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The Orientational Optical Non-linearity of Liquid Crystals. I. Nematics

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The optical non-linearity of a nematic liquid crystal (NLC) is considered. The strongest effect studied is due to the local turn of NLC director. The possibility of wavefront conjugation by four-wave interaction in a NLC is considered. Strong effects of self-focusing in a NLC due to the orientational mechanism of non-linearity are predicted.

INTRODUCTION

It is well known that in the isotropic phase of liquid crystals (LC) near the transition point, the third order optical non-linearity (non-linearity of self-focusing type) is large.[1-3] This is connected with mesophase embryos (nematic, for example) which appear in the isotropic phase near the transition point, and which may be oriented rather easily by the light fields. These embryos are responsible also for the large value of the cross section of spontaneous light scattering in the isotropic phase near the transition point.[4-7] There is a general relation between the cross-section of spontaneous scattering and the third order non-linearity.[7-10] This relation is an analogue of the fluctuation-dissipation theorem.

Below the transition point, i.e., in a nematic phase, the spontaneous scattering cross-section is also quite large. The spontaneous light scattering in a NLC for a single domain specimen is mainly due to thermal fluctuations of the local orientation of the director.[11] Optical homogeneity of appreciable volumes of a NLC can be achieved by the orientation action of an external magnetic field of $H \sim 10^3$ gauss.[11] It is clear that the third-order optical non-linearity of a NLC must also be large due to the strong influence of light fields on the orientation of the director. Indeed, in all real cases, both forces returning the director to the initial direction, i.e., the Frank energy force and the force of magnetic orientation, are rather small, and the system responds effectively even to weak light waves.

However, as far as we know, the cubic optical non-linearity of a NLC has not been discussed so far. The aim of the present paper is to fill this gap. Besides, two particular non-linear optical effects are discussed. The first is the wave front reversal (WFR) by four-wave interaction (FWI) (about WFR, see;[12,13] especially about WFR-FWI see [14,15]). The second is the self-focusing of light in a NLC due to the orientational mechanism of non-linearity. Both effects should be very large in our estimation, but only for a properly chosen experimental geometry.

FREE ENERGY, DISSIPATION FUNCTION, AND EQUATIONS OF MOTION

For the density of free energy $F(\text{erg/cm}^3)$ in a NLC we use the form:

$$F = \frac{1}{2}\left[K_{11}(\operatorname{div}\mathbf{n})^2 + K_{33}(\mathbf{n} \times \operatorname{rot}\mathbf{n})^2 + K_{22}(\mathbf{n}\operatorname{rot}\mathbf{n})^2 \right.$$
$$\left. - \kappa_a(\mathbf{nH})^2 - \frac{\varepsilon_a}{8\pi}(\mathbf{nE})(\mathbf{nE}^*)\right]. \quad (1)$$

Here $\mathbf{n}(r, t)$, is the unit vector $|\mathbf{n}| = 1$ defining the local direction of the mean molecular orientation (director); K_{11}, K_{22}, K_{33} (dyn), are the Frank constants; $\kappa_a = \kappa_{\parallel} - \kappa_{\perp}$, is the magnetic susceptibility anisotropy ($\mu_{ik} = 1 + 4\pi\kappa_{ik}$); $\mathbf{H}$, is the vector of a static magnetic field; ε_a, is the anisotropy of dielectric susceptibility at light frequencies, $\varepsilon_a = \varepsilon_{\parallel}(\omega) - \varepsilon_{\perp}(\omega) \cdot \mathbf{E}$ is the complex vector amplitude of the light field connected with the real vector $\mathbf{E}_{\text{real}}$ by:

$$\mathbf{E}_{\text{real}}(r, t) = 0.5[\mathbf{E}\exp\{-i\omega t\} + \mathbf{E}^*\exp\{i\omega t\}].$$

In Eq. (1) we omitted some terms, which are independent of the director orientation $\mathbf{n}$.

The dissipation function density R (erg cm^{-3} sec^{-1}) takes the form:

$$R = \tfrac{1}{2}\gamma\dot{n}_i\dot{n}_i. \quad (2)$$

Here we neglect the effects of hydrodynamic motion and the hydrodynamic mechanism of orientation relaxation. Therefore, the free energy and dissipation function in Eqs. (1) and (2) do not include the macroscopic velocity $\mathbf{v}$.†

Motion equations have the form

$$\frac{\delta F}{\delta n_i} - \frac{\partial}{\partial x_j}\frac{\delta F}{\delta(\partial n_i/\partial x_j)} + \lambda n_i = -\frac{\delta R}{\delta \dot{n}_i}. \tag{3}$$

where λ is the Lagrange multiplier, to be determined from the condition $\mathbf{n}^2 = 1$. Insertion of Eqs. (1) and (2) into Eq. (3) yields rather lengthy expressions.

Let us consider the case where, in the zero approximation, the NLC is homogeneously oriented by an external magnetic field $\mathbf{H}$, i.e., $\mathbf{n}^0 = \mathbf{H}/H$. Then, for small variations $\mathbf{n}(\mathbf{r}, t) = \mathbf{n}^0 + \delta\mathbf{n}(\mathbf{r}, t)$ we obtain from Eqs. (1)–(3):

$$\begin{aligned}\gamma\frac{\partial \delta n_i}{\partial t} &+ K_{22}[\nabla_i(\nabla\delta\mathbf{n}) + (\mathbf{n}^0\nabla)^2\delta n_i - \Delta\delta n_i] - K_{11}\nabla_i(\nabla\delta\mathbf{n}) - K_{33}(\mathbf{n}^0\nabla)^2\delta n_i \\ &+ (K_{11} - K_{22})(\mathbf{n}^0\nabla)(\nabla\delta\mathbf{n})n_i^0 + \kappa_a H^2\delta n_i \\ &= \frac{\varepsilon_a}{16\pi}(\delta_{il}n_m^0 + \delta_{im}n_l^0 - 2n_i^0 n_l^0 n_m^0)E_l E_m^*.\end{aligned} \tag{4}$$

The values of the tensor product $E_l(\mathbf{r}, t)E_m(\mathbf{r}, t)$ is taken at the point $\mathbf{r}$ and at the moment t. The general solution of Eq. (4) has the form:

$$\delta n_i(\mathbf{r}, t) = \int D_{ilm}(\mathbf{r} - \mathbf{r}', t - t')(E_l E_m^*)_{\mathbf{r}', t'}\, d^3\mathbf{r}'\, dt'. \tag{5}$$

It is convenient to make an explicit calculation of the response function D_{ilm} in Fourier space:

$$(E_l E_m^*)_{\mathbf{r}, t} = \int A_{lm}(\mathbf{q}, \Omega)\exp\{i\mathbf{q}\mathbf{r} - i\Omega t\}d^3\mathbf{q}\,d\Omega,$$

$$\delta\mathbf{n}(\mathbf{r}, t) = \int \tilde{\mathbf{n}}(\mathbf{q}, \Omega)\exp\{i\mathbf{q}\mathbf{r} - i\Omega t\}d^3\mathbf{q}\,d\Omega. \tag{6}$$

Then rather lengthy calculation yields

$$\tilde{n}_i(\mathbf{q}, \Omega) = \tilde{D}_{ilm}(\mathbf{q}, \Omega)A_{lm}(\mathbf{q}, \Omega), \tag{7a}$$

$$\tilde{D}_{ilm}(\mathbf{q}, \Omega) = \frac{\varepsilon_a}{16\pi}\Gamma_{ik}(\delta_{kl}n_m^0 + \delta_{km}n_l^0), \tag{7b}$$

† Note that by omitting the terms containing $\mathbf{v}(\mathbf{r}, t)$ in the dissipiation function we come across the paradox of the dissipation for the homogeneous translation or rotation of a NLC *as a whole*. For a more accurate expression for R see Refs. 7 and 16, but Eq. (2) will be sufficient for us.

where

$$\Gamma_{ik} = [q^2 - (\mathbf{nq})^2]^{-1}[\Gamma_2 q^2(\delta_{ik} - n_i^0 n_k^0) - (\mathbf{n}^0\mathbf{q})^2(\Gamma_2 \delta_{ik} - \Gamma_1 n_i^0 n_k^0$$

$$+ (\mathbf{n}^0\mathbf{q})(\Gamma_2 - \Gamma_1)(n_i^0 q_k + n_k^0 q_i) - (\Gamma_2 - \Gamma_1)q_i q_k], \tag{7c}$$

$$\Gamma_1 = (-i\gamma\Omega + K_{11}q^2 + (K_{33} - K_{11})(\mathbf{n}^0\mathbf{q})^2 + \kappa_a H^2)^{-1}, \tag{7d}$$

$$\Gamma_2 = (-i\gamma\Omega + K_{22}q^2 + (K_{33} - K_{22})(\mathbf{n}^0\mathbf{q})^2 + \kappa_a H^2)^{-1}. \tag{7e}$$

In the above calculations, it is convenient to use the coordinate system with the following system of unit orthogonal vectors:

$$\mathbf{e}_3 = \mathbf{n}^0; \mathbf{e}_2 = \frac{[\mathbf{n}^0 \times \mathbf{q}]}{|[\mathbf{n}^0 \times \mathbf{q}]|}; \mathbf{e}_1 = [\mathbf{n}^0 \times \mathbf{e}_2]. \tag{8a}$$

In such a coordinate system the matrix Γ_{ik} has the form:

$$\hat{\Gamma} = \begin{pmatrix} \Gamma_1 & 0 & 0 \\ 0 & \Gamma_2 & 0 \\ 0 & 0 & 0 \end{pmatrix} \tag{8b}$$

In subsequent considerations, we should hardly use the Fourier integral transformation (6), because in cases of interest to us, the field $E(\mathbf{r}, t)$ consists of a small number of plane monochromatic components.

THE NLC OPTICAL NON-LINEARITY AND ITS POLARIZATION PROPERTIES

Small perturbations of the director $\delta\mathbf{n}$ produce perturbations of the dielectric susceptibility tensor at the frequency of light:

$$\delta\varepsilon_{ik}(\mathbf{r}, t) = \varepsilon_a[n_i^0 \delta n_k(\mathbf{r}, t) + n_k^0 \delta n_i(\mathbf{r}, t)], \tag{9a}$$

and $\mathrm{Tr}(\delta\hat{\varepsilon}) = 0$ because of the condition $\mathbf{n}\delta\mathbf{n} = 0$. The non-linear term for the complex amplitude of the displacement $\mathbf{D}(\mathbf{r}, t)$ at the frequency of light equals

$$(\mathbf{D}^{NL})_i = \delta\varepsilon_{ik}(\mathbf{r}, t)E_k(\mathbf{r}, t). \tag{9b}$$

The insertion of Eqs. (7), (8), and (9a) into Eq. (9b) allows us to obtain the third-order polarizability tensor χ_{iklm}. Let us consider more carefully the situation, where, in the tensor product $(E_l E_m^*)_{\mathbf{r}, t}$, we pick out the term with the space-time dependence $\exp\{i\mathbf{qr} - i\Omega t\}$. This corresponds to the situation, when, for example, the following kind of light waves propogate in the medium:

$$\mathbf{E} = \mathbf{E}_1 \exp\{i\mathbf{K}_1\mathbf{r} - i\omega_1 t\} + \mathbf{E}_3 \exp\{i\mathbf{K}_3\mathbf{r} - i\omega_3 t\}$$

with $\mathbf{q} = \mathbf{K}_1 - \mathbf{K}_3, \Omega = \omega_1 - \omega_3, A_{lm}(\Omega, \mathbf{q}) = (\mathbf{E}_1)_l(\mathbf{E}_3^*)_m$. Then one obtains:

$$\delta\varepsilon_{ik}(\mathbf{r}, t) = 4\pi \exp\{i\mathbf{qr} - i\Omega t\}\chi_{iklm}(\Omega, \mathbf{q})E_{1l}E_{3m}^*, \tag{9c}$$

$$\chi_{iklm}(\Omega, \mathbf{q}) = \frac{\varepsilon_a}{4\pi}(n_i D_{klm} + n_k D_{ilm}) = \frac{\varepsilon_a^2}{64\pi^2}[q^2 - (\mathbf{nq})^2]^{-1}.$$

$$\cdot [\Gamma_2(f_{il}n_k n_m + f_{mi}n_k n_l + f_{lk}n_i n_m + f_{km}n_i n_l - 4q^2 n_i n_k n_l n_m)$$
$$- \Gamma_1(p_i q_l n_k n_m + p_m q_i n_k n_l + p_l q_k n_i n_m + p_k q_m n_i n_l - 4(\mathbf{nq})^2 n_i n_k n_l n_m)]. \tag{9d}$$

Here and below we omit the index 0 in the notation of the vector $\mathbf{n}^0$ of the unperturbed director. In addition, the following notations were introduced in Eq. (9d)

$$\mathbf{p} = 2\mathbf{n}(\mathbf{qn}) - \mathbf{q}; \qquad f_{\alpha\beta} = [q^2 - (\mathbf{qn})^2]\delta_{\alpha\beta} + p_\alpha q_\beta$$

(note, that $f_{\alpha\beta} \neq f_{\beta\alpha}$). The tensor χ_{iklm}, as well as either of the two terms in Eq. (9d) (proportional respectively to Γ_1 and Γ_2), have zero trace and are symmetrical relative (a) to the permutation indices i, k, and (b) to the permutation indices l, m. Moreover, these tensors are invariant under the change $(i, k) \rightleftarrows (l, m)$, the last property being connected with the even dependence of tensor χ on the vector $\mathbf{q}$. Equation (9a) will be used in Section 4, where the WFR–FWI is discussed.

Another case corresponds to the problem of the self-action of the lightwave. If we consider an approximately plane wave, then, neglecting the Frank enegy effects (for the smooth field profile), we obtain from Eq. (3):

$$\delta\varepsilon_{ik} = \frac{\varepsilon_a^2}{16\pi\kappa_a H^2}(\delta_{il}n_k n_m + \delta_{im}n_k n_l + \delta_{kl}n_i n_m + \delta_{km}n_i n_l - 4n_i n_k n_l n_m)\mu$$
$$\times \int_{-\infty}^{t} \exp\{-\mu(t - t')\}(E_l E_m^*)_{\mathbf{r},t'}\,dt', \tag{10}$$

where $\mu = \kappa_a H^2/\gamma$. It is interesting to note that in the limit $|\mathbf{q}| \to 0$, Eqs. (9) taking account of Eq. (7), transform into Eq. (10).

Let us discuss now the polarizational properties of the optical non-linearity of the NLC. Let us remember that, with respect to linear optics, the NLC is a strongly anisotropic, uniaxial crystal. There are, therefore, two independent waves—the ordinary wave ("o" type wave for which $(\mathbf{nE}) = 0$), and the extraordinary wave ("e" type wave for which the electric field is in the plane of $\mathbf{n}$ and the wave vector $\mathbf{K}$).

The non-linear term $\delta\varepsilon_{ik}$ from Eq. (10) also equals zero if the light field corresponds to the ordinary wave. This is valid for any orientation of the ordinary wave vector $\mathbf{K}$. Besides, the non-linear term from Eq. (10) equals zero for the extraordinary wave in two important, special cases when: (1) the wave vector $\mathbf{K}_e$ is in the plane perpendicular to $\mathbf{n}$ (the electric polarisation vector is parallel to $\mathbf{n}$), and (2) $\mathbf{K}_e \| \mathbf{n}$; in that case, for propagation along the axis, any wave is ordinary. The physical reason for the absence of non-linearity in all the above situations is that the light field cannot change, in the first non-vanishing approximation, the director orientation either for $\mathbf{E}\|\mathbf{n}$ or for $\mathbf{E} \perp \mathbf{n}$. Only for an oblique, mutual orientation of vectors $\mathbf{E}$ and $\mathbf{n}$ does the light field affect the orientation in the first order by $|\mathbf{E}|^2$.

Thus, the self-focusing type of non-linearity, appearing due to director reorientation in the NLC, is very large, but may reveal itself only for the extraordinary wave which falls obliquely relative to the director.

Consider now the four-wave non-linearity which is important for the WFR–FWI problem. The non-linearity also turns to zero in this problem, if 4 or even 3 interacting waves are ordinary.

Thus in the FWI problem, the orientational non-linearity of the NLC is large, but only if certain conditions for the types of polarization of interacting waves are fulfilled.

ON WAVE FRONT CONJUGATION POSSIBILITIES BY FOUR-PHOTON INTERACTION IN A NLC

(a) WFC-4FI kinematics

The aim of WFR[12,13] is to create the "reversed" wave from some signal wave $\mathbf{E}_3(\mathbf{r}) \exp\{i\mathbf{K}_3\mathbf{r} - i\omega t\}$ by means of non-linear optical effects, where the "reversed" wave is proportional to

$$E_4 \sim [E_3(\mathbf{r}) \exp\{i\mathbf{K}_3\mathbf{r}\}]^* \exp\{-i\omega t\}. \tag{11}$$

The wave E_4 is the "time reversed" solution of the wave equation. The realization of WFR procedure given by Eq. (11) allows the solution of such practically important problems as the improvement of radiation divergence for a powerful optically inhomogeneous laser amplifier, self-targeting of radiation, etc. The WFR method by four-wave interaction (FWI)—(also called WFR in dynamic holography) consists of the following [13–15].

The medium with cubic optical non-linearity is illuminated by two plane opposite waves $\mathbf{E}_1 \exp\{i\mathbf{K}_1\mathbf{r}\}$ and $\mathbf{E}_2 \exp\{i\mathbf{K}_2\mathbf{r}\}$, $\mathbf{K}_2 = -\mathbf{K}_1$, called the reference waves, see Figure 1. The signal wave, $\mathbf{E}_3(\mathbf{r}) \exp\{i\mathbf{K}_3\mathbf{r}\}$, is also directed at the same medium; here $\mathbf{E}_3(\mathbf{r})$ is the slowly varying, complex amplitude envelope. Among different terms of the cubic response of main importance for WFR–FWI are:

$$P_4 = XE_1E_2E_3^*\mathbf{r}) \exp\{i\mathbf{K}_4\mathbf{r} - i\omega t\}, \tag{12}$$

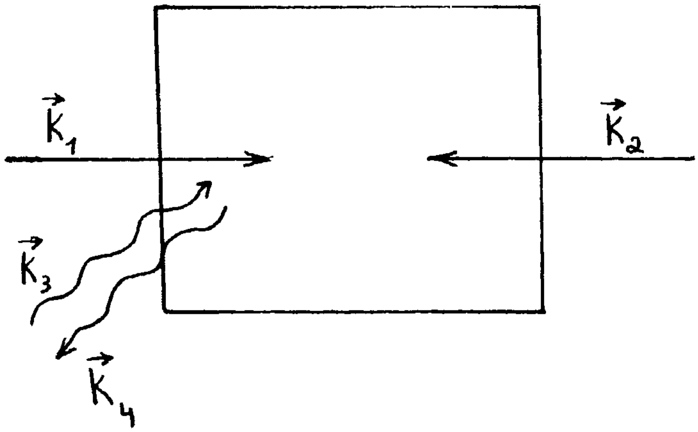

FIGURE 1 Kinematics of four-wave interaction for wave front reversal: $\mathbf{k}_1$ and $\mathbf{k}_2$, the directions of strong reference waves; $\mathbf{k}_3$, the direction of the signal to be reversed; $\mathbf{k}_4$, the direction of the reversed wave.

for which the synchronism condition is satisfied

$$\mathbf{K}_3 + \mathbf{K}_4 = \mathbf{K}_1 + \mathbf{K}_2; \qquad |\mathbf{K}_4| = \frac{\omega}{c}\sqrt{\varepsilon}. \tag{13}$$

Just those terms are responsible for the WFR–FWI.

For an optically uniaxial medium (an oriented NLC), the synchronism condition (13) is satisfied for the following schemes of interaction:

1) $\mathbf{K}_1(e) + \mathbf{K}_2(e) = \mathbf{K}_3(o) + \mathbf{K}_4(o) = 0$
2) $\mathbf{K}_1(e) + \mathbf{K}_2(e) = \mathbf{K}_3(e) + \mathbf{K}_4(e) = 0$
3) $\mathbf{K}_1(o) + \mathbf{K}_2(o) = \mathbf{K}_3(e) + \mathbf{K}_4(\mathrm{e}) = 0$
4) $\mathbf{K}_1(o) + \mathbf{K}_2(o) = \mathbf{K}_3(o) + \mathbf{K}_4(o) = 0$

Here the letters o, e map the wave type. Some other cases of synchronism are also possible; for example, for the waves $\mathbf{K}_1$, $\mathbf{K}_2$, $\mathbf{K}_3$, $\mathbf{K}_4$ propagating almost collinearily in the plane perpendicular to the director, the synchronism is realized for

5) $\mathbf{K}_1(e) + \mathbf{K}_2(o) = \mathbf{K}_3(e) + \mathbf{K}_4(o) = \Delta\mathbf{K}$.

Schemes (1) and (3) seem to be the most interesting. Indeed, as is known, the requirement of a large non-linearity contradicts the requirement of preventing the strong reference waves for self-focusing. As shown above, for strong waves $\mathbf{E}_1$ and $\mathbf{E}_2$ of e-type propagating in the plane perpendicular to the director (i.e. for $(\mathbf{nK}_1) = 0$ and $(\mathbf{nK}_2) = 0$), there is no orientational self-focusing. In case 3), with the o-type reference waves, self-focusing does not occur for any direction of propagation $\mathbf{K}_1$ and $\mathbf{K}_2$. In case (4), in accordance with our results from a previous section, the four-wave orientational nonlinearity equals zero.

(b) Reversal coefficient calculation and numerical estimates

Let us consider the particular Scheme (1) (see Figure 2), when two strong, oppositly directed reference waves $\mathbf{E}_1$ and $\mathbf{E}_2$ with orientation of the electric vector along the director (*e*-type waves), fall normally to the planar oriented cell. The *z*-axis is taken along the director, the *x*-axis is disposed along the normal to the cell boundary, and the *y*-axis is perpendicular to the plane of Figure 2. Let the signal wave have *o*-type polarization, and its wave vector

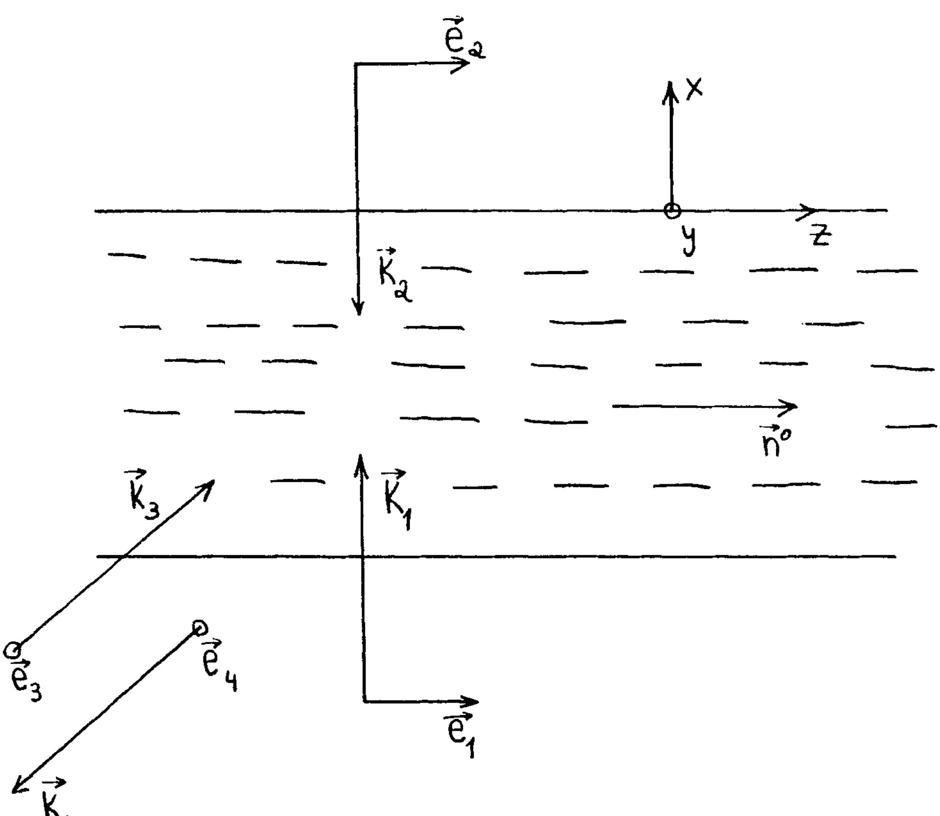

FIGURE 2 Particular scheme of four-wave reversal. The y-axis and the polarization vectors $\mathbf{e}_3$ and $\mathbf{e}_4$ are perpendicular to the figure plane.

be equal to:

$$\mathbf{K}_3 = (K_{3x}, K_{3y}, K_{3z}) = \frac{\omega}{c} n_0(-\sin\phi_3 \cos\psi_3, \sin\phi_3 \sin\psi_3, \cos\phi_3). \quad (14)$$

Here ϕ_3 is the angle between $\mathbf{K}_3$ and director, while ψ_3 together with ζ_3 defines the angle α_3 formed by $\mathbf{K}_3$ with the normal $\mathbf{e}_x$ to the cell boundary: $\cos\alpha_3 = -\sin\phi_3 \cos\psi_3$. The non-linearity under consideration will generate in that scheme only the o-type conjugated wave E_4. The equation for the slowly varying amplitude E_4 has the form:

$$\cos\alpha_3 \frac{dE_4}{dx} = i \frac{2\pi\omega}{cn_0} E_2 E_1 E_3^* \cdot e_{4i}^* e_{2k} e_{1l} e_{3m}^* \{\mathrm{X}_{iklm}(\mathbf{q}_1) + \mathrm{X}_{ilkm}(\mathbf{q}_2)\}. \quad (15)$$

Here we assume that all four waves are monochromatic and have the same frequency ω; therefore it is necessary to put $\Omega = 0$ in Eq. (7) for $\mathrm{X}(\mathbf{q}, \Omega)$. We introduced notations $\mathbf{q}_1 = \mathbf{K}_1 - \mathbf{K}_3, \mathbf{q}_2 = \mathbf{K}_2 - \mathbf{K}_3$ in Eq. (15). Remember that in our case the waves $\mathbf{E}_1$ and $\mathbf{E}_3$ are of different types; therefore $|\mathbf{K}_1| = |\mathbf{K}_2| > |\mathbf{K}_3| = |\mathbf{K}_4|$.

From the terminology of dynamic holography, the term $\mathrm{X}(\mathbf{q}_1)$ from Eq. (15) describes the hologram recorded by the interference of the $\mathbf{E}_1$ and $\mathbf{E}_3^*$ waves reconstructed by the wave $\mathbf{E}_2$. The second term in Eq. (15) describes the recording of the hologram by $\mathbf{E}_2$ and $\mathbf{E}_3$ and reconstruction by $\mathbf{E}_1$. Vectors of polarization $\mathbf{e}_i$ ($i = 1, 2, 3, 4$) in Eq. (15) are pure and real, and equal in pairs: $\mathbf{e}_1 = \mathbf{e}_2 = \mathbf{e}_z, \mathbf{e}_3 = \mathbf{e}_4$.

From Eqs. (9) and (15) and taking account of the chosen polarization directions, we get:

$$\frac{dE_4}{dx} = i \frac{2\pi\omega}{cn_o \cos\alpha_3} E_1 E_2 E_3^* \{\mathrm{X}_{ef}(\mathbf{q}_1) + \mathrm{X}_{ef}(\mathbf{q}_2)\}. \quad (16a)$$

Here $\mathrm{X}_{ef}(\mathbf{q}_1)$ and $\mathrm{X}_{ef}(\mathbf{q}_2)$ correspond to the first and second terms in Eq. (15) and

$$\mathrm{X}_{ef}(\mathbf{q}_1) = \frac{\varepsilon_a^2}{64\pi^2} [\Gamma_1(\mathbf{q}_1)\sin^2\psi_3 + \Gamma_2(\mathbf{q}_1)\cos^2\psi_3], \quad (16b)$$

$$\mathrm{X}_{ef}(\mathbf{q}_2) = \frac{\varepsilon_a^2}{64\pi^2} [\Gamma_1(\mathbf{q}_2)\cos^2\psi_3 + \Gamma_2(\mathbf{q}_2)\sin^2\psi_3]. \quad (16c)$$

Let us consider in more detail the $\mathbf{E}_3$ signal wave falling normally on the NLC layer, i.e. the case $\phi_3 = 90°$, $\psi_3 = 180°$. Then $\mathbf{K}_3 = (\omega n_0/c)\mathbf{e}_x$, $\mathbf{q}_1 = \mathbf{K}_1 - \mathbf{K}_3 = (\omega/c)(n_e - n_0)\mathbf{e}_x$, $\mathbf{q}_2 - \mathbf{K}_2 - \mathbf{K}_3 = -(\omega/c)(n_e + n_0)\mathbf{e}_x$. In this case $|\mathbf{q}_2| \gg |\mathbf{q}_1|$, and the largest contribution is due to $\mathrm{X}_{ef}(\mathbf{q}_1)$. In other words, the dynamic hologram in such a geometry is recorded most strongly by the waves $\mathbf{E}_1$ and $\mathbf{E}_3^*$, because their interference has the larger spatial period and requires smoother distortions of the NLC oriented structure.

Let us make numerical calculations. Let $H = 10^3$ gauss and $\lambda_{vac} = 0.7\ \mu m$ (in vacuum). For 4-azoxyanisole (PAA) in the nematic phase ($T = 125°C$, see[11]), $n_0 = 1.5$, $n_e = 1.8$, $n_e - n_o = 0.3$, $\varepsilon_a = n_e^2 - n_o^2 = 1$, $K_{11} = 4.5 \cdot 10^{-7}$ dyn, $\kappa_a = 1.2 \cdot 10^{-7}$. The expression for $\Gamma_1(\mathbf{q}_1)$ in such a geometry contains two terms in the denominator, $\kappa_a H^2$ and $K_{11}\mathbf{q}_1^2$, corresponding to the restoring forces of the magnetic and inhomogeneous distortions respectively. For the chosen numerical values

$$\kappa_a H^2 = 0,12\ \mathrm{erg/cm^3},\ K_{11} q_1^2 = 4 \cdot 10^2\ \mathrm{erg/cm^3}$$

i.e., the magnetic restoring force can be neglected. Indeed, the interference structure period is $\lambda_{int} = 2\pi/|\mathbf{q}_1| \sim 20\ \mu m$, which is much smaller than the magnetic coherence length in the NLC $l_M = \sqrt{K_{11}/\kappa_a H^2}$ for such values of H. Under such conditions, the expression for X_{ef} can be written in the form:

$$X_{ef} \approx X_{ef}(\mathbf{q}_1) = \frac{(n_e^2 - n_o^2)^2}{64\pi^2 K_{11} q_1^2} = \frac{\lambda_{vac}^2 (n_e + n_o)^2}{256\pi^4 K_{11}}. \tag{17}$$

It is interesting to note that Eq. (17) for X_{ef} turns out to be independent of the value of the anisotropy $n_e - n_o$. The reason for this is that with increasing $n_e - n_o$, the orientating forces of light fields increase (proportionally to $\varepsilon_a^2 = (n_e^2 - n_o^2)^2$, but the Frank restoring force increases approximately at the same rate due to the fact that the interference period becomes smaller, $\lambda_{int} = \lambda_{vac}(n_e - n_o)$. The numerical estimation from Eq. (17) gives $X_{ef} = 0.5 \cdot 10^{-5}\ \mathrm{cm^3/erg}$. This non-linearity is approximately 10^6 times stronger than the nonlinearity of CS_2.

In the Born approximation, the reversal coefficient $R = |E_4|^2/|E_3|^2$ may be obtained from Eq. (16a) and is equal to $R = |2\pi\omega/cn_0 L X_{ef} E_1 E_2|^2$. Let us estimate the power density of the reference waves which is necessary to obtain $R \sim 1$ at the cell thickness $L = 10^{-2}$ cm. For $P_1 = P_2 = P$ one obtains $P \sim 10^4\ \mathrm{watt/cm^2}$. The establishment time can be estimated from Eqs (7) as $\tau \sim \gamma/K_{11}q^2$, and for given conditions and $\gamma \approx 5.10^2$ puas[11] we have $\tau \sim 10^{-4}$ s.

Let us estimate also the role of thermal effects which are accumulated during that time. Even for a relatively large absorption coefficient, $\beta \sim 0.1\ \mathrm{cm^{-1}}$, the temperature increase will be $\Delta T \sim P\tau\beta/\rho c_p \sim 0.1$ deg for $P \sim 10^4$ watt/cm^2 and $\rho c_p \sim 1\ \mathrm{j/cm^3 \cdot deg}$. In relation to $dn/dT \sim 10^{-3}\ \mathrm{deg^{-1}}$ [17], the contribution of thermal effects to the light field phase $(\omega/c)(dn/dT)L\Delta T$ will be of the order of 0.1 rad. This means that the effects of thermal self-action do not distort the WFR–FWI process. Estimations for other possible experimental geometries may also be obtained. If the signal wave propagates almost perpendicular to the reference waves, the non-linearity falls by about $(n_0/(n_e - n_o))^2$ times (as the square of the relation of the $|\mathbf{q}|$ values). This gives a factor of ~ 25 times for PAA. The power density which is necessary for achievement of $R \sim 1$ increases and the establishment time decreases by the same factor. Therefore, even in that case, the thermal effects would not disturb the observation of the wave front reversal.

POLARIZATION PROPERTIES OF SELF-ACTION AND SELF-FOCUSING IN A NLC

Let us consider now the light self-focusing in a NLC. As mentioned above, the orientational mechanism of non-linearity works only for oblique incidence of the extraordinary light wave. Consider the incidence of light at the planar oriented NLC cell, so that the wave vector $\mathbf{K}$ constitutes the angle α with the x-axis inside the medium (Figure 3). Then, for $n_e - n_0 \ll 1$, the polarization vector of the e-type light field can be written in the form: $\mathbf{e}_e = \{-\sin\alpha, 0, \cos\alpha\}$. Thus, we shall follow the light field $\mathbf{E}(\mathbf{r})$ in the form

$$\mathbf{E}(\mathbf{r}) = \mathbf{e}_e E(\mathbf{r}) \exp\{i\mathbf{K}_e\mathbf{r}\}, \tag{18}$$

where $E(\mathbf{r})$ is a slowly varying amplitude depending on both the transverse and longitudinal coordinates. Insertion of Eq. (18) and of the particular form of the polarization unit vector $\mathbf{e}_e$ into Eq. (10) gives $\delta\varepsilon_{ik}(\mathbf{r}) \sim |E(\mathbf{r})|^2$.

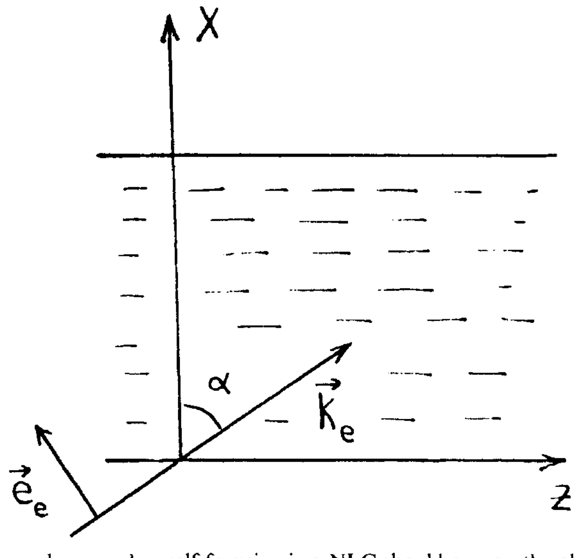

FIGURE 3 The only case when self-focusing in a NLC should occur—the oblique incidence of the extraordinary wave.

For the effective change of refractive index we obtain

$$\delta n = \frac{\delta\varepsilon_{ef}}{2n_o} = \frac{(\mathbf{e}_e)_i\delta\varepsilon_{ik}(\mathbf{e}_e)_k}{2n_o} = \frac{\varepsilon_a^2 \sin^2\alpha\cos^2\alpha}{8\pi n_o \kappa_a H^2}|E(\mathbf{r})|^2. \tag{19}$$

The local character of the coupling of $\delta\varepsilon_{ef}(\mathbf{r})$ and $|E(\mathbf{r})|^2$ in Eqs. (10) or (19) arises from neglecting of the Frank energy K_{ii}/l^2; here l is the characteristic size of the light field inhomogeneities. If we assume $l_M = \sqrt{K_{ii}/\kappa_a H^2} \sim 20\ \mu$m, then the coupling nature of Eq. (19) will be true, if typical sizes of the field inhomogeneities are greater than 20 μm. This assumption is valid for most of the experimental situations.

If we write the relation (19) in the form $\delta\varepsilon_{ef} = \varepsilon_2|E|^2$, then, for $\alpha = 45°$ and $H = 10^3$ gauss we obtain for PAA $\varepsilon_2 = 0.14$ cm^3/erg. This quantity is about 9 orders of magnitude greater than the non-linearity for CS_2 (note, that in this section we characterize non-linearity by the constant ε_2, while in the previous section the constant X was used). There would be a relation of the type $\varepsilon_2 = 4\pi X$ for a substance with local non-linearity. The phase shift in the cell of thickness L, for the type of geometry shown in Figure 3, due to $\delta\varepsilon_{ef}$ from Eq. (19) is

$$\delta\phi = \frac{\omega L\delta\varepsilon_{ef}}{2cn\cos\alpha} = \frac{\varepsilon_a^2 L\cos\alpha\sin^2\alpha}{4n\kappa_a H^2\lambda_{\text{vac}}}|E(\mathbf{r})|^2. \tag{20}$$

This quantity has a maximum for the angle $\alpha = 53°$. However, for $\alpha = 45°$, the value of $\delta\phi$ is different by only 9 %. For $\lambda_{\text{vac}} = 0.7\ \mu$m and $L = 10^{-2}$ cm, the value of $\delta\phi$ is ~ 1 rad for the power density $P \sim 1$ watt/cm^2. The condition $\delta\phi \sim 1$ rad corresponds to a change of beam divergence by an order of diffraction after the beam passes through the non-linear medium. This corresponds to the threshold of the external self-focusing.

If the cell has a greater thickness, the beam can be trapped inside the medium. The critical power of the self-focusing estimated by the known formula[18]

$$W_c = \lambda_{\text{vac}}^2 c n_o / 32\pi^2\varepsilon_2$$

for $\varepsilon_2 = 0.14$ cm^3/erg is $4.9 \cdot 10^{-7}$ watt. The beam of power W and transverse sizes a must be trapped at the length[18]

$$l = \frac{Ka^2}{\sqrt{W/W_c - 1}},$$

which is 40 μm for $\lambda_{\text{vac}} = 0.7\ \mu$m, $a = 0.1$ cm, $W = 0.49$ watt.

The orientational non-linearity establishment time $\tau \sim \gamma/\kappa_a H^2$ for the example considered is $\tau \sim 0.5$ s, which is quite a large quantity. However, since the required powers are quite small, the effects we are interested in can be observed in a clear form without the masking action of thermal effects.

Above we considered the case of relatively weak light fields, when $\kappa_a H^2 \gg \varepsilon_a|E|^2/8\pi$. In the opposite case, when $\kappa_a H^2 \gtrsim \varepsilon_a|E|^2/8\pi$, the orientational action of the light field surpasses the action of the magnetic orientational force. Then, in a stationary regime, the director is oriented along the direction of the light wave electrical vector. As a result, the tilted nematic structure arises from the initial planar orientation of the NLC. The local angle between the director and Z-axis is defined from the equilibrium equation and is:

$$\phi(\mathbf{r}) = \tfrac{1}{2}\arctan\frac{(\varepsilon_a/8\pi)|E(\mathbf{r})|^2\sin^2\alpha}{\kappa_a H^2 + (\varepsilon_a/8\pi)|E(\mathbf{r})|^2\cos^2\alpha} + \frac{\pi}{2}m. \tag{21}$$

In the case under consideration, $\varepsilon_a > 0$, and we must put $m = 0$. Thus, there is a gradient of effective refractive index only on the wings of a beam. In the beam centre, however, $\delta\mathbf{n}$ comes to the plate. As a result, only the extreme part of the beam is self-focused, while the central part passes without change in direction. In the intermediate non-stationary case, the self-focusing picture becomes more complicated.

Numerically the light power density corresponding to saturation $\sim 1(\delta\phi \sim 22°$ for $\alpha = 45°)$ is $P \sim 6.10^2$ watt/cm^2.

CONCLUSIONS

Thus, the extraordinary strong effects of cubic optical non-linearity in an oriented NLC due to director reorientation under the action of light fields are predicted in the present paper. The estimations show a real possibility of the observation of self-focusing in a NLC at a power density level $W \sim 1$ watt/cm^2, $\tau \lesssim 1$ s, and WFR realization for a power density level $P \sim 10^4$ watt/cm^2, $\tau \lesssim 10^{-2}$ s. There are also many other important problems where the usage of the large orientational non-linearity of a NLC can radically change the situation. For example, note the bistable optical devices constructed on the basis of the Fabry-Perot resonator.

We hope that the predicted effects of orientational non-linearity of a NLC will be discovered in the near future.†

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