then, to a linear accuracy in $|E|^2$, we have near the stable point

$$\theta_m = \pm\left(\frac{2C}{V}\right)^{1/2} + \frac{B}{2C}(1 - e^{-2C\Gamma t}) \tag{10.8}$$

The GON constant and the establishment time (relative to the problem without radiofrequency field) are increased by the factor

$(2C)^{-1}$ for slightly above the threshold, $C > 0$. Thus, near the FT the nonlinear optical susceptibility shows the Curie-Weiss behavior: as $|C|^{-1}$ below the threshold and as $(2C)^{-1}$ above the threshold. The case of far above the FT threshold requires more accurate equations than (10.2), (10.3); we do not dwell on this in more detail.

The experimental investigation of the GON constant dependence on C near the radio frequency FT, has been carried out in the paper [42]. A cell of thickness $L = 50$ μm had the director homeotropic anchoring on the walls. The FT took place when an electric tension at frequency 200 Hz was applied. Unfortunately the FT threshold was rather spread out. The GON was observed in the absence of the field $\mathscr{E}$: $N = 3$ fringes of external self-focusing and the divergence increase to the values $7.5.10^{-2}$ rad were registered for a light beam power of 30 mW. The divergence and fringe number increase when the applied field $\mathscr{E}$ is close to the threshold value; also the GON-self-focusing establishment time increases, see Figure 25. According to the theory, the diagrams of the quantities $\delta\varphi^{-1}$ and τ^{-1} depending on C near the FT threshold must be a) straight lines, b) passing through the coordinate origin and c) having twice as different tangent of the inclination angle. The experiment is in good agreement with the points a), b) and c) of the theory for the time τ; somewhat worse agreement for points a) and c) for the phase shift $\delta\varphi$.

In the experiment described the GON constant was increased, due to approaching the radio frequency FT threshold, but only by a factor of 2.1; the reasons obviously were the FT threshold width caused by transverse inhomogeneity and the cell instability. The radio frequency field must very strongly fix the director and the GON constant must tend to zero at $\mathscr{E}^2 >> \mathscr{E}^2_{th}$. Experimentally at $\mathscr{E}_z^2 \gtrsim 2\mathscr{E}^2_{th}$, the GON

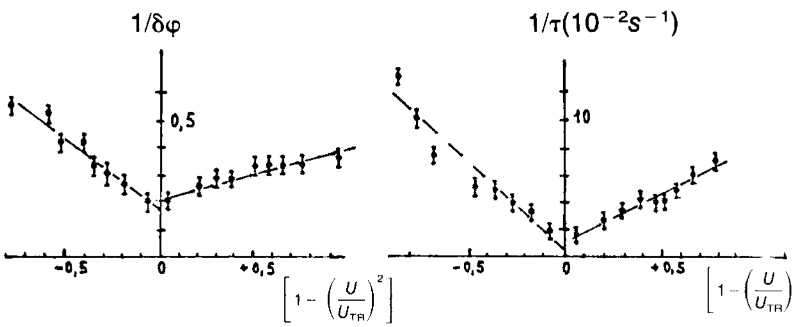


FIGURE 25 The increase of the phase nonlinear shift (δy) and the GON establishment time ($\mathscr{T}$) near the threshold of quasistatic field induced Fredericksz transition [42].

constant was approximately half of the value of GON for the cell without the field $\mathscr{E}$.

Very interesting experimental results are presented in the paper [43], where the GON constant was registered by the self-diffraction method and the FT was induced by a magnetic field in a cell with a homeotropic aligned MBBA. The author observed a very abrupt threshold ($\Delta\mathscr{H} \sim 0.5$ G) in the diffracted waves appearance. The value of the threshold field turned out to be suprisingly small: $\mathscr{H}_{\mathrm{th}} \sim 175$ G for $L = 50$ μm and $\mathscr{H}_{\mathrm{th}} = 70$ G for $L = 100$ μm (if we have correctly understood the notation 70 G in [43]). If one uses the standard theory of the FT in a magnetic field with the director strong anchoring on the boundaries, see [5], then the value of the threshold at $K_3 = 7.5.10^{-7}$ dyne and $\chi_a = 10^{-7}$ is $\mathscr{H}_{\mathrm{th}} = (\pi/L)\sqrt{K_3/\chi_a} = 1700$ G at $L = 50$ μm and $\mathscr{H}_{\mathrm{th}} = 850$ G at $L = 100$ μm.

The cell with a nematic SCB of thickness $L = 250$ μm and with the homeotropic orientation ($\mathbf{n} = \mathbf{e}_z$) inducing walls has been used in the experiment [44]. A very strong magnetic field $\mathscr{H} = 1450$ G was applied on the sample at an angle $\beta_{\mathscr{H}} = 60° =$ to the z axis. As a result the director was considerably inclined from the undisturbed direction acquiring quite a complicated profile $\theta(z)$; by a numerical calculation $\theta(z = L/2) = 53°$. Two plane light waves having a small angle $\beta \sim 10^{-2}$ rad to each other were incident practically normal to the cell. In such a geometry the GON is absent without the magnetic field and self-diffraction is not observed. If the magnetic field is switched on, the director acquires an inclination with respect to the electric field in a considerable part of the cell and the GON arises.

Up to the ± 6 orders of diffraction was observed. For the intensities of each of the two interfering beams 100 w/cm authors of [44] have observed up to twelve (± 6) diffracted beams.

Unfortunately an accurate quantitative calculation of the director GON-perturbation is very difficult here because of the complicated initial profile of $\theta(z)$. However the common behaviour of the diffracted beam intensity dependence on the incident waves intensity (parameter η in expression (2.34)) is in good correspondence with the picture of the phase modulation in the form (2.35). Further, the authors have investigated the diffraction efficiency dependence on the period $\Lambda = \lambda/\beta$ of the interference picture. In the experiment [43] the self-diffraction efficiency decreases with the decrease of the interference grating period Λ.

In the paper [45] a cell with a planarly oriented nematic OCB of thickness 150 μm has been used. A quasistatic electric field $\mathscr{E} = \mathbf{e}_z\mathscr{E}$

was destabilizing the planar structure ($\varepsilon_a^0 > 0$ for OCB) beginning from the threshold voltage $V_{\text{th}} = 1V$. The structure remains planar at $V < V_{\text{th}}$, and for the light beam, normally incident to the cell, the GON was absent. The structure of $\theta(z)$ acquires a complicated profile and the GON arises with a very strong self-focussing at $V > V_{\text{th}}$. The further increase of voltage leads to the more rigid orientation of director by the quasistatic field, so that at $V^2 \gtrsim 2V_{\text{th}}^2$ the GON constant begins to decrease. An interesting aspect of the experiment carried out in [45] was the use of not only a weak but also a comparatively strong light beam (up to 75 mW). In the last case the optical nonlinearity is partially attenuated and the angular divergence decrease, dependent on V, takes place at $V^2 \gtrsim 3V_{\text{th}}^2$. Note that in the experiment [45] the GON was absent below the threshold of the static FT and is moderately increased above the FT threshold, because of the light wave normal incidence. Unlike this, the wave was incident obliquely in the experiment [42]; this leads to the GON without the field and to the GON increase by the Curie-Weiss law near the FT threshold. The main difference, thus, consists not of the planar orientation versus the homeotropic one, but of the light wave oblique incidence to the director relative to the normal.

An homeotropic cell with MBBA of thickness 70 μm has been used in one of the first experiments [46] on the LC orientational nonlinearity. A magnetic field $\mathcal{H} = \mathbf{e}_x\mathcal{H}$ of a value ~70 G (if we correctly understand the notation 70 G used by the authors of [46]) induced the FT. The cell was illuminated by an argon laser radiation ($\lambda = 0.488$ μm) consisting of two plane waves incident practically normal to the cell and at an angle about 0.3° with each other. The GON was absent below the threshold of the magnetic FT: no changes have been experimentally registered in the beams transmitted through the cell. The reference wave of power density $S_1 \approx 2$ W/cm^2 recorded a phase grating with a weak signal wave even for a small excess over the threshold. The reference wave diffraction on this grating, besides other effects, amplified the signal beam (compare with expressions (2.35)). The weak beam was amplified approximately by 11 per cent in the experiment. The authors of [46] have compared their results with the calculations of one of the first theoretical papers on the orientational nonlinearity [47] and have found good agreement.

There is a number of other physical situations where the initially present GON is increased near the threshold of some instability. The director instability, for example, can arise from an inhomogeneous hydrodynamic flow. Depending on the particular conditions the earliest instability may be either an homogeneous or a periodical one (rolls). These instabilities can arise in LC with low conductivity under an electrical potential difference. A great number of instabilities takes place in the presence of temperature gradients.

Light field represents a unique possibility to measure the system's response to periodic space distortions of period Λ, which can be precisely tuned within the limits from $\Lambda >> L$ to $\Lambda = \lambda/2n$, here λ is the light wavelength. It is sufficient to illuminate the cell by the field of two interfering plane waves and to measure the self-diffraction parameters.

In some cases the unstable mode of the perturbation corresponds not to the simple exponential gain but to the evolution with a quasiharmonic oscillations in time, $\sim\exp(\Gamma' t) \cdot \sin(\Gamma'' t)$. The strongest increase of the susceptibility then will take place for illumination by two waves with a shifted frequency, $\omega_1 - \omega_2 = \Gamma''$. A number of instabilities available to GON-investigations is realized in deformed LC-structures. Let us note several examples: planarly twisted nematic (the cell $0 \leqslant z \leqslant L$ and the director has the form $\mathbf{n} = \mathbf{e}_X \cos\varphi(Z) + \mathbf{e}_y \sin\varphi(z)$, $\varphi(0) = 0$, $\varphi(L) \approx \pi$); hyperhybrid aligned nematic ($\mathbf{n} = \mathbf{e}_X \cos\theta(z) + \mathbf{e}_Z \sin\theta(z)$, $\theta(0) = 0$, $\theta(L) \approx \pi$); a cholesteric, homeotropically aligned at the plates of the cell ($q_0 L \approx \pi$) and lots of other ones. GON near the threshold of electrohydrodynamic instabilities was considered in the paper [107].

10.2 External field influence on LIFT

Assume that a broad light beam is incident on a cell with a homeotropic aligned NLC and, in addition, an external electric field $\mathcal{E} = \mathbf{e}_z \mathcal{E}$ and (or) magnetic field $\mathcal{H} = \mathbf{e}_z \mathcal{H}$ is applied. It is not difficult to check that the condition on the resulting FT threshold (which already is not purely induced by the light, electric or magnetic field) in this case takes the form

$$\frac{\varepsilon_a \varepsilon_\perp |E|^2}{8\pi\varepsilon_\parallel} = \tilde{K}_3 \left(\frac{\pi}{L}\right)^2 \tag{10.9}$$

$$\tilde{K}_3 = K_3 \left[1 + \frac{\chi_a \mathcal{H}^2}{K_3}\left(\frac{L}{\pi}\right)^2 + \frac{\varepsilon_a^0 \mathcal{E}^2}{4\pi K_3}\right] \tag{10.10}$$

Moreover, the problem of the LIFT *threshold* in the presence of $\mathcal{E}$ and $\mathcal{H}$ is reduced to the problem on the LIFT *threshold* without the fields $\mathcal{E}$ and $\mathcal{H}$ (see § 5.1) by the substitution $K_3 \to \tilde{K}_3$. The above-threshold structures, however, can be considerably different in the mentioned problems. In particular, the LIFT hysteresis must appear stronger if $\chi_a\mathcal{H}^2 > 0$, see § 9.1. The expression (10.9) for a broad light beam can be rewritten in the form

$$\frac{S_{\text{th}}}{S_1} = 1 + \frac{\chi_a}{|\chi_a|}\frac{\mathcal{H}^2}{\mathcal{H}_1^2} + \frac{\varepsilon_a^0}{|\varepsilon_a^0|}\frac{\mathcal{E}^2}{\mathcal{E}_1^2} \qquad (10.11)$$

where $\mathcal{H}_1$ and $\mathcal{E}_1$ are the corresponding threshold values of the quasi-static fields.

An experimental investigation of the external fields influence on the LIFT has been carried out in the paper [45]. A homeotropic cell of thickness $L = 150$ μm was investigated. An external field of frequency 10 kHz was applied. The argon laser radiation had a diameter of the waist spot of 80 μm. For the nematic OCB at $T = 35°$C $\varepsilon_a^0 = 6.8$ and the external field increased the LIFT threshold power; in the experiment up to 1.7 times. For MBBA $\varepsilon_a^0 = -0.4$ at $T = 30°$C a LIFT threshold power decrease was observed. Near the threshold of electric FT a threshold decrease by a factor 5 was successfully observed.

The exact comparison with the theory here is difficult since the beam diameter is considerably smaller than the cell thickness; in the latter case the effects of inhomogeneity make the LIFT threshold calculation essentially difficult without external fields. The authors of the paper [45] note a good accordance of the experimental results with the theoretical model obtained in the single-constant approximation for a beam of the flat-top type.

It seems to be very interesting to study the interaction of the LIFT and the transition between homogeneous and inhomogeneous state of CLC with homeotropic orienting walls at $qL \sim 1$. In the absence of the field such a cell has stable homogeneous orientation $\mathbf{n} = \mathbf{e}_z$ at $qL << 1$. At $qL \geqslant \pi(K_2/K_3)^{1/2}$ an instability arises and the director describes an inhomogeneous helix in the volume, as a result. It can be expected that both instabilities—the LIFT and the cholesteric one will help each other.

To consider this problem it is convenient to represent the director perturbations in the form $\mathbf{n}(z,t) \approx \mathbf{e}_z + \mathbf{e}_x\theta_x(z,t) + \mathbf{e}_y\theta_y(z,t)$. The linearized variational equations then take the form

$$-\gamma\frac{\partial\theta_x}{\partial t} + K_3\frac{\partial^2\theta_x}{\partial z^2} + 2K_2q\frac{\partial\theta_y}{\partial z} = -\frac{\varepsilon_a}{16\pi}(\theta_x|E_x|^2 + E_xE_z^* + E_x^*E_z), \qquad (10.12a)$$

$$-\gamma \frac{\partial\theta_y}{\partial t} + K_3 \frac{\partial^2\theta_y}{\partial z^2} - 2K_2 q \frac{\partial\theta_x}{\partial z}$$

$$= -\frac{\varepsilon_a}{16\pi} (\theta_y|E_y|^2 + E_y E_z^* + E_y^* E_z) \quad (10.12b)$$

with the boundary conditions $\theta_i(z = 0,t) = \theta_i(z = L,t) = 0$. In the problem of the LIFT threshold in a circularly polarized radiation field (of any sign of the circularity) or in a field of depolarized radiation, the right hand sides in (10.12) can be replaced by $(-\varepsilon_a\varepsilon_\perp/16\pi\varepsilon_\parallel)\theta_x|E|^2$ and $(-\varepsilon_a\varepsilon_\perp/16\pi\varepsilon_\parallel)\cdot\theta_y|E|^2$, respectively, where $|E|^2 = 2|E_x|^2 = 2|E_y|^2$. Then, using the linear combination $\theta_+ = \theta_x + i\theta_y$ and $\theta_- = \theta_x - i\theta_y$ it is possible to obtain the general solution of the system:

$$\theta_+(z,t) = \sum c_{m+} \exp\left(-\Gamma_m t + i\frac{K_2}{K_3}qz\right) \sin\frac{m\pi z}{L}, \quad (10.13a)$$

$$\theta_-(z,t) = \sum c_{m-} \exp\left(-\Gamma_m t - i\frac{K_2}{K_3}qz\right) \sin\frac{m\pi z}{L}, \quad (10.13b)$$

$$\Gamma_m = \frac{K_3}{\gamma}\left[\left(\frac{m\pi}{L}\right)^2 - \left(\frac{K_2 q}{K_3}\right)^2 - \frac{\varepsilon_a\varepsilon_\perp|E|^2}{32\pi\varepsilon_\parallel K_3}\right] \quad (10.14)$$

where c_{m+} and c_{m-} are arbitrary constants. The threshold (the change of the sign of Γ_m) is achieved first of all for $m = 1$:

$$|E|^2_{\text{th}} = \frac{16\pi\varepsilon_\parallel K_3}{\varepsilon_a\varepsilon_\perp}\left[\left(\frac{\pi}{L}\right)^2 - \left(\frac{K_2 q}{K_3}\right)^2\right] \quad (10.15)$$

Thus, near the threshold of cholesterical instability the LIFT threshold decreases

$$\frac{S_{\text{th}}}{2S_0} = \left[1 - \left(\frac{K_2 q}{K_3}\frac{L}{\pi}\right)^2\right] \quad (10.16)$$

where S_0 is the LIFT threshold in the linearly polarized beam ($2S_0$— in the unpolarized beam).

The problem becomes much more difficult for linear polarization of the incident beam. We represent the answer only at $0 < 1 - q/q_{th} << 1$:

$$\frac{S_{th}}{S_0} \approx \frac{8}{3}\left(1 - \frac{K_2}{K_3}\frac{qL}{\pi}\right) \tag{10.17}$$

Consider now the GON in the CLC-cell under consideration. The investigation of the corresponding solution of the system (10.12) shows that there also would be a strong increase of the GON constant near the point $q = q_{th}$ (at $q < q_{th}$):

$$\varepsilon_2(q) \approx 0.34\ \varepsilon_2(q = 0)(1 - q/q_{th})^{-1}$$

From our point of view it would be very interesting to investigate the influence of the cholesterical instability on the GON and LIFT. Detailed theoretical calculations have shown that the cholesterical transition itself must have a hysteresis.

11 NONLINEARITIES CONNECTED WITH ABSORPTION

We considered above the orientational nonlinearity of transparent LC, in which the light quanta are not absorbed. A natural question arises: from where is taken the energy necessary for the director reorientation? The answer is that in the average the light quanta slightly turn red in nonlinear processes. For example, if the intensity is switched on abruptly, the light passed through the GON-cell has a time dependence

$$E_{tr}(t) = E_{in}(t)\exp[i\varphi_0 + i\delta\varphi f(t) - i\omega_0 t] \tag{11.1}$$

where $f(t) = 1 - \exp(-\Gamma t)$. Since $\delta\varphi > 0$, it follows from (11.1) that the instantaneous frequency $\omega_{ins} = -\partial\varphi/\partial t$ is shifted by the quantity

$$\omega_{ins} - \omega_0 = -\Gamma e^{-\Gamma t}\delta\varphi < 0 \tag{11.2}$$

Thus, this quantum defect $h(\omega_0 - \omega_{ins})$ is spent to produce the energy of the equilibrium orientation. Let us be reminded that just the conservation of the number of quanta corresponds to the conservation of the adiabatical invariant U_E/ω, compare with the discussion of the variational principle in § 3.4. Here we see clearly that neither the energy density nor the Poynting vector are conserved. Usually this defect is a very small quantity: at $\delta\varphi \sim 20.2\pi \sim 120$ and $\Gamma \sim 10^{-1}\text{s}^{-1}$ the relative frequency shift $|\delta\omega|/\omega$ is smaller than 10^{-14}. If we multiply the typical power density 10^3 W/cm^2 = 10^{10} erg/cm^2 s by the time $\tau \sim 10$ s and by the factor 10^{-14}, we obtain an energy expenditure about 10^{-3} erg/cm^2. The Frank energy for the director's 50% reorientation in a cell of thickness $L = 100$ μm is equal to $FL = LK(\pi/L)^2 \approx 10^{-3}$ erg/cm^2.

These estimates show that a very insignificant absorption $1 - \exp(-\sigma L) \sim 10^{-13}$ would be enough for the light beam to release in the medium 10 times as much energy. Hence it follows that if we would succeed to find a suitable mechanism transforming the absorbed energy to the refractive index change, then the nonlinearities generated by the absorption can be much stronger than all the "dynamic nonlinearities" considered in the previous chapters of the survey.

The simplest group of effects is connected with the medium heating caused by the light absorption; we shall begin with these effects.

11.1 Thermal self-focusing and self-defocusing

A very strong dependence of refractive indices on temperature is a distinctive characteristic of the LC mesophase. While in the isotropic phase $\partial n/\partial T \approx (\partial n/\partial\rho)(\partial\rho/\partial T) \approx -(10^{-4} - 10^{-5})$ deg^{-1}, in the MBBA mesophase at $T = 33$°C (i.e. $T - T_c \approx 10$°C) we have $\partial n_{\|}/\partial T \approx 4.10^{-3}$ deg^{-1}, the quantity $\partial n_{\perp}/\partial T$ is positive but about 5 times lower by its modulus. As a result, thermal self-defocussing takes place when an extraordinary wave propagates through the weakly absorbing NLC and thermal self-focusing occurs with an ordinary wave. As an order of magnitude it can be written that

$$\delta n = \sigma \frac{cn|E|^2}{8\pi\rho c_p} \frac{\partial n_i}{\partial T} \Gamma^{-1}(1 - e^{-\Gamma t}) \qquad (11.3)$$

where σ is the absorption coefficient (cm^{-1}), ρc_p(erg/cm^{3}deg) is the unit volume thermal capacity, Γ^{-1} is the stationary temperature profile establishment time, $\Gamma \sim \chi(a_{\perp}^{-2} + \pi^2/L^2)$; $\chi \sim 10^{-3}$ cm^2/s is the thermal conductivity coefficient, $a_{\perp}$ is the transverse beam size, L is the thickness of the cell with good heat removal on the walls. Such effects experimentally have been observed in the paper [48] already in 1974. Both the power and the duration of the argon laser radiation would have allowed the authors to observe the GON. For this it was necessary to incline the cell with respect to the beam, which unfortunately had not been done. From our point of view this example shows convincingly how non-trivial it was to realize the circumstance that a light field can strongly reorient the director.

If the field consists of two interfering waves $E_1\exp(i\mathbf{k}_1\mathbf{r}) + E_2\exp(i\mathbf{k}_2\mathbf{r})$, the thermal interference gratings of the refractive index are strongly damped by thermal conductivity. In the expression (11.3) it is necessary to consider $\Gamma = \chi[(\pi/L)^2 + (\mathbf{k}_1 - \mathbf{k}_2)^2]$. These gratings lead to the self-diffraction picture (see § 2.4) in a thin cell and give a contribution to the self-defocusing non-stationary picture of both beams. The thermal effects often prevent the observation of the orientational nonlinearity in insufficiently pure samples. For instance, self-diffraction and other thermal effects in cells with NLC have been observed in the papers [14, 49] during investigation of the orientational effects.

The thermal effects for an extraordinary wave lead to defocusing, and the GON leads to self-focusing. In principle these nonlinear effects can be considerably compensated by a choice of the incidence angle.

11.2 Stimulated thermal scattering (STS)

The interference term of the temperature perturbation is shifted by the phase with respect to the interference picture, if in a weakly absorbing medium two waves of different frequencies propagate, $E = E_1\exp(i\mathbf{k}_1\mathbf{r} - i\omega_1 t) + E_2\exp(i\mathbf{k}_2\mathbf{r} - i\omega_2 t)$. Actually it follows from the thermal conductivity equation

$$\frac{\partial T}{\partial t} - \chi\Delta T = \frac{\sigma}{\rho c_p}\frac{cn|E(\mathbf{r},t)|^2}{8\pi} \tag{11.4}$$

that the grating $\delta T \sim E_1E_2^* + c.c.$ has the form

$$\delta T(\mathbf{r},t) = \frac{\sigma c n}{\rho c_p\Gamma(1 + i\Omega/\Gamma)}\frac{E_1^*E_2}{8\pi}e^{-i\mathbf{q}\mathbf{r}+i\Omega t} + c.c. \tag{11.5}$$

where $\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2$, $\Omega = \omega_1 - \omega_2$, $\Gamma = \chi q^2$. The dielectric permittivity perturbation $\delta\varepsilon(\mathbf{r},t) = 2n(\partial n/\partial T)\delta T(\mathbf{r},t)$ reproduces the behaviour of the temperature profile. As it has been shown in detail in § 2.5, the scattering of the wave by the first term in (11.5) leads to amplification of the wave E_2 with the coefficient g_{ef} (cm^{-1}, by intensity) equal to

$$g_{ef} = \frac{\omega}{c}\frac{\partial n}{\partial T}\frac{\sigma}{\rho c_p \Gamma} S_1 \frac{2\Omega/\Gamma}{1 + \Omega^2/\Gamma^2} - \sigma \quad (11.6)$$

Here S_1 is the E_1 wave power density, and from the amplification due to the grating process we have subtracted the absorption σ. The amplification of the wave E_2 at $\partial n/\partial T < 0$ will occur only at $\omega_1 - \omega_2 < 0$. Further, the amplification is maximal at $\Omega = -\Gamma$, and g_{ef} is positive only when the power density S_1 exceeds the threshold value: at $\Omega = -\Gamma$

$$g_{ef} = \sigma(S_1|S_{th} - 1), \qquad S_{th} = \Gamma\rho c_p \lambda[2\pi(\partial n/\partial T)]^{-1} \quad (11.7)$$

The process under consideration is called stimulated temperature scattering (STS) generated by absorption or STS of the second kind (STS-II) [50].

The large value of $|\partial n_{\|}/\partial T|$ for LC mesophase leads to the comparatively small value of S_{th}. For example, for the STS of extraordinary waves counterpropagating perpendicular to the director we have $S_{th} \approx 6.10^5$ W/cm², $\Gamma = 2.10^8$ s^{-1} for $\lambda = 0.5$ μm, $\partial n_{\|}/\partial T = -4.10^{-3}$ deg^{-1}, $\chi = 10^{-3}$ cm²/s, $\rho c_p = 1.5$ J/cm³ deg. If we take the absorption as not very small, $\sigma = 5$ cm^{-1}, $S_1 = 20\ S_{th}$, then the amplification of the wave $|E_2|^2$ becomes $\exp(gz) = e^{10}$ at a length $L \approx 0.1$ cm. During the time $\tau \sim 7\Gamma^{-1} \approx 3.10^{-8}$ s, the medium at such temperatures will be heated by the beam $|E_1|^2$ by about 1.5°C. Thus, the observation of STS-II in NLC appears to be quite realistic by using pulsed lasers.

It is not difficult to obtain the formulae for STS-II in NLC, in CLC and SLC within the general geometry of the interacting waves and with account of absorption dichroism; we do not dwell on the corresponding expressions because there are no qualitative peculiarities here.

11.3 Thermal reorientation

In a homogeneous sample of a nematic, the temperature change leads to the local change of the refractive indices $n_{\|}$ and $n_{\perp}$, but does not change the homogeneous director orientation. If we consider an inhomogeneous nematic, for example a hybrid cell (§ 4.6) or a twist cell, the sample heating caused by absorption must lead to a change of the director profile in the volume due to the dependence of the Frank constants on the temperature. We can expect the appearance of the effects of self-focusing, defocusing, GRON, SS and so on, generated by this mechanism.

There is one more effect in smectics-C apart from the above considered thermal effects: the dependence of the angle ϕ between the n-director and the normal to the smetic layers on the temperature. Heating must lead to the local rotation of the optical axis with all the consequences following from this for nonlinear optics, if in the geometry used the layers orientation can be considered fixed. The following mechanism was suggested for reorientation of by spatially inhomogeneous absorption of laser radiation in the paper [108]. The temperature inhomogeneity gives rise convection both by the Rayleigh-Benard mechanism in the gravitational field or by the Marangoni mechanism due to the dependence of the surface tension on temperature. The coupling between hydrodynamic motion and the director leads to the change of optical properties, i.e. to optical nonlinearity.

11.4 Nonlinearities due to photostimulated transformations

A new and essentially non-thermal mechanism of a large optical nonlinearity generated by absorption has been observed in the papers [51–53]. A planar cell with a solution of cianobiphenyles of thickness 10 μm [51], or with MBBA of thickness 65 μm [52, 53], was illuminated by the field of two plane waves which were incident normally to the director and at a small (up to some degrees) angle α_{12} with respect to each other. The interference of these waves recorded a refractive index grating of spatial period $\Lambda = \lambda/\alpha_{12}$. The intensity of the third (testing) beam diffraction as well as the intensities of the diffraction of the recording waves themselves (compare with § 2.4) were registered. The geometry of the experiment was such that the orientational nonlinearity could not give any considerable contribution to this process. The authors of [51–53] were convinced that the thermal nonlinearities, at the conditions of the experiment, would give several orders of magnitude lower and about 10^3 times faster nonlinearity than the one experimentally observed.

In the papers mentioned above a model of reversible transformation of the molecules in the LC mesophase in the time of the light absorption has been proposed as a mechanism. The highest nonlinearity, $|\varepsilon_a| \sim 5$ cm^3/erg, has been registered at the wavelength $\lambda =$ 0.44 μm (He-Cd laser), at which the MBBA absorption coefficient was $\sigma = 25\ \text{cm}^{-1}$. This nonlinearity decreases about 10 times at the wavelength $\lambda = 0.48\ \mu\text{m}$ for which $\sigma \approx 18\ \text{cm}^{-1}$. If one assumes that the modified state of the molecule lives in the homogeneous medium during time T and diffuses with coefficient $\mathcal{D}$, then for $\delta\varepsilon$ a phenomenological equation can be written

$$\frac{\partial \delta\varepsilon}{\partial t} + \frac{1}{T}\delta\varepsilon - \mathcal{D}\Delta\delta\varepsilon = \frac{1}{2T}\varepsilon_2|E|^2 \qquad (11.8)$$

where the constant ε_2 is proportional to the absorption coefficient. The measurements of the time evolution of $\delta\varepsilon$ and the dependence of $\delta\varepsilon$ on the angle α_{12} between the interfering waves, carried out in the paper [53], have allowed one to determine the values of the parameters from the equation (11.8): $T \approx 1.6$ s, $\mathcal{D}_\perp = 1.3 \cdot 10^{-7}\ \text{cm}^2/s$. The authors were also convinced that the strongest nonlinearity arises when an extraordinary wave is absorbed. Besides, the result of the process is mainly the change of the quantity $\varepsilon_\parallel$, and the quantity $\varepsilon_\perp$ is changed by about half of that for $\varepsilon_\parallel$.

It is interestingly to compare all these results with the results from the previous paper [54], where the processes of deactivation and diffusion of the dye "methyle red" mixed with the MBBA mesophase have been considered. Approximately the same recording technique of amplitude-phase grating by an argon laser radiation was used in [54], but for the grating reconstruction a He-Ne laser beam ($\lambda = 0.628\ \mu\text{m}$) was used because of strong absorption in the green spectral region. The values obtained there were several seconds for the time T and $\mathcal{D}_\parallel \approx 1.5.10^{-7}\ \text{cm}^2/\text{s}$, $\mathcal{D}_\perp \approx 2, 4.10^{-7}\ \text{cm}^2/\text{s}$. Note that in the paper [54] there were also all the experimental conditions allowing one to observe the GON by the beams inclination.

Coming back to the paper [53] let us give the estimate for the change of the MBBA's individual molecule polarizability due to the phototransformation: $\Delta\beta/\beta \sim 10^{-1}$. The method of the experiment [53] did not allow the determination of the signs of $\delta\varepsilon_\parallel$ and $\delta\varepsilon_\perp$, i.e. to refer the effect to self-focusing or self-defocusing. The absence of such a nonlinearity in the isotropic phase of the same samples was a very important experimental fact registered in [53]. Besides, the saturation of the nonlinear addition to the refractive index has been observed for a light power density of the order of $4\ \text{W/cm}^2$. Actually, for absorption $\sigma \sim 25\ \text{cm}^{-1}$ and flow $I \sim 1.5 \cdot 10^{19}$ quanta cm^2 about 6.10^{20} photoexcitations take place in 1 cm^3 during the time $T \simeq 1.5$ s, which constitute a considerable part of the density $N \approx 3 \cdot 10^{21}\ \text{cm}^{-3}$ of the MBBA itself. Therefore, in spite of the large value of the constant ε_2, it turns out to be impossible to obtain a considerable ($\delta\varphi$

$\geqslant 2\pi$) nonlinear phase shift on the basis of the mechanism under consideration in a cell with the MBBA of thickness $L \leqslant 60$ μm.

Recently the same authors showed [109] that the nonlinearity under consideration is largely increased near the phase transition point of NLC to isotropic liquid. The phototransformated molecules decrease the transition temperature like admixtures do; that decrease was registered experimentally.

From our point of view it is very interesting to investigate such type of nonlinearities in the mesophase of other LC. In particular, we think that the pitch of CLC may also be considerably changed due to the photostimulated transformation of molecules.

11.5 Optically controlled LC valves

The creation of light valves, i.e. devices realizing light space modulation, is a very important practical problem and there are many investigations devoted to it. A very important place is occupied by the optically controlled transparents (light valves), i.e. those, where the amplitude-phase transmission coefficient transverse distribution depends on the recording light beam intensity. There is successful creation of such valves on the basis of LC, for example by the scheme of Figure 26. The LC layer and the adjoining layer of weakly absorbing photosemiconductor are squeezed between the transparent

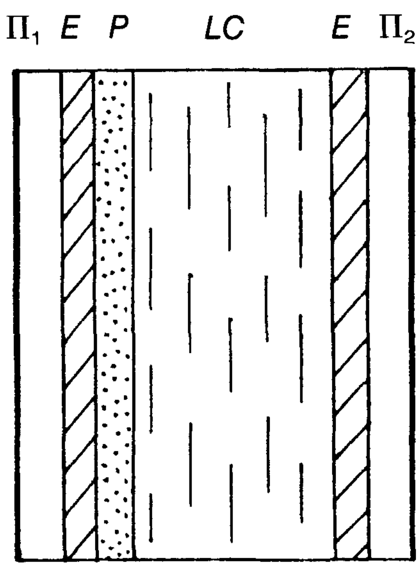


FIGURE 26 Liquid crystalline light valve: E-electrodes, P-photoconducting layer, Π_1, Π_2-transparent glass plates, LC-liquid crystal.

electrodes to which a voltage is applied from a radiofrequency generator. The illumination of the photosemiconductor increases its conductivity and part of the voltage applied to the LC layer increases as a result. Due to this a change of the LC orientation dependent on the incident beam intensity takes place.

A discussion of a great number of very interesting results obtained with the aid of light valves (LV) is outside the scope of the present survey. The LV is an extreme example of a system with absorptional optical nonlinearity where the absorbed energy of the photon is amplified many times by controlling the energy flow from the radiofrequency generator to the LC. We will touch here only upon some experiments where the LVs were carrying out the functions typical for the elements of nonlinear optics.

In the paper [55] a LV supplied with a reflecting mirror was used. Due to this the phase of the reflection coefficient depended on the incident beam local intensity with a very high sensitivity, so that $\delta\varphi \sim 2\pi$ is obtained at $S_{\text{in}} \sim 10^{-5} \cdot -10^{-6}$ W/cm^2. The reference wave E_0 was incident exactly along the normal to the LV plane. The weak signal $E_s(\mathbf{r})$ together with the reference wave were recording a phase hologram of the reflection coefficient $r(\mathbf{r})$ on the LV:

$$r(\mathbf{r}) \approx e_0^{i\varphi}[1 + iAE_0^*E_s + iAE_0E_s^*(\mathbf{r})] \tag{11.9}$$

The reference wave reflected by this hologram contains a component

$$E'(\mathbf{r}) = iAE_0^2E_s^*(\mathbf{r}) \tag{11.10}$$

which propagates exactly contrary to the signal $E_s(\mathbf{r})$ and was phase-conjugated relative to the signal $E_s(\mathbf{r})$. By such a way [55] the operation of phase conjugation was realized using the radiation of a He-Cd laser ($\lambda = 0.44$ μm) with the reference waves of a very small power 10^{-5} W at the power density 10^{-5} W/cm^2. A LV with a nematic BMAOB was used; the layer thickness 5 μm; the establishment time ~8 s; the efficiency of the reflection to the conjugated wave ~1%.

In the paper [56] the LV has been used for a bistable optical device construction. The nematic here had a large dichroism due to the special admixed dye: it was absorbing about 90% in the initial planar orientation and 20–30% after an electric field application which was changing the director orientation in the volume to almost homeotropic. The incident light propagates through the layer of the LC of thickness 10 μm and falls onto the photosemiconductor GaAS of thickness 200 μm. If the input intensity is small, the light absorption in the LC is high, the semiconductor is weakly illuminated and almost all the voltage of the generator (from 20 to 70 V) was applied to the semiconductor layer. The LC layer preserves the planar orientation as a result. If the light intensity is gradually increased, the conductivity of the GaAs layer is increased, the voltage on the LC layer is increased, the director becomes reoriented and the cell transmission increases. The bleached state switching off occurs for a lower intensity of the incident light (compare with the Chapter 9), which corresponds to the optical hysteresis or optical bistability. It is convenient to control such a cell by illuminating directly from the side of the photosemiconductor. The power density required for such a control was about 5.10^{-6} W/cm^2 in the experiment. The use of a semiconductor having a thickness 20 times greater than the LC layer has allowed one, for switching on, to apply a voltage to the LC approximately 20 times greater the FT threshold. The switching on time becomes drastically shorter as a result of this, $\tau_{on} \sim 5.10^{-4}$ s. The relaxation time for switching off was 0.1–0.2 s, which is typical for a cell with thickness 10 μm. The relatively great thickness of the GaAs layer led to the moderate space resolution which was about 10 lines per millimeter.

In the papers [52, 53] LV has served as a nonlinear element inside the Fabry-Perot resonator, and the regime with hysteresis was realized.

12 COMMENTS ON THE BIBLIOGRAPHY

The orientational nonlinearity of the isotropic phase of liquid crystals near the transition point to the mesophase has been investigated in the papers by G. Wang and Y. R. Shen [1], D. Rao and S. Jayaraman [2] and others; see also the survey by Shen [59]. The nonlinear constant ε_2 of the order of 2.10^{-9} cm^3/erg was obtained. In the paper† by B. I. Lembrikov [60], the effect of phase mutual modulation for *e*-and *o*-waves normal propagation in the nematic (GRON- in our terms) has been considered. In the paper by R. M. Herman and R. J. Serinco [47], the process of the NLC director reorientation by a pair of interfering light waves has been considered theoretically with assumption that the cell is close to the Friedericksz transition threshold induced by an external magnetic field. The weak wave amplification process has been considered and the low level of the power required for the observation of the effect, especially near the FT threshold, has been noted. In the paper by B. I. Lembrikov [61], the

†This paper and the subsequent ones are quoted in the order of their submission dates.

stimulated scattering (SS) on the second sound in SLC-A with amplification constant of the same order that for the stimulated Brillouin scattering in ordinary liquids has been considered theoretically. In the paper by B. Ya. Zel'dovich and N. V. Tabiryan [32], the SLC-C orientational optical nonlinearity and the corresponding processes of SS due to the orientational, thermal and orientational-thermal mechanisms have been calculated; the estimates carried out have indicated the large value of the nonlinearity constant. In the paper by B. Ya. Zel'dovich and N. V. Tabiryan [30], the NLC cubic susceptibility tensor in light fields due to the director reorientation has been calculated, the processes of the orientational and thermal SS in NLC have been considered and the amplification coefficient's anomalously large values were noted. In the paper by B. Ya. Zel'dovich, N. F. Pilipetskii, A. V. Sukhov and N. V. Tabiryan [8], the giant orientational nonlinearity (GON) of NLC mesophase—the analog of the Friedericksz effect for the oblique orientation of the field with respect to the director—has been experimentally observed for the first time. The experiment on self-focussing of a weak-power radiation $\sim 10^{-2}$ W was carried out to check the theoretical predictions of the present review's authors, developed by them independently from the investigations by Lembrikov, Herman and Serinko. In our paper [8] these predictions have been confirmed. In the paper [46] by I. C. Khoo and S. L. Zhuang, an experimental verification of the Herman and Serinko predictions [47] has been realized by amplification observation in self-diffraction. The theoretical papers by B. Ya. Zel'dovich and N. V. Tabiryan [62, 63, 64], contain a detailed discussion of the processes GON, GRON, SS and optical phase conjugation due to the orientational nonlinearity mechanism of the mesophase of NLC, CLC and SLC. A. S. Zolotko, V. F. Kitaeva, N. Kroo, N. N. Sobolev and L. Csillag used a homeotropic cell in the paper [13]. They observed experimentally a threshold reorientation of the director at normal incidence of the beam with power of about 0.1 W—light induced Fredericksz transition in a narrow sense (LIFT). In the paper [51], S. G. Odulov, Ju. A. Reznikov, O. G. Sarbey, M. S. Soskin, E. K. Frolova, A. I. Khizhnyak reported the observation of a large non-thermal absorptional nonlinearity of the NLC mesophase by the method of self-diffraction. The theory of the LIFT, including its qualitative differences from the FT theory in quastatic fields, has been given by B. Ya. Zel'dovich, N. V. Tabiryan and Ju. S. Chilingaryan in the paper [65].

During the short period from 1980 to the beginning of 1985 a large number of interesting theoretical and experimental investigations devoted to the LC mesophase orientational nonlinearity have been carried out.

Experiment

Note the experimental papers [66–77] which were not referred to explicitly in the main text of the survey. In [66] a light induced birefringence in a NLC cell with establishment time of about 20 minutes was observed: its nature is not clear at present. In [67] the results of an investigation of the GON-self focusing in a homeotropic cell with MBBA without external fields are reported and the GON standard theory is confirmed. In the papers [21, 68–70] the LIFT process in MBBA is investigated. In the paper [71] the LIFT is investigated for a MBBA layer freely hanging in the air whose orientation was supported by a magnetic field. The threshold power was about 5.10^{-3} W for the thickness $L = 200\ \mu m$; the threshold dependence on the field $\mathcal{H}$ allowed one to estimate the surface energy anisotropy as $\sim 10^{-5}$ erg/cm^2. In the paper [72] the GON in the regime of orientational nonlinearity accumulation from the short pulses sequence was investigated. In the papers [9, 24, 73] a special attention was paid to the fringes of aberrational self-focusing.

Theory

The equations of LC equilibrium and the Maxwell equations in section 3.3 we have obtained from a single Lagrangian following our paper [23]; see also the paper [78] where this method is applied for obtaining the conservation laws. The GON theory in section 4.1 is presented following our papers [8, 9] and [62] the generalization for narrow beams (section 4.2) and for a weak anchoring of the director on the walls was given in the paper [79]. The GON in a hybrid cell is considered in [79] and [80]. The GON in a CLC-cell of large pitch (section 4.8) is investigated theoretically and experimentally in the paper [16]. The theory of the GON and the nonlinear optical activity in smectics-*C* (section 4.9) was developed in the papers [64, 81].

The LIFT theory in sections 5.1–5.5 is presented following our papers [23, 65]; in [23] there is a critical analysis of some versions of the LIFT theory published after [65]. The various aspects of the LIFT theory are discussed also in the papers [14, 22, 24, 27, 82], note that we do not agree with some of the results of these publications, see [23]. The account of the field z-component influence on the LIFT threshold (the factor $\varepsilon_{\parallel}/\varepsilon_{\perp}$) was for the first time carried out in the paper [65]; there the possibility of the LIFT hysteresis in broad beams has been noted. The LIFT theory of an ordinary wave in a planar cell near the threshold has been formulated in the paper [23], which we followed in section 5.8. The transcendental equation (5.49) with the assumption $\Gamma' = 0$, $\Gamma'' = 0$ was obtained in the paper [84] where the integral operator from [23] was used.

The GRON theory we presented here following the papers [62, 64, 30], see also [60, 85, 86]. The orientational SS theory (section 7) is presented after papers [30–32], see also [85], and [29]. The light influence on the LC surface layer was considered theoretically in the paper [34], see the experiment in [35]. The statement of the problem on a CLC helix pitch change as a result of reflected photons angular momentum action has been given in the paper [31]; we follow this paper in section 8.3. The results of section 8.4 are given on the basis of the paper [87].

The hysteresis in cholesterics due to the dynamic untwisting of the helix was considered in [88], where the solution of the self-consistent problem cholesteric + field was obtained. The thermal mechanisms of the hysteresis in CLC (section 9.2) were discussed in [106]. The general review on optical bistability (without LC) including the Fabry-Perot nonlinear resonators, see in [37]. In a discussion of a resonator with a normally placed homeotropic NLC-cell we followed papers [89–90]. The theory of orientational nonlinearity in the presence of other fields in Chapter 10 we followed the papers [47, 45, 42], see also [91]. The cholesterical Friedericksz transition in the cell with the director homeotropic anchoring was considered theoretically and investigated experimentally in the papers [92, 93], see also the theoretical papers [94, 95]. The influence of this transition on the LIFT was discussed theoretically in [96].

The thermally stimulated scattering (TSS) due to absorption, for isotropic liquids was considered by R. M. Herman and M. A. Gray in 1967 [50], see also [97] and [98]. TSS in NLC and SLC-C was considered in the papers [30, 32].

The considerations of the thermal reorientation in section 11.3 belong to the authors of the present review. The survey on the papers about LC light valves is given in [99]. The proposal to use LC light valves for optical phase conjugation was expressed in [91], see the experiment [55].

A large number of papers on LC nonlinear optics partially covering the publications of the same authors in other editions is contained in the collection [100]. We did not include in the list hardly accessible references such as various conferences, theses and so on.

We have known recently about some else papers concerning with the problems under consideration in the present review [110–154].

One can judge about subject of those papers by given titles.

Finishing these comments on the bibliography we would like to note the following. The possibility of the giant nonlinear effects observation in thin (~0.01 cm) layers of LC at a very low level of power ($\sim 10^{-2} - 10^{-3}$ W) was not in the least obvious at the time of the appearance of the first theoretical papers. Note, for example, the detailed review on the LC nonlinear optics of the "pre-orientational era" [101]. Even such well-known authorities in nonlinear optics as G. A. Ascaryan and Y. R. Shen have expressed a doubt on the possibility of GON observation upon first acquaintance with the corresponding theoretical proposals.

The wide investigations of orientational optical nonlinearity were stimulated mostly by the papers [8, 13] in the socialist countries and by the papers [47, 46] in the western hemisphere.

Conclusion

The investigation of the orientational optical nonlinearity is only beginning, as a matter of fact. In a number of cases nonlinear effects allow one to determine LC parameters, which are difficult to measure by other methods. A distinct characteristic of optical action on LC is its high spatial locality, up to sizes $\Lambda/2\pi = \lambda/4\pi n \sim 250$ Å. Short laser pulses allow to obtain very high temporal resolution—up to 10^{-12} s. Besides, nonlinear effects in LC mesophase allow one to simulate many phenomena which are of interest for coherent optics, with the aid of low-power lasers. The role of possible nonlinear effects (bad in some cases) should be carefully taken into account in the construction of LC devices.

In our point of view the detailed experimental verification of the numerous theoretical predictions is very desirable. We are almost sure that a large number of unexpected effects will be found this way; maybe even more interesting than the ones we have been engaged in with pleasure during the last six years.

The authors thank E. I. Katz, R. S. Hakopyan, N. F. Pilipetsky, L. M. Blinov and V. V. Shkunov for valuable discussions. We are deeply thankful to Yu. P. Raizer, and V. V. Titov for support of this work.

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