Enhancement and suppression of light-induced orientational effects and time-dependent instabilities resulting from competition between two waves in a nematic liquid crystal with a threshold nonlinearity

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An analysis is made of the threshold orientational effects in a nematic liquid crystal subjected to a laser field when two waves with orthogonal polarizations propagate in a nonlinear medium. The pattern of instabilities is revealed for the first time and an analysis is made of the various states of such a system with enhancement and suppression of reorientation when two fields should compete with one another purely on geometric grounds. An experiment is carried out for this case. Analytic solutions are obtained for the first time for oscillatory regimes unstable in time, which are observed when a nematic liquid crystal is pumped continuously when circularly (elliptically) polarized light enters it.

1. INTRODUCTION

Excitation of modulated structures (three-dimensional gratings) by laser radiation in an anisotropic medium such as a liquid crystal gives rise, because of the self-interaction effects, to instabilities of nonlinear wave processes associated with the propagation of light (see, for example, Ref. 1). These instabilities are governed by the light-induced changes in the orientation of the liquid crystal director representing the average orientation of molecules in a sample, i.e., the local direction of its optic axis. The parameter which then varies is the angle ψ by which the director $\mathbf{n}$ deviates from its initial unperturbed orientation $\mathbf{n}_0$. It is important to note that in the case of nematic liquid crystals such reorientation has a threshold with respect to the intensity of the incident light (which is the result of competition between the orienting effect of the optical field and the tendency for the elastic forces of the medium to maintain the original orientation set by rigid boundary conditions). The strong anisotropy of a nematic liquid crystal (NLC) and the collective nature of the response of molecules to an external perturbation (which is a consequence of the elastic properties of the medium) are responsible for the very strong optical nonlinearity of the molecules.[2]

The topic currently of the greatest interest is that of wave phenomena in a strongly nonlinear optical medium under conditions when two waves with different (orthogonal) polarizations travel inside the medium. The competition between these waves and the mutual energy exchange give rise to regimes which are unstable in time: undamped oscillations of the intensity and polarization of the light transmitted by a system developed in the case of continuous pumping reaching the entry of the system.[3–6] The fundamental feature is the light-induced deformation of the structure of a liquid crystal which is described by two angles (ψ and φ) in orthogonal planes.[2] Each of these angles has its own spatial structure and its own characteristic growth time: the periods of the spatial structures which are established are different and so are the intensities of the polarization components. In particular, the angle ψ may describe an adiabatic deformation (with a characteristic scale $\Lambda \sim d$), whereas φ may describe a nonadiabatic deformation $[\Lambda \sim 1/(k_e - k_o) \ll d]$, where d is the thickness of the sample and $k_{e,o}$ are the wave vectors of the extraordinary and ordinary waves, respectively. These two angles of reorientation of the director determine two types of time-dependent instabilities. Firstly, these are oscillations of the characteristic aberration pattern of self-focusing which appears in transmitted light for a beam localized in space; they are due to oscillations of the angle ψ representing deviation from its original orientation.[3,5,6] Secondly, in addition to these oscillations, experiments on circularly polarized (incident) light have revealed also[5] oscillations of the polarization which are due to the precession of $\mathbf{n}$ about its local direction (the azimuthal angle φ is varied).

The exact solution of the problem of the appearance of these time-dependent instabilities has not yet been obtained.[5] Therefore, as pointed out already, the physics of the effect is definitely linked to the presence of two waves with orthogonal polarizations and the effective exchange of energy between them.[1] If we use the usual language nonlinear optics, we can describe the situation as four-wave parametric scattering of light: a dynamic grating of the refractive index is formed in the investigated medium.[7] The frequency shift between the interacting waves (within the limits of the width of the line representing the incident light) is then due to dissipation processes. An analysis of the effects of stimulated scattering of light in an NLC was recently made[8] using this approach.

We provide the first detailed analysis of the problem of competition between two waves with orthogonal polarizations interacting in a nonlinear anisotropic medium (in the form of an NLC) and we use the geometric-optics approximation. We first consider the simplest case when waves of the same polarization, but oriented at an angle 90° relative to one another propagate in an anisotropic nonlinear medium in such a way that they act on the NLC in opposite directions in the purely geometric sense (Sec. 2). In this case we demonstrate experimentally and theoretically that there are new regimes involving amplification and suppression of the net reorientation and we reveal unstable states of such a system. We also deal with the problem of excitation of instabilities inside an NLC when one wave is incident on a sample and the interaction of two components of the field inside a non-

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linear medium is due to the anisotropy of this medium. We obtain for the first time an analytic solution of the problem of excitation of oscillations in time (both of the aberration pattern and of the polarization) in the case of threshold reorientation of an NLC in the field of a circularly (elliptically) polarized light (Sec. 3). This solution enables us to explain all the results of a fundamental experiment reported in Ref. 5, including the occurrence of hysteresis.

2. ENHANCEMENT AND SUPPRESSION OF THE ORIENTATION OF A NEMATIC LIQUID CRYSTAL IN THE FIELD OF TWO WAVES WHEN THE DIRECTIONS OF THEIR LINEAR POLARIZATIONS ARE DIFFERENT

Experiment

We used the experimental setup shown as an inset in Fig. 1: two light waves with linear polarizations ($\mathbf{E}_1$ and $\mathbf{E}_2$) in the plane of incidence reached a sample obliquely at an angle of $2\alpha_0$ relative to one another, but symmetrically relative to $\mathbf{n}_0$. We used radiation from a cw YAG:Nd^{3+} laser ($\lambda = 1.06\ \mu$m) which was split into two beams that were focused inside a cell containing a nematic liquid crystal (4-*n*-pentyl-4'-cyanobiphenyl, usually abbreviated to 5CB). The ratio of the intensities of these two beams was $I_2/I_1 \approx 1.5$ or 2.7 (see the caption of Fig. 1). The thickness of the NLC layer was $d \approx 125\ \mu$m; its initial orientation was homeotropic ($\mathbf{n}_0 \| z$). We determined the nonlinear phase advance Φ^{nl} (represented by aberration rings in a transmitted light) experienced by a weak probe beam (provided by an He–Ne laser emitting at $\lambda = 0.633\ \mu$m), which appeared because of reorientation of the NLC director in the pump (strong) field provided by the neodymium laser.

In this experiment the NLC was subjected first to one pump beam (I_1); when a steady-state reorientation pattern was obtained, a second beam I_2 was directed to the sample. When the pattern was established in the field of both beams, one of the beams was shut off. The resultant steady-state and transient characteristics of Φ^{nl} obtained under different conditions are plotted in Fig. 1. Special measurements showed that as a result of reorientation the NLC director did not tilt out of the plane of incidence of the two waves ($\mathbf{E}_1\mathbf{E}_2$ plane), when one would also expect enhancement of the reorientation.

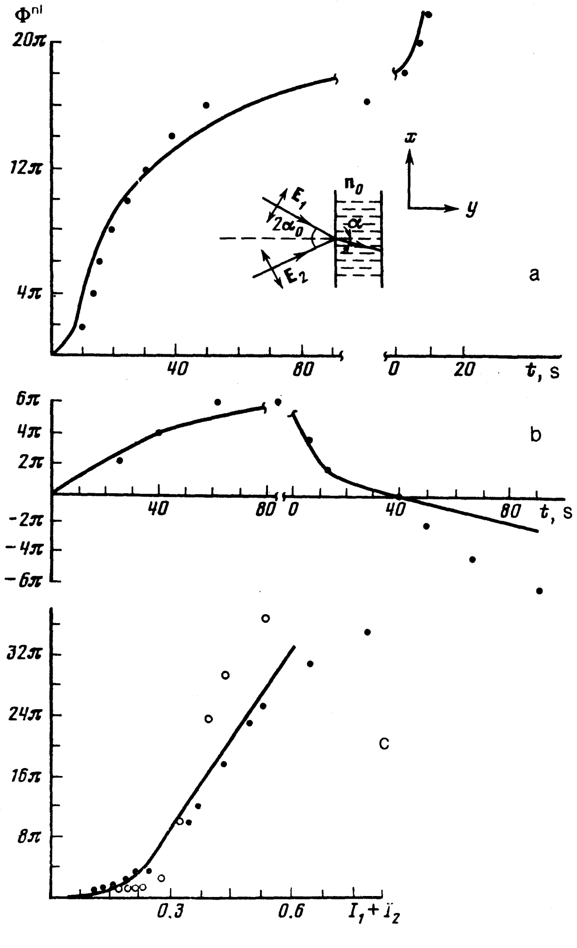

FIG. 1. Nonlinear phase advance Φ^{nl} experienced by a probe beam as a function of the time *t*, obtained for $I_1 = 280$ W/cm² and $I_2 = 700$ W/cm² (a) and $I_1 = 157$ W/cm² and $I_2 = 440$ W/cm² (b), and the dependence of the phase advance on the intensity ($I_1 + I_2$) of the incident light under steady-state conditions (c). The points are the experimental values and the curves are theoretical (calculations were carried out only in the range of validity of the theory: $\psi \ll 1$, $\Phi^{nl} \ll 36\pi$). Values of the parameters in Eq. (6): one beam acting: a) $A = -0.01$, $B = 0.03$, $C = -0.029$, $D = -0.035$; b) $A = -0.0075$, $B = 0.017$, $C = -0.05$, $D = -0.02$; two beams acting: a) $A = -0.05$, $B = -0.046$, $C = 0.08$, $D = 0.05$; b) $A = -0.02$, $B = -0.006$, $C = -0.01$, $D = 0.008$; c) results for two cases: ○) $I_2/I_1 = 1.5$;) ●) $I_2/I_1 = 2.7$. The calculated curve corresponds to the latter case (●).

Theory and calculations

1. Initial equations. The problem under discussion is the interaction of light waves with a variably anisotropic nonlinear medium, so that it is convenient to use the approach based on the geometric-optics approximation.[9] The standard calculation procedure is given in Ref. 10. In this case the reorientation occurs in one plane, defined by just one angle ψ by which the director deviates from its original orientation; the angle in question is assumed to be small, so that perturbation theory can be used.

The equation of motion of the director can in this case be reduced to

$$\frac{\partial}{\partial z}\left[\frac{\partial\psi}{\partial z}(1+K\sin^2\psi)\right]-K\sin\psi\cos\psi\left(\frac{\partial\psi}{\partial z}\right)^2 + \frac{\varepsilon_a\varepsilon_\perp^{1/2}}{c\varepsilon_\| K_{33}}[g_2(\psi)I_{2z}+g_1(\psi)I_{1z}]=\frac{\gamma}{K_{33}}\frac{\partial\psi}{\partial t}. \qquad (1)$$

Here, $\varepsilon_a = \varepsilon_\| - \varepsilon_\perp$ is the optical anisotropy ($\varepsilon_\|$ and $\varepsilon_\perp$ are the values of the permittivity along and across the director, respectively); K_{33} and $K \equiv (K_{11} - K_{33})/K_{33}$ are the elastic constants of the NLC; γ is the viscosity; c is the velocity of light; I_{1z} and I_{2z} are the z-components of the Poynting vectors for each of the waves, which are constants of motion (we shall ignore the interference between the waves resulting in distortions which are nonadiabatic in z and which disappear when this situation is averaged over the thickness);

$$g_{1,2}(\psi)=\mathrm{tg}\,\alpha_{1,2}/\cos\psi\left(1+\frac{\varepsilon_\perp}{\varepsilon_\|}\mathrm{tg}\,\alpha_{1,2}\,\mathrm{tg}\,\psi\right)\left(1+\frac{\varepsilon_\perp}{\varepsilon_\|}\mathrm{tg}^2\,\alpha_{1,2}\right)^{1/2},$$

$$\mathrm{tg}\,\alpha_{1,2}=\{\varepsilon_\|\sin\psi\cos\psi\pm[\varepsilon_\|-\sin^2\alpha_0+\mathrm{tg}^2\psi(\varepsilon_\perp-\sin^2\alpha_0)]^{1/2}\}/(\varepsilon_\|\cos^2\psi-\sin^2\alpha_0);$$

α_0 is the angle of incidence in air. The boundary conditions of the problem are stringent: $\psi(z=0) = \psi(z=d) = 0$. In the case of ψ we shall use the approximation of light-induced adiabatic deformations which appear in this geometry for each of the light waves:

$$\psi=\sum_{l=1}^{\infty} C_l \sin(\pi l z/d)$$

and, as is usual, we shall retain only the first term ($l=1$) of this expansion, which is energetically favorable:

$$\psi=\psi_m \sin(\pi z/d) \tag{2}$$

where ψ_m is the maximum angle of reorientation at the center of the sample ($z=d/2$).

The following relationship can be easily derived for the expression that contains the functions $g_{1,2}(\psi)$ in Eq. (1): expanding as a series and retaining terms up to $\sim\psi^3$, we find that

$$h^{1/2}[g_2(\psi)I_{2z}+g_1(\psi)I_{1z}]=(I_{2z}-I_{1z})\,\mathrm{tg}\,\alpha$$
$$+(I_{2z}+I_{1z})(2-h)\psi+(I_{2z}-I_{1z})(3\varepsilon_a/\varepsilon_\parallel-2)\psi^2\,\mathrm{tg}\,\alpha$$
$$+(I_{2z}+I_{1z})[6h^2-h-9(\varepsilon_\perp/\varepsilon_\parallel)h^2+(8-21h)]\psi^3(h-1)/6, \tag{3}$$

where

$$h=1+\frac{\varepsilon_\perp}{\varepsilon_\parallel}\mathrm{tg}\,\alpha,\qquad \mathrm{tg}^2\,\alpha=\frac{\varepsilon_\parallel}{\varepsilon_\perp}\frac{\sin^2\alpha_0}{\varepsilon_\parallel-\sin^2\alpha_0}.$$

2. *Qualitative description.* Some qualitative results can be obtained even from Eq. (3). For if $I_{1z}=I_{2z}$, only the terms proportional to ψ and ψ^3 remain in Eq. (3). Then, Eq. (1) reduces to the usual case of the threshold light-induced reorientation of an NLC caused by one normally incident linearly polarized wave of intensity $I_z=(I_{1z}+I_{2z})(2-h)h^{1/2}$ (compare with Ref. 11). We are considering here only determination of the threshold intensity. A specific selection of the orientation of the NLC on its substrate can ensure that the threshold-free reorientation condition $2-h<0$ is satisfied. The value of the threshold is given by

$$(I_{1z}+I_{2z})_{\text{th}}=K_{33}\varepsilon_\parallel c h^{1/2}\pi^2/\varepsilon_a\varepsilon_\perp^{1/2}(2-h)d^2. \tag{4}$$

Therefore, reorientation occurs if $2-h>0$, which can be rewritten in the form $\sin^2\alpha_0<\varepsilon_\parallel/2$. In the case of real NLCs, we have $\varepsilon_\parallel\sim3$, so that light-induced reorientation is possible for any value of α_0. It is important to note that in this case both fields tend to increase ψ. We shall analyze a different case for which $I_{1z}\neq I_{2z}$. The most interesting effect is linked to the relative importance of the first term (of zeroth order) in Eq. (3), which contains the difference $I_{2z}-I_{1z}$. At low light intensities it plays the dominant role (zeroth approximation) when the two fields are oppositely directed: the reorientation is weakened compared with the case of one field. At high intensities (high values of ψ) the two fields begin to enhance together the reorientation [the terms linear in ψ should then be included in Eq. (3)]. This enhancement of the reorientation effect occurs for angles

$$|\psi|>\left|\frac{(I_{2z}-I_{1z})\,\mathrm{tg}\,\alpha}{(I_{2z}+I_{1z})(2-h)}\right|.$$

When the intensity is increased still further, the mutual suppression and enhancement of the reorientation effect begins to be influenced also by those terms in Eq. (3) which are proportional to ψ^2 and ψ^3, respectively. This provides a qualitative explanation of the experimental results plotted in Fig. 1.

The feasibility of mutual enhancement of the reorientation in crossed fields (at first sight, from the purely geometric point of view we would expect suppression) is due to the fact that when an anisotropic system is considered from the microscopic point of view and an external field induces a polarization in this system, only the projection of the external field along the direction of the strongest polarization (**n**) is important and this projection acts as an external force independent of the mutual orientations of **E** and **n** (compare with Ref. 12).[1)] However, in the case of propagating waves there is one important feature which distinguishes this situation from the case of static fields: when two waves are added, we must include also the phase coefficients proportional to $\exp(i\mathbf{k}\cdot\mathbf{r})$; these coefficients give rise to spatially small (compared with d) interference terms which disappear as a result of averaging and the net effect is governed only by the term proportional to $(|E_1|^2+|E_2|^2)$, i.e., by the total intensity.

3. *General analysis.* We now carry out a more rigorous analysis of the possible reorientation regimes in the case under discussion, in a fairly general form.

It is convenient to write down Eqs. (1) and (2) in the form

$$d\psi_m/dt=A\psi_m^3+B\psi_m^2+C\psi_m+D, \tag{5}$$

where the coefficients of the various powers of ψ_m are

$$A=\frac{K_{33}}{\gamma}\left\{\frac{3}{4}a\left[\frac{1}{6}(5-9\delta)-\frac{h-1}{2}\left(h+\frac{11}{3}-5\delta-h\delta\right)\right]\cdot(I_{1z}+I_{2z})-\frac{K}{2}\left(\frac{\pi}{d}\right)^2\right\}$$
$$B=\frac{K_{33}}{\gamma}\frac{8a}{3\pi}\mathrm{tg}\,\alpha\left(3\frac{\varepsilon_a}{\varepsilon_\parallel}-2\right)(I_{2z}-I_{1z}),$$
$$C=\frac{K_{33}}{\gamma}\left[a(2-h)(I_{1z}+I_{2z})-\left(\frac{\pi}{d}\right)^2\right], \tag{6}$$
$$D=\frac{K_{33}}{\gamma}\frac{4a\,\mathrm{tg}\,\alpha}{\pi}(I_{2z}-I_{1z}),$$

and the parameters are $\delta=\varepsilon_\perp/\varepsilon_\parallel$, and $a=\varepsilon_a\varepsilon_\perp^{1/2}/K_{33}\varepsilon_\parallel c h^{1/2}$.

Let us assume that the initial distribution $\psi_{m,\text{in}}\equiv\psi_{\text{in}}$ determined by which field is first applied and by the material parameters of the medium) corresponds to the maximum angle of reorientation of **n** in the middle of a sample: at $t=0$ the second field is applied and the two fields interact with an NLC. Substituting a variable $x=\psi_m-\psi_{\text{in}}$, we can transform Eq. (5) to

$$dx/dt=F_1(x,\psi_{\text{in}})x+F_2(\psi_{\text{in}}), \tag{7}$$

where $F_2(\psi_{\text{in}})$ can be written in the form

$$F_2(\psi_{\text{нач}})=A\psi_{\text{in}}^3+B\psi_{\text{in}}^2+C\psi_{\text{in}}+D,$$

so that F_1 is now given by the right-side of Eq. (5). For $x\ll1$, the first term on the right-hand side of Eq. (7) can be ignored, for the sake of simplicity, and then integration yields

$$x=F_2(\psi_{\text{in}})t. \tag{8}$$

Therefore, if $F_2(\psi_{\text{in}})>0$, then $x=\psi_m-\psi_{\text{in}}$ increases with time (i.e., we can speak of an increase in ψ_m), whereas for $F_2(\psi_{\text{in}})<0$ the angle ψ_m decreases. The actual regime is

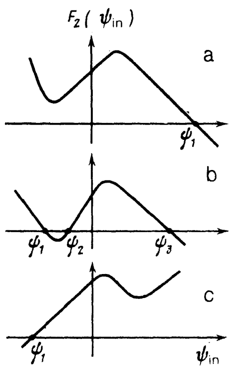

FIG. 2. Dependence $F_2(\psi_{in})$ for the cases of one ψ_1 (a corresponds to $A<0$; c corresponds to $A>0$) and three $\psi_{1,2,3}$ (b corresponds to $A<0$) steady-state real roots of Eq. (5). The explanation is given in text.

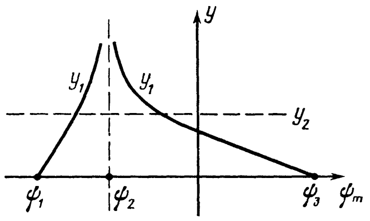

FIG. 3. Graphical solution of Eq. (9). The explanation is given in text.

determined by the values of the coefficients in front of the powers of ψ_{in}.

We shall begin with the case $A<0$. Then the dependence $F_2(\psi_{in})$ has the form shown in Fig. 2 for two cases: a) one real root ψ_1; b) three real roots $\psi_{1,2,3}$; in the case of two real roots we effectively have the case shown in Fig. 2a and $\psi_2 \equiv 0$ corresponds to an unstable state. It is easy to analyze the stability of these solutions, which correspond to the steady-state solutions of Eq. (5).

In fact, as in the $F_2(\psi_{in})>0$ case, we observe an increase in ψ_m [see Eq. (8)], so that in the case shown in Fig. 2a subject to the condition $\psi_{in}<\psi_1$ we can expect an increase in ψ_m, which tends to the value ψ_1; for $\psi_{in}>\psi_1$, we have $F_2(\psi_{in})<0$ and on reduction we reach the limit $\psi_m \rightarrow \psi_1$. Therefore, this case is relatively simple: as a function of time the director of an NLC always rotates toward the direction set by the angle ψ_1; depending on ψ_{in}, the application of the second field can enhance or suppress the reorientation.

The case shown in Fig. 2b is more interesting. The existence of three real roots $\psi_{1,2,3}$ is possible only if the total intensity $I_{1z}+I_{2z}$ exceeds a certain threshold value given by Eq. (4). The solutions ψ_1 and ψ_3 represent stable states, whereas ψ_2 represents an unstable state. It is easy to show that if $\psi_{in}>\psi_2$, the angle ψ_m rises approaching a steady-state value ψ_3; for $\psi_{in}<\psi_2$, the value of ψ_m decreases and we then have $\psi_m \rightarrow \psi_1$.

It follows that, irrespective of which of the intensities I_1 or I_2 is higher, the reorientation angle ψ_m can both increase or decrease. Although the actual existence of three states is typical of the usual geometry of the threshold reorientation of an NLC in an external field (Fréedericksz transition in a static or optical field—see Ref. 2),[2)] in the case under discussion there is an important difference. In fact, below the threshold value of the field the usual Fréedericksz transition always gives rise to a second state which, in spite of the presence of an external field, is stable ($\psi_m \equiv 0$); however, in the present case of the threshold-free reorientation in the presence of the second field when $\psi_{in} \neq 0$, we find that even if we ignore the dependence of ψ_2 on $I_{1z,2z}$ for $I_{1z}+I_{2z}<I_{th}$, the value of ψ_m may not approach ψ_2. Moreover, for $I_{2z}<I_{1z}$ (i.e., for $D<0$), then in the case of low light intensities ($C<0$)/steady-state value is $\psi_{st} \equiv -D/C<0$; the value of ψ_2 is always positive (ψ_2 is a real number). It should be noted that the transition to one of the stable states of the system may be responsible for optical bistability in those cases when there is hysteresis (see Ref. 13).

We shall consider briefly the case $A>0$. The regime with one solution (ψ_1) corresponds to an unstable state shown in Fig. 2c; for three solutions we have one stable state (ψ_2) and two unstable states ($\psi_{1,3}$); this should be compared with Fig. 2a. The first regime is particularly interesting: the absence of a stable steady-state solution which the system would tend to reach finally means essentially that time-dependent instabilities can appear; the answer as to what happens then can be found by an analysis of the problem in the next approximation.

4. Graphical solution. The solution of the problem can also be found directly by integrating Eq. (5). For example, in the case of three real roots and $A<0$, we obtain

$$\frac{|\psi_m-\psi_1|^{\alpha}|\psi_m-\psi_3|^{\beta}}{|\psi_m-\psi_2|^{\gamma}}=\frac{|\psi_{in}-\psi_1|^{\alpha}|\psi_{in}-\psi_3|^{\beta}}{|\psi_{in}-\psi_2|^{\gamma}}e^{At}. \qquad (9)$$

Here α, β, and γ are certain positive constants which are solely combinations of the quantities $\psi_{1,2,3}$. The solution of Eq. (9) with ψ_m can be obtained conveniently by a graphical method. Representing the left-hand side of Eq. (9) by y_1, we obtain the dependence shown by a continuous curve in Fig. 3; the right-side of Eq. (9) is represented by the straight line y_2 parallel to the ψ_m axis (dashed in Fig. 3). The solution is given by the points of intersection of y_1 and y_2.

In the case when $A<0$, an increase in t, beginning from $t=0$, causes a reduction in y_2 and in the limit $t\rightarrow\infty$ we have two stable steady-state solutions $\psi_{1,3}$. Depending on the value of ψ_{in} (to the left or right of the vertical line $y=\psi_2$), the final state of the system is governed by ψ_1 or ψ_3, respectively, i.e., the overall reorientation of the investigated NLC decreases or increases ($\psi_1<\psi_2<\psi_3$). The change in the orientation of the director toward the direction given by ψ_3 is now much faster.

For $A>0$ (corresponding to an increase in y_2 with time t), in principle we can have a jump between these two states ($\psi_m<\psi_2$ and $\psi_m>\psi_2$); its realization requires, firstly, that the solution be finite at $y=\psi_2$: the curves to the left and right of the $y=\psi_2$ line should join (we now have to include the higher terms in the expansion ψ_m). Secondly, the rates of switching from left to right and the rates of transition between different states on the same branch should be similar. The existence of such a jump in the case of transient orientation gives rise to oscillations in time and to instabilities.

In our analysis we shall be interested in real steady-state solutions (we obtained by numerical calculations of the general expressions without expanding in the small parameter ψ). The absence of these solutions is a clear sign that time-dependent instabilities appear in the system. However, the process of finding transient solutions is fairly complicated even numerically, and this has not yet been done.

5. Estimates and calculations. We shall now compare the theory and experiment. Using the parameters of our NLC crystal (5CB), for which $K_{33} \approx 4.4 \times 10^{-7}$ dyn,$\varepsilon_{\parallel} \approx 3.02$ and $\varepsilon_{\perp} \approx 2.31$ (Ref. 2), and assuming that $d = 125\ \mu$m, we find from Eq. (4) that $(I_{1z} + I_{2z})_{\rm th} \approx 320$ W/cm^2 (after allowance for the Fresnel reflection, this value becomes ≈ 360 W/cm^2). Therefore, the total intensity in our experiments did indeed exceed the threshold. Enhancement of the reorientation in the field of two beams (Fig. 1a) corresponded to $|\psi| > 0.12$.

Under our experimental conditions we were dealing with the case $A < 0$ (see the caption of Fig. 1). Then, the geometry with one beam corresponds to one real root of Eq. (5) ($\psi_1^{(1)}$ in Fig. 1 and $\psi_1^{(1)'}$ in Fig. 1b), whereas the case of two beams corresponds to three such roots $\psi_{1,2,3}^{(2)}$ (strong fields, Fig. 1a) or one root $\psi_1^{(2)}$ (weak fields, Fig. 1b). The steady-state values are determined by the stable solution $\psi_{1,3}$. The numerical values of these solutions are as follows: $\psi_1^{(1)} = -2.4$; $\psi_1^{(1)'} = -0.35$; $\psi_3^{(2)} = 1.17$; $\psi_2^{(2)} = -0.05$; $\psi_1^{(2)} = -1.54$; $\psi_1^{(2)'} = 1.00$.

The calculated curves corresponding to the light intensities used in our experiment are represented by continuous curves in Fig. 1. The agreement between the theory and experiment is good in the range of validity of the theory ($\psi < 1$). For $\psi \gtrsim 1$ (which corresponds to $\Phi^{\rm nl} \gtrsim 36\pi$), we can only speak of qualitative agreement (a certain role may be played here also by the finite transverse dimensions of the laser beam, by the influence of thermal effects, by the difference between the material parameters of our NLC sample and the parameters given in the published literature).

It should also be pointed out that the threshold nature of the reorientational process is manifested more strongly (steeper dependence) for similar values of $I_{1,2}$ (Fig. 1c), as is clear from the above discussion.

3. TIME-DEPENDENT INSTABILITIES IN EXCITATION OF THRESHOLD REORIENTATION OF A NEMATIC LIQUID CRYSTAL IN THE FIELD OF A CIRCULARLY (ELLIPTICALLY) POLARIZED WAVE

1. Basic equations. We shall now analyze theoretically the process by which oscillations of the polarization as a function of time and of the aberration pattern of self-focusing of light-induced NLC reorientation are excited (we shall assume that a circularly or elliptically polarized wave is incident normally on an NLC with the homeotropic orientation). Earlier we discussed only the oscillations of the polarization in the first case[5]; the theory of oscillations of the aberration pattern for such polarizations of the incident light has not been considered at all.

The calculation procedure is again based on an approach developed in Ref. 10. We shall not go into details, but write down directly the principal equations which reduce, firstly, to transport equations relating the orthogonal components of a light field inside an NLC and, secondly, to the equations of motion of the director in an optical field. In the former case they are

$$\frac{\partial B_1}{\partial z} = \left(\frac{\varepsilon_{\parallel}}{\varepsilon_{\perp}}\right)^{3/4}\left(1 - \frac{\varepsilon_a}{\varepsilon_{\parallel}}\sin^2\psi\right)^{1/4} e^{-i\omega\Delta g/c} A_1 \frac{\partial \varphi}{\partial z}, \qquad (10)$$

$$\frac{\partial A_1}{\partial z} = -\left(\frac{\varepsilon_{\parallel}}{\varepsilon_{\perp}}\right)^{-3/4}\left(1 - \frac{\varepsilon_a}{\varepsilon_{\parallel}}\sin^2\psi\right)^{-1/4} e^{i\omega\Delta g/c} B_1 \frac{\partial \varphi}{\partial z}. \qquad (11)$$

Here,

$$B_1 = B\left(1 - \frac{\varepsilon_a}{\varepsilon_{\parallel}}\sin^2\psi\right)^{1/4}\left(\frac{\varepsilon_{\parallel}}{\varepsilon_{\perp}}\right)^{1/4}$$

and $A_1 = A$ are the amplitudes of the waves with the extraordinary (e) and ordinary (o) polarizations inside a medium [if $\mathbf{E}$ is the amplitude of the incident field along a selected direction, then $\mathbf{E}_e = (B;\ 0;\ B(\varepsilon_{\perp}/\varepsilon_{\parallel}) \times \tan\psi)$, and $\mathbf{E}_o = (0;\ A;\ 0)$];

$$\Delta g = \int_0^z \varepsilon_{\perp}\left(1 - \frac{\varepsilon_a}{\varepsilon_{\parallel}}\sin^2\psi\right)^{-1/2} dz - \varepsilon_{\perp}^{1/2} z$$

represents a change in the difference between the eikonals of the waves with the orthogonal polarizations inside the medium.

In the second case, when we are dealing with optical excitation of nonadiabatic (in the transverse direction of a sample) distortions of the NLC structure giving rise to a three-dimensional (along z) gratings, we obtain

$$K\frac{\partial^2\psi}{\partial z^2} - K\sin\psi\cos\psi\left(\frac{\partial\varphi}{\partial z}\right)^2 + \frac{\varepsilon_a\varepsilon_{\perp}}{8\pi\varepsilon_{\parallel}}|B|^2 \operatorname{tg}\psi = \gamma\frac{\partial\psi}{\partial t}, \qquad (12)$$

$$K\frac{\partial}{\partial z}\left(\frac{\partial\varphi}{\partial z}\sin^2\psi\right) + \frac{\varepsilon_a\varepsilon_{\perp}}{16\pi\varepsilon_{\parallel}}\frac{\sin^2\psi}{\cos\psi}(A^*Be^{i\omega\Delta g/c} + AB^*e^{-i\omega\Delta g/c}) = \gamma\sin^2\psi\frac{\partial\varphi}{\partial t}. \qquad (13)$$

As before, the angle ψ describes the deviation of $\mathbf{n}$ from the initial homogeneous orientation of the sample ($\mathbf{n}_0 \| z$) and this angle is associated with oscillations of the aberration pattern. The angle φ describes azimuthal rotation of $\mathbf{n}$ in the $xy \perp z$ plane, which gives rise to oscillations of the polarization[3]; K is the elastic constant (for the sake of simplicity we shall ignore the anisotropy of this constant and assume that $K_{11} \equiv K_{33} \equiv K$). In the case under discussion when the incident light is circularly polarized, $\mathbf{E}' = \mathbf{E}(1 + i)$, we have the following boundary condition (at $z = 0$):

$$B \equiv B_b = E, \qquad A \equiv A_b = iE. \qquad (14)$$

The solution of Eqs. (10) and (11) was found iteratively in the approximation $\partial\varphi/\partial z \ll 1/d$. We shall assume that $B_1 = B_{10} + B_{11}$ and $A_1 = A_{10} + A_{11}$, where B_{11} and A_{11} are small quantities of the same order as $\partial\varphi/\partial z$; then, using Eq. (14), we obtain

$$B_{10} = E(\varepsilon_{\parallel}/\varepsilon_{\perp})^{1/4}, \qquad A_{10} = iE. \qquad (15)$$

Next, assuming that $\psi \ll 1$ and using Eq. (15), we find the solution of Eqs. (10) and (11):

$$B_1 = \left(\frac{\varepsilon_{\parallel}}{\varepsilon}\right)^{1/4}\left\{E + iE\int_0^z \frac{\partial\varphi}{\partial z}\left(1 - \frac{\varepsilon_a}{4\varepsilon_{\parallel}}\sin^2\psi\right) e^{-i\omega\Delta g/c} dz\right\},$$

$$A_1 = iE - E\int_0^z \frac{\partial\varphi}{\partial z}\left(1 - \frac{\varepsilon_a}{4\varepsilon_{\parallel}}\sin^2\psi\right) e^{i\omega\Delta g/c} dz. \qquad (16)$$

It follows from Eq. (16) that energy is transferred directly between the orthogonal components of light inside the nonlinear medium; this transfer is a consequence of the condition $\partial\varphi/\partial z \neq 0$. This condition is ignored in Ref. 5, and the theory given there cannot account for oscillations of the aberration pattern [see Eq. (24) below].

Using Eq. (16), we can rewrite Eqs. (12) and (13) to first order in ψ using the following relationships:

$$K\frac{\partial^2\psi}{\partial z^2} - K\psi\left(\frac{\partial\varphi}{\partial z}\right)^2 + \frac{\varepsilon_a\varepsilon_\perp}{8\pi\varepsilon_\parallel}\psi|E|^2 \cdot\left\{1+2\int_0^z \frac{\partial\varphi}{\partial z}\sin\left(\frac{\omega}{c}\Delta g\right)dz\right\} = \gamma\frac{\partial\psi}{\partial t},$$

$$K\frac{\partial}{\partial z}\left(\frac{\partial\varphi}{\partial z}\psi^2\right) + \frac{\varepsilon_a\varepsilon_\perp}{8\pi\varepsilon_\parallel}\psi^2|E|^2\sin\left(\frac{\omega}{c}\Delta g\right) = \gamma\psi^2\frac{\partial\varphi}{\partial t}. \qquad (17)$$

2. Oscillations of the polarization. We shall first consider the second equation of the system (17). In this equation we shall ignore the first term (because it is a small quantity in the next order), which gives

$$\varphi(z,t) - \varphi(z,t_0) = \frac{\varepsilon_a\varepsilon_\perp}{8\pi\varepsilon_\parallel}|E|^2\int_{t_0}^{t}\sin\left(\frac{\omega}{c}\Delta g\right)dt, \qquad (18)$$

where the time t_0 corresponds to $\psi = 0$.

In the approximation $\psi = \text{const}$, i.e., also $\Delta g = \text{const}$, averaging over z yields the time dependence of φ:

$$\varphi(t) - \varphi(t=0) = \left\{\frac{1}{d}\int_0^d \frac{\varepsilon_a\varepsilon_\perp}{8\pi\varepsilon_\parallel\gamma}|E|^2\sin\left(\frac{\omega}{c}\Delta g\right)dz\right\}(t-t_0), \qquad (19)$$

where

$$\varphi(t) = \frac{1}{d}\int_0^z \varphi(t,z)\,dz, \qquad \varphi(t=0) = \frac{1}{d}\int_0^z \varphi(t=0,z)\,dz.$$

The relationship (19) can be written in the form

$$\Delta\varphi = \varphi(t) - \varphi(t=0) = \Omega(t-t_0). \qquad (20)$$

Therefore, we can expect the director to process in time in the azimuthal plane with an angular rotation frequency

$$\Omega = \frac{\varepsilon_a\varepsilon_\perp}{8\pi\varepsilon_\parallel}\frac{|E|^2}{d\gamma}\int_0^d \sin\left(\frac{\omega}{c}\Delta g\right)dz. \qquad (21)$$

This gives rise to rotation of the plane of the elliptic polarization of the transmitted light.

When an allowance is made for the time dependence of ψ, $\psi = \psi(t)$, we find that Eq. (18) yields [on condition that $\partial\varphi(z,t=0)/\partial z = 0$]

$$\frac{\partial\varphi}{\partial z} = \frac{\varepsilon_a^2\varepsilon_\perp^{1/2}}{8\pi\varepsilon_\parallel^2\gamma}|E|^2\frac{\omega}{c}\int_{t_0}^{t}\psi^2\cos\left(\frac{\omega}{c}\Delta g\right)dt, \qquad (22)$$

which we shall use later. It corresponds to the condition of continuity of φ at the boundaries of our sample. In fact, the boundary conditions are specified only for ψ: $\psi(z=0) = \psi(z=d) = 0$; they leave free the conditions for the director in the azimuthal plane, so that we shall assume that

$$\frac{\partial\varphi}{\partial z}(z=0,t) = \frac{\partial\varphi}{\partial z}(z=d,t) = 0.$$

3. Oscillations of the aberration pattern. We now turn to the first equation in the system (17). We solve it by means of perturbation theory using a small parameter $\partial\varphi/\partial z$. In the zeroth approximation ($\partial\varphi/\partial z = 0$) it is readily [solved subject to the condition given by Eq. (2)]. We then obtain

$$\psi_m = \text{const}\,e^{\Gamma t}, \quad \Gamma = \frac{1}{\gamma}\left[\frac{\varepsilon_a\varepsilon_\perp}{8\pi\varepsilon_\parallel}|E|^2 - \left(\frac{\pi}{d}\right)^2 K\right], \qquad (23)$$

where $\text{const} \equiv \psi_0$ is governed by the thermal rms fluctuations of the reorientation angle. The condition $\Gamma \geqslant 0$ governs the threshold (I_{th}) of the light-induced reorientation process considered in this approximation.

Substituting the solution of Eq. (23) into the first equation in the system (17), we can readily obtain the solution in the next approximation:

$$\psi_m = \text{const}\cdot\exp[\Gamma(t-t_0) - \alpha + \beta], \qquad (24)$$

where

$$\alpha = \frac{2K}{\gamma d}\int_{t_0}^{t}\int_0^d\left(\frac{\partial\varphi}{\partial z}\right)^2\sin^2\frac{\pi z}{d}\,dz\,dt$$

$$= \frac{\omega^2}{c^2}\frac{2K}{\gamma d}\left(\frac{\varepsilon_a^2\varepsilon_\perp^{1/2}}{8\pi\varepsilon_\parallel^2\gamma}|E|^2\right)^2 \cdot\int_{t_0}^{t}dt\int_0^d\left[\int_{t_0}^{t}\psi^2\cos\left(\frac{\omega}{c}\Delta g\right)dt\right]^2\sin^2\frac{\pi z}{d}\,dz,$$

$$\beta = \frac{\varepsilon_a\varepsilon_\perp|E|^2}{2\pi\varepsilon_\parallel\gamma d}\int_{t_0}^{t}dt\int_0^d\left[\sin^2\frac{\pi z}{d}\int_0^z\frac{\partial\varphi}{\partial z}\sin\left(\frac{\omega}{c}\Delta g\right)dz\right]dz$$

$$= \frac{\varepsilon_a^3\varepsilon_\perp^{\frac{3}{2}}}{16\pi^2\varepsilon_\parallel^3\gamma^2 d}\frac{\omega}{c}|E|^4 \cdot\int_0^d\left\{\sin^2\frac{\pi z}{d}\int_{t_0}^{t}dt\int_0^z\sin\left(\frac{\omega}{c}\Delta g\right)\left[\int_{t_0}^{t}\psi^2\cos\left(\frac{\omega}{c}\Delta g\right)dt\right]dz\right\}dz.$$

Here we have used Eq. (22).

We shall now consider in greater detail the solution (24). Various regimes appear depending on the values of the parameters Γ, α, and β. Let us assume that at the initial time $t = t_0$, when $\psi = \psi_0 \sim 0$, we have $\Gamma \geqslant 0$, i.e., $I > I_{\text{th}}$. In this case we find $\alpha = \beta = 0$. As t is increased ($t > t_0$), the parameters α and β begin to increase and in the case of α the process is faster (the time integral in the square brackets occurs in the second power in the expression for α, whereas in the expression for β it is in the first power). We can therefore assume that $\beta = 0$ and then Eq. (24) readily shows that there is always time t_1 when α becomes equal to Γ [$\alpha = \Gamma(t - t_0)$ at $t = t_1$] and for $t > t_1$, the angle ψ_m begins to decrease to its fluctuation value $\psi_0 \sim 0$ (or some other value). The process is then repeated.

This gives rise to oscillations of the reorientation angle ψ with time, i.e., oscillations of the aberration pattern. It is important to stress that these oscillations are related to the condition $\partial\varphi/\partial z \neq 0$, i.e., the rotation of the polarization of the light across the thickness of a sample is nonuniform.

4. Hysteresis: discussion. This solution also gives rise to hysteresis of the dependence $\psi_m(I)$. In fact, for $I > I_{th}$, when the reorientation is excited in an NLC ($\psi_m > 0$), we now begin to reduce I. When $\Gamma < 0$ we find that α approaches zero faster than does β. We can therefore assume that $\beta > \alpha$ applies in this range; then, as long as $\Gamma(t - t_0) + \beta \geqslant 0$, the angle ψ_m does not tend to zero and the system is excited. The system returns to its initial state ($\psi_m \equiv \psi_0 \sim 0$) at a different threshold $I'_{th} < I_{th}$, i.e., hysteresis appears. It should be stressed that in this case ($\Gamma < 0$) the reorientation is weak and the aberration rings are no longer observed, so that we can observe experimentally only the oscillations of the polarization of light.

Our analysis therefore accounts for all the experimental results reported in Ref. 5 (oscillations of the polarization and of the aberration pattern, the appearance of hysteresis).

We shall now estimate the characteristic frequency Ω of the oscillations we have found for the experimental conditions of Ref. 5. In the case of the nematic liquid crystal 5CB used in Ref. 5 ($d = 65\,\mu$m, $\lambda = 0.5\,\mu$m, which gives $I_{th} \approx 2.1$ kW/cm^2), we have $\Omega = 0.0034/\psi_m^2$ Hz. This value of Ω satisfies the results of Ref. 5 ($2\pi/\Omega \approx 40$ s) when $\psi_m \approx 0.14$. This value of ψ_m corresponds approximately to the number of the aberration rings, which is of the order of or even less than one, in agreement with the experimental observations showing that oscillations of the polarization are observed only near the threshold when the nonlinear phase advance is $\Phi^{nl} < 2\pi$ (and the aberration rings are no longer visible). This does not violate the condition of validity of our theory and, in particular, we find from Eq. (22) that the inequality $(\partial\varphi/\partial z)d \sim 0.01 \ll 1$ is indeed satisfied and our approximation based on an expansion in terms of this small parameter is correct. When the excess above the threshold is large, so that the aberration rings are observed, the pulsations of these rings become dominant (and oscillations of the polarization are then difficult to observe experimentally against this background).

We shall conclude by noting that a similar analysis can be carried out also in the case of elliptic polarization of the incident light ($\mathbf{E} = \mathbf{E}_1 + i\mathbf{E}_2, \mathbf{E}_1 \neq \mathbf{E}_2$). Then, instead of the relationships given by Eq. (14), we have to write down $B_b = E_1$ and $A_b = iE_2$. Since inside an NLC the propagating light has the elliptic polarization in either case, all the results should remain qualitatively valid. Oscillations in the case of elliptic polarization of the incident light are more pronounced (they become undamped) when the values of $\mathbf{E}_1$ and $\mathbf{E}_2$ are close, which is in agreement with the experimental results.[6]

4. CONCLUSIONS

These results demonstrate that the main reason for the appearance of time-dependent instabilities is a nonlinear interaction which occurs inside a medium between waves with different (orthogonal) polarizations. We are in fact dealing with the nonlinear dynamics of a system with a small number of degrees of freedom (these degrees of freedom are the two components of the polarization of light) and a transition of the system to a chaotic state. The classical problem for which processes of this kind have been thoroughly investigated is that of the dynamics of a nonlinear (anharmonic) oscillator. The threshold orientational effects in liquid crystals make it possible to represent the equations in the form of spatial or temporal analogs of the equation for an anharmonic oscillator, although even then there are some difficulties.[1]

When two orthogonal components of the polarization of light propagate inside an NLC, the situation is analogous to that of oscillations of two coupled oscillators[6]; the coupling between the polarization components is due to the nonlinearity of the medium. Two aspects are important. Firstly, the periodic exchange of energy between the components (dependent on the strength of the coupling) is of a competitive nature, i.e., we can say that these components make opposite contributions to the nonlinearity of the medium. Secondly, a system of this kind can be reduced to two oscillators with different natural oscillation frequencies (spatial periods of modulated structures); this gives rise to different relaxation times of the nonlinear response for the two components of the field. Therefore, the conditions are satisfied for the appearance of regenerative pulsations in the system.[14] In a system with two coupled oscillators we can of course expect stochastic regimes; in optics this leads to polarization chaos.[15]

A remarkable feature of liquid crystals is that the time τ_f which characterizes the feedback effects is related to a nonlocal response of the medium to an external field and is governed by relaxation times (we are speaking here of the internal feedback in the absence of mirrors—see Ref. 1). The response time can be fairly long (it can vary from 10^{-5} s to several seconds), so that in purely optical systems we have conditions favoring the appearance of Ikeda instabilities for which τ_f must be greater than the relaxation time) discussed in Ref. 16.[4)] Experimental demonstration of these instabilities is a matter for the near future.

[1)]For example, in the case of magnetic systems subjected to two mutually orthogonal fields, the magnetic field experienced by the anisotropic magnetic center is directed not in the direction of the magnetic fields but in the direction of the maximum polarizability of the center which experiences the fields as if they were mutually parallel.

[2)]The initial state ($\psi_{in} \gtrless 0$) is in this case governed by the random thermal fluctuations of the director; there are three possible states of the system: $\pm\psi_m, 0$.

[3)]They are governed essentially by the well-known effects of the appearance of a rotational momentum in a body illuminated with elliptically polarized light (Sadovskii effect), which leads to precession.[12]

[4)]In the case of systems with resonators, τ_f is the round-trip time (the time taken to travel there and back across the resonator), which is usually less than 10^{-8} s.

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