Self-focusing of laser radiation in the course of the Fréedericksz transition in the nematic phase of a liquid crystal

A. S. Zolot'ko, V. F. Kitaeva, N. N. Sobolev, and A. P. Sukhorukov

P. N. Lebedev Physics Institute, Academy of Sciences of the USSR, Moscow
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An aberration theory of self-focusing of light beams is developed for a homotropically oriented nematic liquid crystal. Theoretical and experimental results are compared in the case of normally incident narrow beams (when the transverse dimensions of a beam are comparable with the thickness of a crystal). A qualitative agreement is obtained.

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1. INTRODUCTION

Among the great variety of nonlinear mechanisms of self-interaction of light beams (Kerr effect, a electrostriction, heating, etc.[1-3]), there has been a considerable interest in the specific change in the refractive index of a nematic liquid crystal (NLC) due to the distortion of the director field under the action of the electric field of a laser beam.[4,5] Reorientation of the NLC director, known as the Fréedericks transition, was discovered,in the thirties employing a static magnetic field,[6] and it has been observed subsequently in static and low-frequency electric fields.[7,8] Quite recently the Fréedericksz transition was observed when the electric field of a light wave interacted with an NLC.[5] All the main features of the transition were observed in this last case: there was a threshold of the effect in the $\mathbf{E} \perp \mathbf{n}_0$ case, the degree of reorientation of the director depended on the electric field intensity $\mathbf{E}$ and on the angle between $\mathbf{E}$ and the unperturbed director $\mathbf{n}_0$, etc. A theoretical analysis of a number of topics related to the Fréedericksz transition in the field of a light wave was reported later.[9]

Reorientation of the director alters the optical properties of an NLC. In fact, and NLC is uniaxial with the optic axis at a point $\mathbf{r}$ given by the director $\mathbf{n}(\mathbf{r})$. A change in the director orientation naturally alters the orientation of the local optic axis and this changes the refractive index of the extraordinary wave at this point. Consequently, self-interaction effects may appear in an NLC. In an NLC with a positive optical anisotropy the indiced change in the refractive index results in self-focusing of the transmitted light beam. It should be stressed that in the $\mathbf{E} \perp \mathbf{n}_0$ case there is a specific self-focusing threshold field which is due to the Fréedericksz transition threshold (this makes it possible to distinguish it from the self-focusing threshold associated with the suppression of diffraction[1-3]).

Several investigations have already been made of the self-focusing effect in NLCs. For example, self-focusing of an He–Ne laser beam in a planar sample of complex composition was investigated in Ref. 4 for small angles of rotation of the director in the $\mathbf{n} \cdot \mathbf{E} \neq 0$ case (a local reorientation mechanism was considered and saturation was ignored). The saturation effects in the case of large angles of rotation of the director in strong optical fields were considered theoretically in Ref. 10. The Fréedericksz transition of the nematic phase of an octylcyanobiphenyl liquid crystal, investigated using an Ar^+ laser beam,[5] was also accompanied by self-focusing. In this case the nonlinear divergence of the beam reached 40° for a crystal thickness of $L = 150$ μ and a laser power of $P = 150$ mW. A characteristic aberration structure (alternation of bright and dark rings, whose number and dimensions depended on the beam power) was observed in the transverse cross section of the beam. Similar results were reported also for MBBA liquid crystals.[11] Clearly, the same orientational self-interaction mechanism was observed in Ref. 12.

We shall develop an aberration theory of self-focusing of light beams in a homotropically oriented NLC subjected to an external magnetic field. The nonlinear response of a liquid crystal will be considered for an arbitrary angle of incidence of wide laser beams (this will be done using a local model) and of narrow beams of normal incidence (this will be done using a nonlocal relationship between the angle of rotation of the director and the electric field vector). In both cases we shall determine the dependence of the number of the aberration rings and of the nonlinear divergence on the laser beam power. We shall compare the results of theoretical calculations with experimental data.

2. REORIENTATION OF THE DIRECTOR OF A NEMATIC LIQUID CRYSTAL UNDER THE ACTION OF A BOUNDED LIGHT BEAM

Let us assume that there is an NLC film of thickness L in which the director is oriented at right-angles to the film boundaries (Fig. 1). We shall assume that the

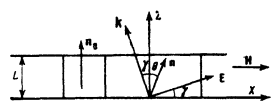

FIG. 1. Experimental geometry: $\mathbf{k}$ is the wave vector of the incident beam, $\mathbf{n}$ is the director, $\mathbf{n}_0$ is the unperturbed director, $\mathbf{E}$ is the electric field vector, $\mathbf{H}$ is the magnetic field vector, and L is the crystal thickness.

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director $\mathbf{n}_0$ is oriented along the Z axis and that a static magnetic field H is applied along the X axis which lies in the plane of Fig. 1.

The behavior of a unit vector $\mathbf{n}(\mathbf{r})$ ($n_\alpha n_\alpha = 1$) in the presence of electric E and magnetic H fields is described by the system of equations[7]

$$\frac{\partial F}{\partial n_\alpha} - \partial_\beta\left(\frac{\partial F}{\partial(\partial_\beta n_\alpha)}\right) = \lambda(\mathbf{r})\, n_\alpha \tag{1}$$

subject to the following conditions on the plane boundaries:

$$n_z|_{z=0} = n_z|_{z=L} = 1. \tag{2}$$

Here, n_α is the projection of $\mathbf{n}(\mathbf{r})$ onto the α axis ($\alpha = x, y, z$); $\partial_\beta = \partial/\partial x_\beta$; $\lambda(\mathbf{r})$ is an unknown function (undetermined Lagrange multiplier); $F(n_\alpha, \partial_\beta n_\alpha)$ is the free-energy density which has the following form in the one-constant approximation:

$$F = \frac{K}{2}(\mathrm{div}^2\,\mathbf{n} + \mathrm{curl}^2\,\mathbf{n}) - \frac{\Delta\varepsilon(\mathbf{nE})^2}{8\pi} - \frac{\chi_a(\mathbf{nH})^2}{2} \tag{3}$$

where K is the Frank elastic modulus; $\Delta\varepsilon = \varepsilon_\parallel - \varepsilon_\perp$ is the optical anisotropy; χ_a is the anisotropy of the magnetic susceptibility.

Writing down the electric field intensity in terms of a slowly varying complex amplitude $\mathbf{A}$,

$$\mathbf{E} = {}^1/_2\mathbf{A}(\mathbf{r})\exp[i(\mathbf{kr} - \omega t)] + \text{c.c.}, \tag{4}$$

we find after substitution of Eq. (4) into Eq. (3) and subsequent averaging with respect to time that the free energy density is given by

$$F = \frac{K}{2}(\mathrm{div}^2\,\mathbf{n} + \mathrm{curl}^2\,\mathbf{n}) - \frac{\Delta\varepsilon|(\mathbf{nA})|^2}{16\pi} - \frac{\chi_a(\mathbf{nH})^2}{2}. \tag{5}$$

If the optical wave is polarized in the XZ plane, the rotation of the director considered in the one-constant approximation again occurs parallel to the plane. In this case the orientation of the director can be described by a single angular coordinate $\theta(\mathbf{r})$ (Fig. 1):

$$n_x = \sin\theta(\mathbf{r}), \quad n_y = 0, \quad n_z = \cos\theta(\mathbf{r}). \tag{6}$$

The quantity $\theta(\mathbf{r})$ is described by the following expression derived from Eqs. (1), (2), and (5):

$$\Delta\theta + \frac{1}{\xi_E^2}\sin(\theta+\gamma)\cos(\theta+\gamma) + \frac{1}{\xi_H^2}\sin\theta\cos\theta = 0,$$

where

$$\theta|_{z=0} = \theta|_{z=L} = 0;$$

$$\frac{1}{\xi_E^2} = \frac{\Delta\varepsilon|A|^2}{8\pi K}, \quad \frac{1}{\xi_H^2} = \frac{\chi_a H^2}{K}.$$

The above equation can be rewritten in the form

$$\Delta\theta + \frac{1}{\xi^2}\sin(\theta+\beta)\cos(\theta+\beta) = 0, \tag{7}$$

where

$$\theta|_{z=0} = \theta|_{z=L} = 0;$$

$$\frac{1}{\xi^4} = \frac{1}{\xi_E^4} + \frac{1}{\xi_H^4} + \frac{2\cos 2\gamma}{\xi_E^2\xi_H^2}, \quad \sin 2\beta = \frac{\xi^2}{\xi_E^2}\sin 2\gamma.$$

Solution of Eq. (7) for a light wave with a spatial inhomogeneity is a difficult task. We shall consider two special but important cases. We shall denote the characteristic transverse size of a beam by w.

a) Propagation of a wide beam at an arbitrary angle ($L \ll w$)

If the amplitude of a light wave varies sufficiently slowly in space, i.e., if the coherence length ξ is much smaller than the transverse beam size w, Eq. (7) can be solved in the quasihomogeneous approximation assuming that the parameter ξ is independent of the spatial coordinates. The solution of Eq. (7) for a homogeneous field is well known.[13]

$$\sin(\theta+\beta) = \sin(\theta_m+\beta)\,\mathrm{sn}\left(\frac{z+z_0}{\xi}, \sin(\theta_m+\beta)\right), \tag{8}$$

where su(n, k) is an elliptic sine; θ_m is the angle of deviation of the director at the point $z = L/2$, given by

$$\frac{L}{2\xi} = \mathcal{F}\left(\frac{\pi}{2}, \sin(\theta_m+\beta)\right) - \frac{z_0}{\xi}; \tag{9}$$

$\mathcal{F}(\varphi, k)$ is an incomplete elliptic integral of the first kind;

$$\sin\zeta_0 = \frac{\sin\beta}{\sin(\theta_m+\beta)}, \quad \frac{z_0}{\xi} = \mathcal{F}(\zeta_0, \sin(\theta_m+\beta)).$$

If $\beta = 0$, there is a threshold for the appearance of the distortion of the director. The threshold field is found from Eq. (9) which in this case becomes $L/\xi = \pi$. Immediately above the threshold (when $\theta_m \ll 1$) the distribution of the director is

$$\theta = \theta_m \sin(\pi z/L).$$

b) Normal incidence of a narrow beam ($L \sim w, \gamma = 0$)

In this case we cannot regard the rotation of the director as simply locally related to the field intensity. The distribution of the director in a Gaussian field of a beam

$$E = E_0\exp(-\rho^2/w^2) \tag{10}$$

(ρ is the distance from the beam axis) can be found by the variational method. Substituting Eq. (6) into Eq. (5), we find that Eq. (10) allows us to determine the free energy density

$$F = \frac{K}{2}\left\{\left(\frac{\partial\theta}{\partial\rho}\right)^2 + \left(\frac{\partial\theta}{\partial z}\right)^2 - \left[\frac{1}{\xi_0^2}\exp\left(-\frac{2\rho^2}{w^2}\right) + \frac{1}{\xi_H^2}\right]\sin^2\theta\right\}, \tag{11}$$

where $1/\xi_0^2 = \Delta\varepsilon E_0^2/8\pi K$. In an equilibrium state the free energy of a crystal

$$\Phi = 2\pi\int_0^L dz\int_0^\infty \rho F(\rho, z)\, d\rho \tag{12}$$

has a minimum. The function $\theta(\mathbf{r})$ will be found in the form

$$\theta(\rho, z) = B\exp(-\rho^2/\alpha^2)\sin(\pi z/L), \tag{13}$$

where B and α are variable parameters. We shall expand $\sin^2(\theta)$ in Eq. (11) as a series in terms of θ^2 and we shall retain only the first two terms of the expansion. Substituting Eq. (13) into the resultant expression and integrating over the volume of the sample, we find from Eq. (12) that

$$\Phi=\frac{\pi KLB^2}{4}\left\{1+\frac{\pi^2\alpha^2}{2L^2}\left(1-\delta_E^2\frac{1}{1+\alpha^2/w^2}-\delta_H^2\right)+\frac{B^2\pi^2\alpha^2}{16L^2}\left(\delta_E^2\frac{2}{2+\alpha^2/w^2}+\delta_H^2\right)\right\}, \tag{14}$$

where $\delta_E=L/\pi\xi_E$, $\delta_H=L/\pi\xi_H$.

Having determined the parameter α from the condition $\partial\Phi/\partial(\alpha^2)=0$, we find that

$$\alpha^2=w^2\left[\frac{\delta_E}{(1+\delta_H^2)^{1/2}}-1\right]. \tag{15}$$

Substituting Eq. (15) into Eq. (14) and retaining the terms quadratic in B, we can use the condition $\Phi<0$ to find the threshold for the appearance of the distortion of the director under the action of a Gaussian beam:

$$E_{\text{th}}=E_{\text{th hom}}[(1-\delta_H^2)^{1/2}+g],\quad E_{\text{th hom}}=\frac{\pi}{L}\left(\frac{8\pi K}{\Delta\varepsilon}\right)^{1/2},$$

where $E_{\text{th hom}}$ is the transition threshold in the homogeneous field of a plane wave; $g=\sqrt{2L/\pi w}$ is a geometric factor. In the absence of a magnetic field, we have

$$E_{\text{th}}=E_{\text{th hom}}(1+g). \tag{16}$$

Substituting Eq. (15) into Eq. (14) and minimizing the resultant expression with respect to B, we find that when $H=0$, then

$$B=\left(2\frac{[(\delta_E-1)^2-g^2](\delta_E+1)}{(\delta_E-1)\delta_E^2}\right)^{1/2}.$$

If $\eta=(E-E_{\text{th}})/E_{\text{th}}\ll 1$, we obtain

$$B=2\eta^{1/2}G^{1/2},\quad G=(2+g)(1+g)^{-1}.$$

Thus, the required variational function is

$$\theta(\mathbf{r})=2\eta^{1/2}G^{1/2}\exp\left\{-\frac{\rho^2}{w^2[g+\eta(1+g)]}\right\}\sin\frac{\pi z}{L}. \tag{17}$$

3. NONLINEAR ABERRATIONS IN THE CASE OF SELF-INTERACTION OF A LIGHT BEAM IN A NEMATIC LIQUID CRYSTAL

We shall now use the director distribution obtained above (Sec. 2) to find the parameters of a light beam transmitted by an NLC film. The refractive index of an extraordinary wave traveling at an angle γ to the Z axis depends on the angles γ and $\theta(\rho,z)$ and it is given by

$$n_e(\gamma,\theta)=\frac{n_0n_e}{[n_0^2\sin^2(\theta+\gamma)+n_e^2\cos^2(\theta+\gamma)]^{1/2}}, \tag{18}$$

where $n_0^2=\varepsilon_\perp$, $n_e^2=\varepsilon_\parallel$. If we regard an NLC with this distribution of n_e as a thin nonlinear lens, the phase of the light beam transmitted by the crystal is

$$S=\frac{2k_0}{\cos\gamma}\int_0^{L/2}n_e(\gamma,\theta)\,dz$$

(where $k_0=2\pi/\lambda_0$ is the wave vector in vacuum) or, using Eq. (18), we find that

$$S=\frac{2k_0n_0n_e}{\cos\gamma}\int_0^{L/2}\frac{dz}{[n_0^2\sin^2(\theta+\gamma)+n_e^2\cos^2(\theta+\gamma)]^{1/2}}. \tag{19}$$

A qualitative dependence of the phase S on the distance ρ of a given ray from the beam axis is shown in Fig. 2a. The angle of deviation ψ of a ray is[1]

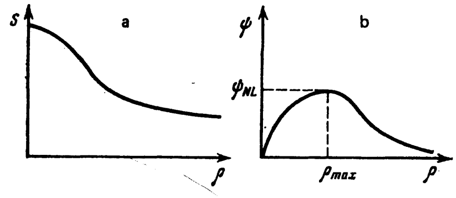

FIG. 2. Nature of the dependence of the phase advance (a) and of the angle of deviation (b) of rays in a beam on the transverse coordinate of the beam.

$$\psi=\frac{1}{k}\frac{\partial S}{\partial\rho} \tag{20}$$

(see Fig. 2b). In this case, we have

$$\psi=\frac{k_0n_0n_e(n_e^2-n_0^2)}{k\cos\gamma}\int_0^{L/2}\frac{\sin(2\theta+2\gamma)\,\partial\theta/\partial\rho}{[n_0^2\sin^2(\theta+\gamma)+n_e^2\cos^2(\theta+\gamma)]^{3/2}}dz. \tag{21}$$

Using $\Delta n=n_e-n_0\ll n_e, n_0$, we find that Eq. (21) can be simplified to

$$\psi=\frac{2\Delta n}{n_0\cos\gamma}\int_0^{L/2}\sin(2\theta+2\gamma)\frac{\partial\theta}{\partial\rho}dz. \tag{22}$$

At those points in the beam where $\xi\lesssim L$, and $\theta=\pi/2-\gamma$, we can then expect an increase in the beam power to result in effective deviation of the rays further from the axis.

Intersection of rays gives rise to an aberration pattern.[1] The total number of the aberration rings is related in a simple manner to the phase advance:

$$N=\frac{S(\rho=0)-S(\rho=\infty)}{2\pi}. \tag{23}$$

In the absence of a magnetic field but in electric fields sufficiently high for the director on the beam axis oriented parallel to the electric field, we find that the number of rings is

$$N_{max}=\frac{k_0n_en_0L}{2\pi\cos\gamma}\left[\frac{1}{n_0}-\frac{1}{(n_0^2\sin^2\gamma+n_e^2\cos\gamma)^{1/2}}\right]. \tag{24}$$

The angular magnitude of the outer aberration ring ψ_{NL} (Fig. 2b), which represents the total nonlinear beam divergence, can be found by differentiating Eq. (20) with respect to ρ, equating the derivarive to zero, finding hence the coordinate ρ_{max} of a ray with the greatest deviation, and substituting ρ_{max} into the equation for ψ.

Specific expressions for the main parameters of nonlinear aberrations (total nonlinear divergence and number of aberration rings) in the cases a and b considered in the present study are naturally governed by the distribution of the director field in these two cases, i.e., by Eqs. (8) and (17).

In the case of a narrow light beam incident normally on a crystal (i.e., when $\gamma=0$), which corresponds—in particular—to the experimental conditions in Ref. 5, these expressions are

$$N=2\Delta n\frac{L}{\lambda}G\left(\frac{E-E_{\text{th}}}{E_{\text{th}}}\right), \tag{25}$$

$$\psi_{NL}=\frac{4\Delta nLG}{e^{1/2}wn_0}(E-E_{\text{th}})/E_{\text{th}}\left[g+\left(\frac{E-E_{\text{th}}}{E_{\text{th}}}(1+g)\right)\right]^{1/2}. \tag{26}$$

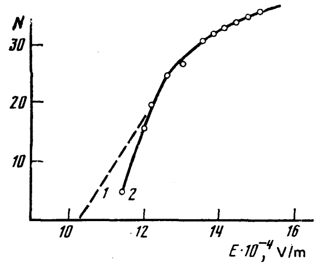

FIG. 3. Dependence of the number of the aberration rings on the electric field of a light wave on the beam axis. Crystal thickness $L = 150\ \mu$. 1) Theoretical dependence; 2) experimental dependence.

4. COMPARISON OF THEORETICAL CALCULATIONS AND EXPERIMENTAL RESULTS OBTAINED FOR OCTYLCYANOBIPHENYL CRYSTALS

The aberration structure of a light beam due to reorientation of the director in a field of a light wave was first observed by Zolot'ko *et al.*,[5] who investigated in detail the nature of the divergence of a beam from an argon ion laser transmitted by the homotropically oriented nematic phase of a liquid crystal of octylcyanobiphenyl (OCBP).

A cell with OCBP was placed at a constriction of a laser beam. The diameter of the constriction, calculated using the geometric parameters of the laser resonator and optical system, was 32 μ. Samples were 150 and 50 μ thick.

We shall now compare the experimental results of Ref. 5 with the theoretical estimates obtained in our study. The parameters of an OCBP crystal needed for this purpose were taken from Refs. 14 and 15. In addition to the results given in Ref. 5, we plotted the experimental dependences of the number of the aberration rings and the nonlinear divergence on the electric field intensity. These are plotted in Fig. 3 and 4. These figures include also (curves 1) the theoretical dependences. These dependences are calculated for fields slightly above the threshold value ($\eta < 0.2$), i.e., they are within the range of validity of Eqs. (25) and (26). It is clear from Figs. 3 and 4 that the theoretical curves are qualitatively correct representations of the dependences of the number of aberration rings and of the divergence on the electric field intensity on the beam axis. However, it should be pointed out that in our approximation the theory predicts a slower rise of N and ψ on increase in the electric field than that actually observed. This may be due to, for example, the fact that self-focusing distorts the electric field of a light beam which we ignored in determining the distribution of the director, or due to the fact that the attachment of molecules to the walls is not sufficiently rigid.[8] The variational method used in the present study suffers from its own errors.

The maximum number of aberration rings estimated from Eq. (24) is 42. In the saturation region the experimental patterns exhibited up to 39 rings.

The threshold of the Fréedericksz transition estimated from Eq. (16) is 10.3×10^4 V/m. The experimental value of this threshold (Fig. 3) is 11.3×10^4 V/m. The ratio of the thresholds for crystals of thickness 50 and 150 μ is approximately 3. The same ratio calculated from Eq. (16) is 2.7.

It follows that the theoretical results obtained in our study using a simple model are in reasonable agreement with the experimental data.

5. CONCLUSIONS

A theory of self-focusing in the course of the Fréedericksz transition presented in the present paper should be developed further. This applies to the description of nonlinear properties of NLCs and to the process of wave propagation. In addition to analytic methods, it would be undoubtedly interesting to apply numerical methods.

We shall conclude by noting that self-focusing in the course of the Fréedericksz transition is a new and very important technique for investigating liquid crystals. It makes it possible to study, above all, the dynamics of the Fréedericksz transition in the field of a light wave; in particular the transition threshold can be used to estimate the elastic constants of liquid crystals, etc. A definite advantage of the method is the fact that the effect is observed for relatively low laser radiation powers.

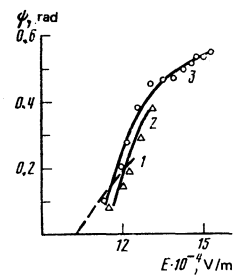

FIG. 4. Dependence of the divergence of the laser beam on the electric field: 1) Theoretical curves; 2) results of measurements on the minor semiaxis of the outer aberration ring; 3) on the major semiaxis. Crystal thickness $L = 150\ \mu$.

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