

EFFECT OF SPATIAL DISPERSION OF NONLINEARITY  
ON SELF-FOCUSING OF LASER RADIATION IN LIQUID  
CRYSTALS: THEORY AND NUMERICAL EXPERIMENTS

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UDC 530.145:530.18

Analytic calculations (in the aberrationless approximation) are made for the threshold conditions and changes in width of a light beam with self-focusing (SF) in the isotropic phase of a liquid crystal near the phase transition point. It is shown that the orientational and thermal mechanisms of spatial dispersion (SD) of the nonlinearity ensure the possibility of quasiwaveguide propagation of a powerful beam in the medium. The method of finite elements is used to develop a numerical solution program for the SF equations in a medium with SD of nonlinearity, and the characteristics of light beams propagating in the medium are investigated over a wide range of parameters.

§1. For a variety of reasons, there is considerable interest in investigating self-focusing (SF) of light in substances which, in a certain range of thermodynamic parameters, have a liquid-crystal structure characterized by macroscopic ordering of the molecular structure.

1) In many liquid crystals (LC), the nonlinearity greatly exceeds that of ordinary liquids in magnitude; the same is true of the relaxation time  $\tau_T$  of LC nonlinearity, the increase in  $\tau_T$  being particularly prominent as the temperature approaches the transition point  $T^*$  "isotropic liquid/nematic LC" [1-4]. † The main consequences of this are as follows: low threshold of SF [1-4], stability of SF relative to separation of intense beam into filaments (something that has been recorded experimentally [1] and has a theoretical explanation based on allowance for spatial dispersion (SD) of the nonlinearity [5, 6]). The importance of these features in applications is unquestionable. The sensitivity of  $\tau_T$  to temperature changes can also be effectively used in research for the purpose of studying generation techniques for short pulses and the general properties of nonstationary self-action [7].

2) The influence of the structural properties of LC and of powerful radiation is reciprocal; the latter can be used to effect controlled rearrangement of the structure of the medium [6, 8, 9] (see also [10]). This makes nonlinear optical effects — in particular, SF effects — into indicators of thermodynamic changes in the LC phase near the phase transition point. Statistical methods of nonlinear optics also provide additional possibilities here (see [11]).

3) The pronounced nature, diversity, and controllability of nonlinear self-action in LC, on the one hand, and the very real possibility of setting up the appropriate experiments over a wide range of parameters, on the other, make these effects convenient for solving an extensive set of problems in the theory of nonlinear waves.

The aim of this paper is to provide, first of all, an analytic calculation (in the aberrationless approximation [12]) of the threshold conditions for SF and of changes in intensity and width of the light beam in the isotropic phase of LC near the phase transition point; second, a numerical analysis of SF effects, in the course of which we find the regions of the radiation and medium parameters that define the qualitative behavior of SF (saturation effects, quasiwaveguide modes, multiple-focusing structure). Particular emphasis is laid on thermal and orientational mechanisms of SD of nonlinearity.

§2. The initial equations for analyzing SF of radiation in the isotropic phase of LC are the equations

† Self-action effects of light in cholesteric LC have their own specific features, of course, and should be examined separately.

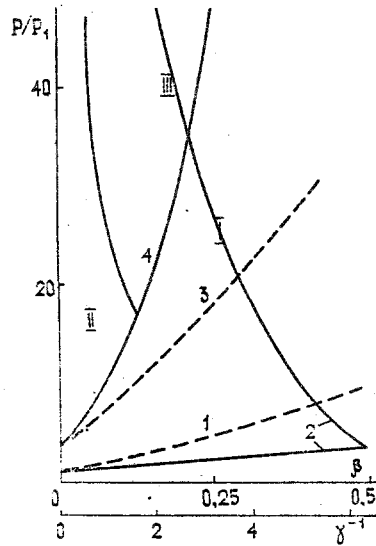


Fig. 1

that link the slow complex amplitude  $E$  of the linearly polarized electromagnetic wave, the temperature  $T$ , and the order parameter  $Q$  that characterizes the degree of orientation ordering of the molecules.

In the first place, we have the parabolic equation of nonlinear diffraction [12, 2], whose nonlinearity results from the fact that  $Q$  depends on the field:

$$\left( \frac{\partial}{\partial z} + \frac{i}{2k} \Delta_{\perp} + \frac{1}{v} \frac{\partial}{\partial t} + \delta \right) E = - \frac{4\pi ik}{3n^2} \chi_a Q E. \quad (1)$$

Here  $z$  is the propagation coordinate;  $k$  is the wave number;  $\Delta_{\perp} = \partial^2/\partial x^2 + \partial^2/\partial y^2$ ;  $v$  is the group velocity; and  $\chi_a$  is the anisotropy of linear susceptibility. We ignore the fact that the absorption coefficient  $\delta$  depends on the temperature and the radiation intensity; according to estimates, it becomes important only in the non-transparency region of LC ( $T < T^*$ ).

The Landau expansion of the free nematic energy as  $T \rightarrow T^*$ , taking account of the effect of the field, has the form [13]†

$$F = F_0 + \frac{a}{2} (T - T^*) Q^2 + \frac{b}{2} (\text{grad } Q)^2 - \frac{2}{9} \chi_a Q |E|^2. \quad (2)$$

Here  $F_0$  is the energy of the isotropic phase ( $T \gg T^*$ );  $a$  is a constant that is independent of temperature. The constant  $b$ , which depends weakly on temperature, is related to the characteristic correlation scale  $L_C$  for the LC as follows:  $b = a(T - T^*)L_C^2/4$ ; the third term in (2) describes the SD of the order parameter. The relaxation equation can be obtained by variation of (2) with respect to  $Q$ :

$$\eta \partial Q / \partial t + a(T - T^*)Q = b \Delta_{\perp} Q + \frac{2}{9} \chi_a |E|^2, \quad (3)$$

where  $\eta$  is the viscosity coefficient. Near  $T^*$ , the relaxation time of the parameter, of order  $\tau_r = \eta/a(T - T^*)$ , varies strongly with temperature (whereas  $\eta$  is weakly dependent on  $T$  [3]), and therefore the effect of laser heating may become decisive.

The change in temperature of the medium in a radiation field is described by the heat equation

$$C \rho \frac{\partial T}{\partial t} = \kappa \Delta_{\perp} T + \frac{\delta c n |E|^2}{8\pi}, \quad (4)$$

where  $\kappa$  is the coefficient of thermal conductivity. This equation takes account of a second (thermal) mechanism of spatial nonlocalness of the nonlinear response. According to estimates, the temperature dependence of the density  $\rho$  of the medium and of the heat capacity  $C$  in the range of interest to us has little effect on the solution of system (1), (3), (4).

† Generally speaking, the order parameter is a tensor, but SF effects for a linearly polarized wave depend only on one scalar combination of the components of this tensor.

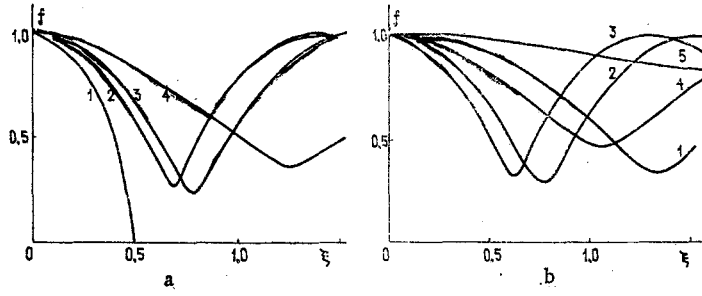


Fig. 2

To be specific, we will assume that a Gaussian beam is incident on the boundary of the medium ( $z = 0$ ),

$$E(r, z = 0) = E_0 \exp\left(-\frac{r^2}{2r_0^2}\right) \quad (r^2 = x^2 + y^2), \quad (4')$$

and let us consider the steady-state mode of self-focusing which occurs if the duration of the incident pulse is greater than  $\tau_T$  and the settling time of the temperature distribution  $\tau_T = C \rho r_0^2 / 4\kappa$  (the qualitative features of SF that manifest themselves under steady-state conditions are also maintained under unsteady-state ones; a detailed investigation of nonstationary SF in LC will be taken up in a subsequent paper).

§3. Following the familiar procedure of the aberrationless approximation [12], we can derive from system (1), (3), (4) under steady-state conditions ( $\partial/\partial t = 0$ ) equations that describe the change in the dimensionless beam width with the propagation coordinate (under the assumption that the Gaussian shape is maintained [12], the intensity on the beam axis is inversely proportional to the square of the width  $f$ ). If we allow only for the orientational mechanism of SD ( $\delta = 0$ ), we have

$$f^3 \frac{d^2 f}{d\xi^2} = 1 - \alpha \gamma f^2 [1 + \gamma f^2 \text{Ei}(-\gamma f^2) \exp(\gamma f^2)]. \quad (5)$$

Here  $\xi = z/L_d$  (the diffraction length  $L_d = \kappa r_0^2$ );  $\text{Ei}(x)$  is an integral exponential function; the parameter  $\alpha = P/P_1$  is the ratio of the beam power  $P = c n E_0^2 r_0^2 / 8$  to the threshold SF power without allowance for SD;  $P_1 = 27\pi c n^3 a (T - T^*) / 2(8\pi \kappa \chi_a)^2$ ; the SD parameter  $\gamma = (r_0/L_c)^2$ .

For a purely thermal mechanism ( $L_c = 0$ )†

$$f^3 \frac{d^2 f}{d\xi^2} = 1 - \alpha \left(1 - \frac{0.1 \alpha \beta}{f^2}\right) \left(1 + \frac{0.4 \alpha \beta}{f^2}\right)^{-2}, \quad (5')$$

where  $\beta = P_1/P_2$ ,  $P_2 = 2\pi \kappa (T_0 - T^*)/\delta$ ,  $T_0$  is the temperature of the thermostat.

It follows from (5) and (5') that SF of the laser beam occurs if its power exceeds the threshold value  $P > P_{th}$  ( $\alpha > \alpha_{th}$ ), this being given by the following expressions, respectively:

$$\alpha_{th} = [\gamma (\gamma \exp(\gamma) \text{Ei}(-\gamma) + 1)]^{-1}; \quad (6)$$

$$1 = \alpha_{th} (1 - 0.1 \alpha_{th} \beta) (1 + 0.4 \alpha_{th} \beta)^{-2}. \quad (6')$$

In the absence of SD resulting from the nonlocal nature of the oriented response ( $L_c = 0$ ) and of absorption ( $\delta = 0$ ), expressions (6) and (6') become the ordinary condition  $P_{th} > P_1$  [12].

Figure 1 shows expressions (6) and (6'), representing the threshold power as a function of the radiation parameters and the parameters of the medium. Curve 1 represents  $\alpha_{th}(\gamma)$ , while curve 2 represents  $\alpha_{th}(\beta)$ . As can be seen from the figures, SD effects make the SF conditions for a beam with specified parameters more rigid; the presence of the upper branch on curve 2 indicates that, in the aberrationless approximation, SF does not occur if the beam power exceeds some value (fairly large on an actual scale). This is to be explained by the fact that the thermal mechanism saturates the nonlinearity, the saturation being nonlocal. The self-action picture can be investigated in greater detail through an exact numerical analysis of steady-state conditions (1), (3), (4) (see §4).

We can integrate Eqs. (5) and (5') once:

† The variation in intensity as a result of absorption in the isotropic LC phase can be ignored:  $\delta L \ll 1$ ,  $L$  is the length of the LC.

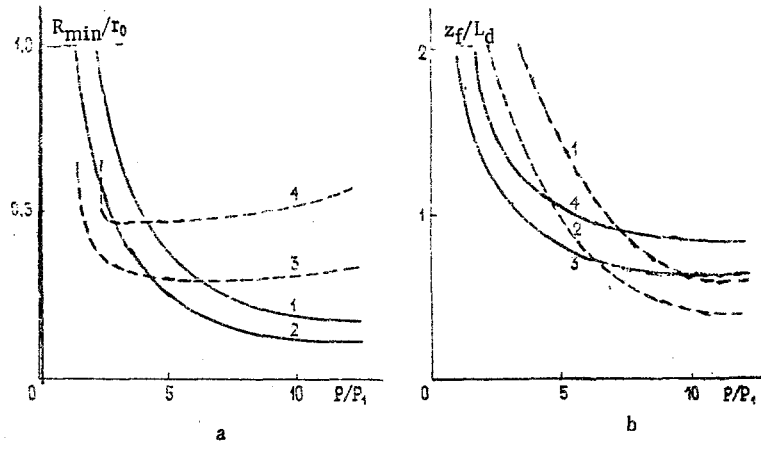


Fig. 3

$$\left(\frac{df}{d\xi}\right)^2 = 1 - \frac{1}{f^2} + \alpha\gamma [\text{Ei}(-\gamma) \exp(\gamma) - \text{Ei}(-\gamma f^2) \exp(\gamma f^2)]; \quad (7)$$

$$\left(\frac{df}{d\xi}\right)^2 = 1 - \frac{1}{f^2} - \frac{5\alpha}{4} \left(1 - \frac{1}{f^2}\right) / \left(1 + \frac{2\alpha\beta}{5}\right) \left(1 + \frac{2\alpha\beta}{5f^2}\right) + \frac{5}{8\beta} \ln \left( \frac{1 + \frac{2}{5}\alpha\beta}{1 + \frac{2\alpha\beta}{5f^2}} \right). \quad (7')$$

The solutions of (7) are shown in Fig. 2a. As  $\gamma \rightarrow \infty$  (ordinary Kerr mechanism without saturation) the beam collapses to a point (curve 1). Spatial dispersion of the nonlinearity causes the beam width to oscillate with respect to  $z$ , the period of the oscillations increasing as  $\gamma^{-1}$  increases (curves 1-4 in Fig. 2a are plotted for  $\alpha = 5$  and  $\gamma$  equal to  $\infty$ , 10.1, 5.3, and 5.1, respectively).

The thermal mechanism has a similar effect on the change in  $f$  (Fig. 2b); SF becomes softer and is also characterized by a multifocal structure. Curves 1-3 are plotted for  $\beta = 0.1$  and  $\alpha = 2; 5; 10$ ; curves 4 and 5 are for  $\alpha = 5$  and  $\beta = 0.2; 0.4$ . The minimum cross section  $R_{\min}^2$  of the beam is given by the following equation, allowing only for SD of Q:

$$\frac{r_0^2}{R_{\min}^2} = \alpha\gamma \left[ \text{Ei}(-\gamma) \exp(\gamma) - \text{Ei}\left(\frac{-\gamma R_{\min}^2}{r_0^2}\right) \exp\left(\frac{\gamma R_{\min}^2}{r_0^2}\right) \right] + 1, \quad (8)$$

and, when only the thermal mechanism is allowed for, by the equation

$$\frac{r_0^2}{R_{\min}^2} = 1 - \frac{5\alpha}{4} \left(1 - \frac{r_0^2}{R_{\min}^2}\right) / \left(1 + \frac{2}{5}\alpha\beta\right) \left(1 + \frac{2\alpha\beta r_0^2}{5R_{\min}^2}\right) + \frac{5}{8\beta} \ln \left( \frac{1 + \frac{2}{5}\alpha\beta}{1 + \frac{2\alpha\beta r_0^2}{5R_{\min}^2}} \right). \quad (8')$$

Figure 3 shows  $R_{\min}$  (Fig. 3a) and the oscillation period  $z_f$  of the beam width (Fig. 3b) as a function of beam power (curves 1 and 2 are for  $\gamma = 1$  and 2; curves 3 and 4 are for  $\beta = 0.1$  and 0.2). The beat depth for  $f$  increases with the power; the effect of SD of Q is manifest in the attempt to stabilize the amplitude of these beats (quasiwaveguide propagation);  $R_{\min}$  is a nonmonotonic function of power when the thermal mechanism is allowed for; when the Kerr threshold is slightly exceeded ( $\alpha = 1$ ) the heating of the medium does not yield any marked contribution to stabilization of  $f$ ; for large power values ( $\alpha \geq 8$ ) the nonlocal nature of the heating hinders the contraction of the beam and the beat depth of  $f$  decreases.

Thus, competition between the Kerr mechanism of SF and the mechanisms of SD of the nonlinearity can account for the experimentally recorded stabilization of  $R_{\min}$  [7] under self-action in LC.

§4. The aberrationless approximation makes possible a qualitative treatment of the process of SF and yields estimates for the critical parameter values. Successive solution of (1), (3), (4) with arbitrary boundary conditions can be attained only by means of advanced numerical methods such as the finite-difference method (for applications to SF, see, e.g., [14]) or the method of finite elements, used in this study and capable of attaining a specified accuracy with less computation (described in detail in [15]; regarding applications to nonlinear diffraction problems, see [16]).

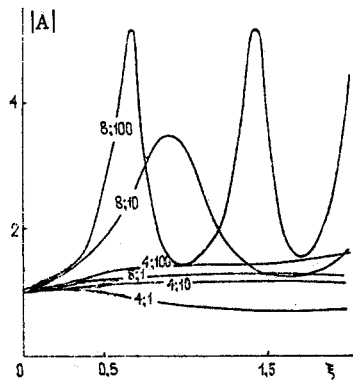


Fig. 4

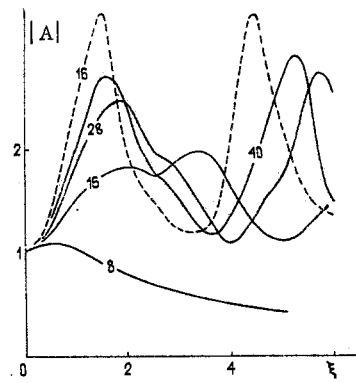


Fig. 5

Let us now analyze our numerical results; first we will consider the effect on SF of SD of the order parameter. Figure 4 shows the normalized modulus of the field amplitude  $|A|$  on the beam axis as a function of crystal thickness  $\xi$  for different  $\alpha$  and  $\gamma$  values; these values are shown in the gaps of the curves [with (4') being used as a boundary condition]. Three of the curves were obtained for  $\alpha = 8$ , this corresponding to a power that exceeds the threshold of Kerr SF by a factor of 2 (as we know [17], the aberrationless approximation decreases the SF threshold by a factor of 4). Curves 3 and 4 in Fig. 1 show  $\alpha_{th}(\gamma)$  and  $\alpha_{th}(\beta)$  as obtained for numerical solution of the initial equations; the solution method described takes account of the aberrations. If the LC correlational scale  $L_C$  is considerably less than  $r_0$  ( $\gamma = 100$ ), a multifocus structure arises. With increasing  $L_C$  ( $\gamma = 10$ ) the distance between foci increases, while the maximum amplitude value simultaneously decreases. For  $\gamma = 1$  we encounter conditions that recall stationary waveguide propagation conditions. The remaining curves were obtained near the threshold of Kerr SF ( $\alpha = 4$ ).

If the thermal mechanism predominates, the SF picture is determined by three processes: Kerr SF, linear diffraction, and defocusing from heating of the medium. In region I of the medium and radiation parameters (Fig. 1) it is diffraction that plays the major part; the field amplitude on the axis decreases monotonically with increasing  $\xi$ . Regions II and III are characterized by a multifocused structure; examples of such structures are given in Fig. 5 ( $\beta = 0.1$ ; solid curves, with the values of  $\alpha$  being indicated in the gaps in the curves). Near the boundary of regions I and II the multifocus structure of SF is of a complex nature ( $\alpha = 16$ ). A more detailed analysis of the numerical results shows that a prominent process here is that of radial redistribution of the amplitude over the cross section of the beam, with its shape deviating from the Gaussian. A characteristic of region III is the appearance of saturation; further increases in the input power have little effect on the position of the foci and the maximum amplitude value. Decreasing  $\beta$  has a very marked effect on the SF threshold (dashed curve in Fig. 5,  $\beta = 0.075$ ,  $\alpha = 16$ ).

§5. Spatial dispersion of the order parameter becomes more and more prominent as the temperature approaches the critical value; the principal term in (3) becomes the term with the transverse Laplacian. The coefficient  $\gamma^{-1} \sim L_C^2$  diverges critically at the transition point, and therefore detection and measurement of the parameters of the multifocus structure of SF in LC is a realistic way of investigating  $L_C$  — one of the most important characteristics of the phase transition.

The authors are grateful to S. A. Akhmanov for useful discussions.

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EFFECT OF NONRECIPROCAL ELEMENTS ON INTERACTION  
OF ELLIPTICALLY POLARIZED OPPOSING WAVES IN  
RING GAS LASERS

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UDC 621.373.826

Scalar generation equations are obtained for elliptically polarized opposing waves in ring gas lasers with anisotropic resonators when there are nonreciprocal elements. These equations are used as a basis for computing the difference in generation frequencies for the opposing waves in the case of practical importance in which their polarizations differ little from one another and are quasilinear. The formula is analyzed for one- and two-isotope composition of the active medium.

§1. Papers [1, 2] examined nonlinear interaction of elliptically polarized opposing waves in ring gas lasers with resonators having arbitrary polarization anisotropy and not containing nonreciprocal elements. It is known that in such resonators, in each of the opposing directions ( $n = r, l$ ), there are two ( $k = 1, 2$ ) eigenstates of polarization of the traveling wave with different eigenvalues. The frequencies and losses of opposing waves belonging to the same eigenstates are the same, while their polarizations are different. In accordance with [2], polarization eigenvectors of opposing waves belonging to different eigenvalues possess the property of quasiorthogonality (see (1.4) of [2]).

In this paper we will generalize the results of [1, 2] to the case in which there are arbitrary nonreciprocal elements in the resonator that create amplitude, frequency, and polarization nonreciprocity of the opposing waves. In this case the losses, frequencies, and polarizations of all four natural oscillations of the resonator are different and the quasiorthogonality conditions are violated. Scalar generation equations (2.14) and (2.15) of [2] are maintained in the same form, whereas the coefficients of nonlinear interaction of the elliptically polarized waves (2.16) and (2.17) are modified. The modified expressions for these coefficients may be found in the appendix.

We should note the following differences between coefficients (A.1)-(A.3) and the corresponding expressions in [2]:

- 1) Coefficient  $T_0$  from [2], which is independent of the propagation direction of the wave, is replaced by  $T_r$  from (A.3);
- 2) the expression for  $b_r$  is altered;
- 3) a new term proportional to  $a_n$  appears in the expression for  $B_{nn}$ .

If there are no nonreciprocal elements, these differences disappear and Eqs. (A.1)-(A.3) become the corresponding expressions of [2].

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Leningrad State University. Translated from Izvestiya Vysshikh Uchebnykh Zavedenii, Radiofizika, Vol. 22, No. 1, pp. 62-65, January, 1979. Original article submitted July 11, 1977.