Giant optical nonlinearity in the mesophase of a nematic liquid crystal (NCL)

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(Submitted 22 January 1980)

Pis'ma Zh. Eksp. Teor. Fiz. **31**, No. 5, 287–292 (5 March 1980)

A self-focusing, giant optical nonlinearity, which is caused by rotation of the NLC director due to the action of the light wave field, is predicted in the oriented mesophase of a nematic liquid crystal (NLC). The self-focusing effect of He-Ne laser radiation, which has a power of $\sim 10^{-2}$ W and a power density of $\sim 50\ \mathrm{W/cm^2}$, was detected experimentally in a 60-μm-thick, plane oriented NLC layer for oblique incidence of an extraordinary wave. The measured value of the effective nonlinearity constant $\epsilon_2 = 0.14\ \mathrm{cm^3/erg}$, which corresponds to the theoretical predictions, is $\approx 10^9$ times greater than the nonlinearity of carbon bisulfide.

PACS numbers: 42.65.Jx, 78.20.Ek

1. *Theory.* The equations for the director unit vector $\mathbf{n}(\mathbf{r},t) = \mathbf{n}^0 + \delta\mathbf{n}(\mathbf{r},t)$, disregarding the hydrodynamic motion of the medium and allowing for the orienting action of the light field $\mathbf{E}_{\text{sub}} = 0.5\,[\mathbf{E}(\mathbf{r})\exp(-i\omega t) + \mathbf{E}^*(\mathbf{r})\exp(+i\omega t)]$ in an approximation that is nonlinear with respect to $\delta\mathbf{n}$, have the form (see, for example, Refs. 1–4):

$$\gamma\frac{\partial \delta n_i}{\partial t} + K_{22}[\nabla_i(\vec{\nabla}\vec{\delta}\mathbf{n}) + (\mathbf{n}^\circ\vec{\nabla})^2\delta n_i - \Delta\delta n_i]$$
$$- K_{11}\nabla_i(\vec{\nabla}\cdot\delta\mathbf{n}) - K_{33}(\mathbf{n}^\circ\cdot\vec{\nabla})^2\delta n_i \tag{1}$$
$$+ (K_{11} - K_{22})n_i^\circ(\mathbf{n}^\circ\vec{\nabla})(\vec{\nabla}\delta\mathbf{n}) + \kappa_a H^2\delta n_i$$
$$= \frac{\epsilon_a}{16\pi}\left(\delta_{il}n_m^\circ + \delta_{im}n_l^\circ - 2n_i^\circ n_l^\circ n_m^\circ\right)E_l E_m^* \cdot$$

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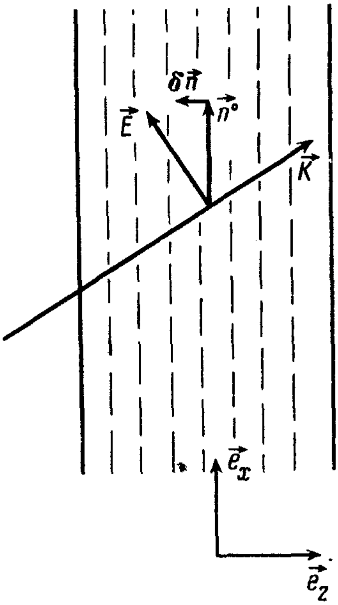


FIG. 1.

Here K_{11}, K_{22}, and K_{33} (ergs/cm) are the Franck constants, γ (poise) is the relaxation constant, $\kappa_\alpha = \kappa_\parallel - \kappa_\perp$ is the anisotropy of the magnetic susceptibility, $\mu = 1 + 4\pi\kappa$, and the intensity of the external field. We shall assume that the field $\mathbf{E}(\mathbf{r})$ is a quasi-plane wave with a polarization that is constant in space (i.e., either an *o* wave or an *e* wave). Let us choose an *x* axis along the director direction $\mathbf{e}_x \equiv \mathbf{n}^0$ (see Fig. 1), assuming that the NLC occupies the region $0 < z < L$. It is convenient to seek a solution of Eq. (1) in the expansion form

$$\delta \mathbf{n}(\mathbf{r}, t) = \sum_{m=1}^{\infty} \mathbf{A}_m(x, y, t) \sin \frac{\pi m z}{L} . \tag{2}$$

Thus, for a plane, light wave **E** of a certain polarization (*o* or *e* wave) it follows from Eq. (1) that

$$\mathbf{A}_m = [1 - \exp(-\Gamma_m t)]\mathbf{e}_z (\mathbf{E} \cdot \mathbf{e}_x) \frac{(\mathbf{E}^* \cdot \mathbf{e}_z)\epsilon_a}{\Gamma_m \gamma 4\pi^2} \frac{1}{m} [1 - (-1)^m] . \tag{3}$$

$$\Gamma_m = \gamma^{-1} [\kappa_a H^2 + K_{11} (\pi m / L)^2] .$$

Thus, for an *o* wave, i.e., for $\mathbf{E} \perp \mathbf{e}_x$, there is no nonlinearity. The coefficient $[1 - \exp(-\Gamma_m t)]$ characterizes the gradual establishment of orientation when the light wave suddenly appears at the time $t = 0$. The nonlinear advance of the phase $\delta\phi$ across the cell at an angle α between the *z* axis and the directional propagation is

$$\delta\phi = \frac{\omega}{c} L \frac{\epsilon_a}{\pi\sqrt{\epsilon}\cos\alpha} \frac{(\mathbf{E} \cdot \mathbf{e}_x)(\mathbf{E} \cdot \mathbf{e}_z)}{|E|^2} \sum_m |A_m| m^{-1} [1 - (-1)^m] . \tag{4}$$

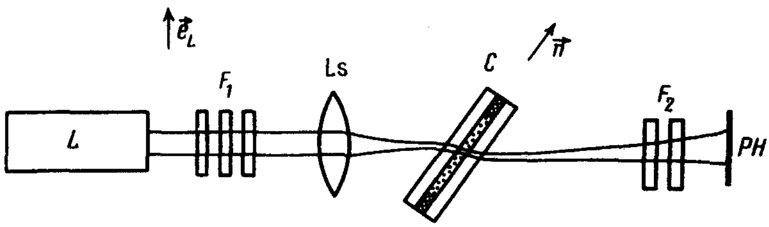


FIG. 2.

The term with $m = 1$ is the most important contribution. If the incident wave is not strictly plane, but has a smooth dependence

$$|\mathbf{E}(\mathbf{r}_\perp)|^2 = |\mathbf{E}_0|^2 (1 - 2|\mathbf{r}_\perp|^2/a^2 + \ldots)$$

on the coordinates across the beam, then Eq. (4) can be used for $\delta\phi(\mathbf{r}_\perp)$ by substituting in it the local value of the intensity $|\mathbf{E}(\mathbf{r}_\perp)|^2$. Self-focusing of the light occurs as a result (see Refs. 5–7), and the reciprocal of the focal length f^{-1} of the nonlinear lens is

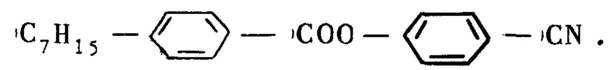
$$f^{-1} = \frac{4c\,\phi(r_\perp = 0)}{\omega a^2} = \frac{4 L \epsilon_a^2}{\pi^3 a^2} \frac{\cos a \sin^2 a}{\sqrt{\epsilon}\,\Gamma_1 \gamma} |E_0|^2 [1 - \exp(-\Gamma_1 t)], \tag{5}$$

where only the term with $m = 1$ is retained. This expression can be compared with the focal length of the nonlinear lens when the dielectric constant of the medium depends on the field in the manner $\epsilon = \epsilon_0 + 0.5\,\epsilon_2 |E|^2$ for the same geometry:

$$f^{-1} = \frac{\epsilon_2 |E_0|^2}{\sqrt{\epsilon_0}} \frac{L}{a^2 \cos a}. \tag{6}$$

2. *Experiment.* The following scheme was assembled to observe the light self-focusing effect in an NLC (Fig. 2).

The radiation of a LG-38 He-Ne laser (L), which lases in a low, transverse mode after passing through the filters F_1, was focused by the lens (*Ls*) with a focal length $f = 25$ cm on the thin glass cell (C) with a nematic liquid crystal (*NLC*). The angular divergence of the transmitted beam in the far zone was recorded by the lensless camera (PH) with a system of light filters F_2. The nematic was comprised of 60% of mixture "*A*" and 40% of the substance



The crystal parameters were as follows:

$$n_0 = 1.51,\ n_e = 1.71,\ K_{11} = 8.5 \cdot 10^{-7}\ \text{dyne}.$$

The crystal was at room temperature of 15–20°C. The 60-μm-thick cell was fabricated from 4-mm-thick glass with a Teflon spacer. The plane of the cell is perpendicular to the plane of the illustration; the electrical polarization vector $\mathbf{e}_L$ of the beam and the NLC director $\mathbf{n}$ are in the plane of the illustration (the polarization was imparted by

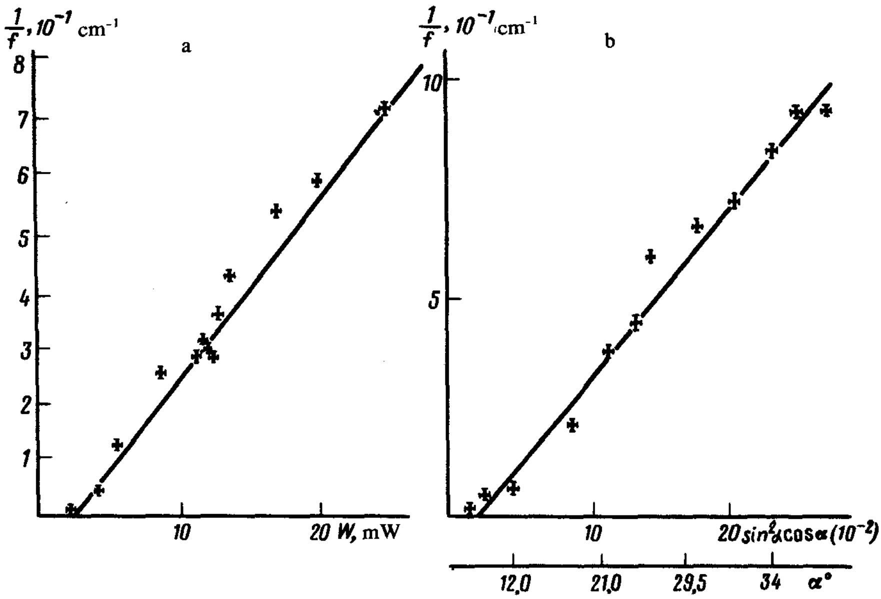


FIG. 3.

means of a Glan prism). The power was varied by changing the filters F_1 and by a slight misadjustment of the laser cavity. The following results were obtained. a) An appreciable increase (by more than a factor of two) in beam divergence is observed as the power is changed from 4 to 35 when the cell is located in the region of the focal constriction. This occurred if the incidence angle of the beam on the cell was different from 0. b) In the $\alpha = 0$ case case no divergence changes were observed with a change in the power. c) When n is perpendicular to $\mathbf{e}_L$, and $\mathbf{e}_L$ lies in the plane of the cell face, no changes of the angular structure were observed at any incidence angles. d) In order to determine the sign of the nonlinearity, the cell was moved to the region beyond the caustic in the diverging beam. In this case the angular divergence decreased with increasing power. Thus, it can be stated that we are dealing with self-focusing, and the orientation-polarization dependence of the effect agrees qualitatively with the theoretical dependence.

The following was done for the purpose of a quantitative comparison of the effect of parameters with theory. A calibration curve was plotted by introducing a series of thin glass lenses with focal lengths from 1.1 to 50 cm into the focal constriction in place of the cell, thereby making it possible to determine from the angular divergence of the beam in the far zone, the optical power of the nonlinear lens induced in the medium. Assuming a Gaussian intensity distribution along the beam cross section, we determine the radius α of constriction from the divergence θ in the far zone as the half-width at a level equal to e^{-2} of the maximum for the distribution $I(r_\perp) \sim \exp[-2r_\perp^2/a^2]$; specifically, $\alpha = HWe^{-2}M = 2c\omega^{-1}[\theta(HWe^{-2}M)]^{-1} = 1.18 \times 10^{-2}$ cm. The intensity at the center of the beam is $|E_0|^2 = (8W/cn\alpha^2)$(ergs/cm^3). The accuracy of the longitudinal placement of the cell in the constriction was ± 0.5 mm. A series of measurements of the power dependence of the beam divergence in the far zone were made at a constant external angle $\alpha_{ext} = 50°$, which corresponds to a refraction angle of $\alpha = 32°$ in the crystal. A graph of the dependence of the power f^{-1} of the nonlinear lens on the beam power was constructed by means of the calibration curve (see Fig. 3a). In an analogous manner the dependence of the power of the nonlinear lens on the parameter $\sin^2\alpha \cos \alpha$, where α is the wave refraction angle inside the cell, was determined for a fixed power $W = 30$ MW (Fig. 3b). Both graphs turned out to be quite linear, in agreement with the theoretical expression (5). A calculation of the proportionality constant in (5) gives the steady-state power of the nonlinear lens (for $H = 0$) that agrees exactly with the experimental value. This exact agreement of the constant may be accidental because of experimental errors and because of imprecise knowledge of the optical and mechanical properties of the NLC. Measurements were made of the variation of the nonlinear-lens power when the beam abruptly turned on; the characteristic time to reach equilibrium amounted to $\tau \sim 10$ sec, whereas a theoretical estimate from Eq. (3) gives $\Gamma_1^{-1} \sim 5$ sec for values of $\gamma \sim 1$ poise, typical for this type of NLC (we did not know the exact value of gamma). If the effective optical nonlinearity constant ϵ_2 is determined from Eq. (6), then the experiment gives $\epsilon_2 = 0.07$ cm^3/erg for an angle $\alpha = 32°$. This value is approximately *nine orders of magnitude greater than* the nonlinearity of a well-known self-focusing medium such as caron bisulfide CS_2, for which $\epsilon_2 = 1.2 \times 10^{-10}$ cm^3/erg.

Thus, in this paper we have theoretically predicted for the first time, as well as detected and studied experimentally, the giant optical nonlinearity of the mesophase of an NLC, which is caused by reorientation of the NLC director due to the action of the fields. This opens up the prospects for developing a whole series of nonlinear optical devices with a very low operating power based on the orientational nonlinearity of liquid crystals; these include bistable optical cavities, wave-front inversion devices, etc.

The authors wish to thank L. M. Blinov, S. M. Arakelyan, N. B. Baranova, E. I. Kats, N. A. Mel'nikov, and Yu. S. Chilingaryan for valuable discussions. In addition, we wish to thank L. M. Blinov for providing the liquid crystal.

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