Strong, nonlinear, optical activity in the nematic phase of a liquid crystal

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Nonlinear optical activity (NOA) in MBBA was detected for the first time by an orientational nonlinearity. Appropriate formulas are given for calculating the NOA parameters in an anisotropic medium. The self-focusing of light (SFL) and defocusing of light (DFL) observed in the experiment are used to explain the NOA. The NOA makes it possible to realize a "single-transmission" polarization of the filled, nonlinear cavities.

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1. The recently discovered giant optical nonlinearity in the mesophase of a nematic liquid crystal (NLC) in an experiment on the self-focusing of light (SLF)[1] leads to several easily observable light-induction effects[2] and makes it possible to investigate nonlinear phenomena by an orientation mechanism in exceptionally small laser fields. We report for the first time in this paper an experimentally observed orientational nonlinear optical activity (NOA)–dependence of the polarization rotation angle on the light intensity[1)]–in thin, plane-oriented (by rubbing) samples (with thickness $d \approx 50$ μm) of the nematic phase of the NLC *N*-(-*n*-methoxybenzylidene)-*n*-butylaniline (MBBA) in the field of a cw argon laser ($\lambda = 4880$ Å) with a power $P \sim 10$ mW (as a result of focusing it on a sample by a lens with $F = 5$ cm). At a power $P \gtrsim 50$ mW the NOA decreases because of the breakup of the NLC orientation by heating. We show that the NOA makes it possible to obtain efficient, "single-transmission" optical elements, which are similar to the filled, nonlinear, Fabry-Perot cavities that have recently been discussed in great detail.[4]

2. The NOA was recorded from the rotation of the polarization of a probing, linearly polarized radiation of an He-Ne laser ($\lambda = 6328$ Å) after turning on the Ar laser with the same polarization that induces the nonlinearity. The measurement procedure, which excluded the influence of various, NOA-simulating factors (associated with variation of the intensity of the transmitted light) was essentially similar to the method described in Ref. 5. The light was obliquely incident on the MBBA layer ($\psi \equiv \widehat{\mathbf{k}\mathbf{m}} \approx 60°$, **k** is the wave vector of the incident **E** wave, and **m** is the director) at $\alpha \equiv \widehat{\mathbf{E}_{np}\mathbf{m}} \approx 10°$ ($\mathbf{E}_{np}$ is the projection of **E** on the plane of the cell).

Figure 1 shows the obtained dependence of the NOA angle for the probing beam on the power P of the Ar laser. We can see that saturation sets in at $P \geqslant 10$ mW. At a constant value of P, ϕ^{nl} was almost independent of the duration Δt of the MBBA irradition (for $\Delta t > 10$ sec). For the "o"($\alpha = 90°$) and "e" polarization ($\alpha = 0°$) of the incident light $\phi^{nl} \approx 0$ (a rotation by more than several tenths of a degree in this case is

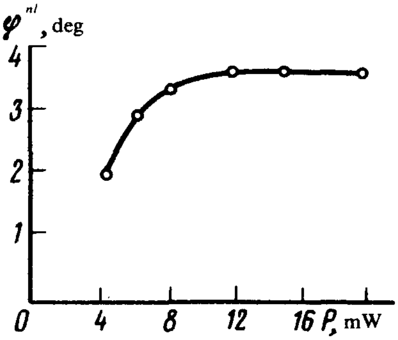

FIG. 1. Dependence of ϕ^{nl} in MBBA for the probing beam on the incident power P of the primary Ar-laser radiation in the geometry $\widehat{\mathbf{kn}} \approx 60°$ and $\widehat{\mathbf{E}_{pn}\mathbf{n}} \approx 10°$.

due to sample imperfection). The value of ϕ^{nl} oscillated as a result of variation of ψ and d. SFL was also observed for the "e" wave at the P values in Fig. 1. At $P \gtrsim 50$ mW an increase of Δt defocused the light (DFL), and the NOA decreased. The P dependence of the "switching" time from SFL to DFL is shown in Fig. 2.

At normal incidence ($\psi = 0°$ and $\alpha = 10°$) a noticeable NOA which was accompanied by SFL, appeared only at $P \sim 100$ mW. However, this was observed only for a short time (~ 100 msec), and DFL appeared with further irradiation. After a prolonged irradiation SFL occurred for the "o" wave in the medium and DFL for the "e" wave. Figure 3 shows the cross-sectional profiles of the probing beam for these cases. Critical opalescence appeared with a further increase of P (at $P \gtrsim 500$ mW). The picture was generally reconstructed after interruption of the Ar laser, although complete reconstruction did not occur at $\Delta t \gtrsim 5$ min.

3. The physical cause of the observed effects is the orienting influence[1,2] of the laser field on the NLC molecules.[2)] It is exceptionally effective for "longitudinal" reorientation (along **d**) for the "e" wave when it is obliquely incident with respect to **m**.[1] A "trans-

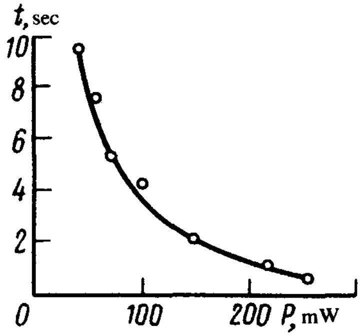

FIG. 2. Time during which DFL develops during prolonged sample irradiation as a function of P ("e" polarization of incident light).

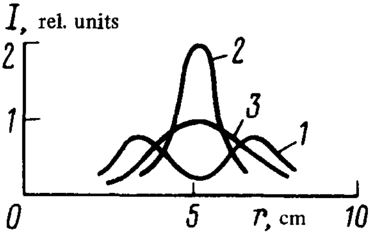

FIG. 3. Cross sections of the probing beam, which passes through MBBA for "e" polarization (1) and for "o" polarization (2) in the presence of Ar-laser radiation and the beam profile (3) in the absence of primary radiation. (The pictures correspond to placement of the NLC cell at the neck behind the focus of the lens; r is the transverse coordinate taken at a distance of 1 m from the cell.)

verse" reorientation, which occurs at $\psi = 0°$ (Ref. 8), is also possible.[3)] These light-induced stimuli also produce NOA because of the different nonlinear corrections, and the refractive index for the "o" and "e" waves.

The characteristics of the examined interaction, in particular ϕ^{nl}, for the "longitudinal" reorientation (along z) can be calculated from the following formulas for the $\mathbf{E}_{np}$ wave transmission through the NLC layer for two, orthogonal polarizations (a single transmission is examined):

$$E_{np}^{\parallel} = \frac{4\sin\psi\,\sqrt{\epsilon_\perp\epsilon_\parallel}\sqrt{\epsilon_\perp - \cos^2\psi}}{(\sqrt{\epsilon_\perp - \cos^2\psi} + \sqrt{\epsilon_\perp\epsilon_\parallel}\sin\psi)^2}\, E_{\text{inc}}^{\parallel} \exp\left\{ i\frac{\omega}{c}\int_0^d (\cos^2\psi + n_z^2)\frac{dz}{n_z} \right\},$$

$$E_{np}^{\perp} = \frac{4\sin\psi\sqrt{\epsilon_\perp - \cos^2\psi}}{(\sqrt{\epsilon_\perp - \cos^2\psi} + \sin\psi)^2}\, E_{\text{inc}}^{\perp} \exp\left\{ i\frac{\omega}{c}\epsilon_\perp d/\sqrt{\epsilon_\perp - \cos^2\psi} \right\}, \tag{1}$$

where

$$n_z = \frac{1}{\epsilon_\perp + \epsilon_\parallel \Delta z^2}\left\{ -\epsilon\cos\psi\,\Delta z + \sqrt{\epsilon_\perp\epsilon_\parallel(1+\Delta z^2)[(\epsilon_\perp - \cos^2\psi) + (\epsilon_\parallel - \cos^2\psi)\Delta z^2]} \right\}$$

is the z component of the $\mathbf{k}n/|\mathbf{k}|$ vector, n is the index of refraction

$$\Delta z = [1 - \exp(-\Gamma t)](E^e\hat{x})(E^e\hat{z})\,\epsilon\,\frac{1}{2\pi^2 y\Gamma}\sin\left(\frac{\pi z}{d}\right)$$

is the nonlinear variation in thickness d of the z component of the director, $\epsilon = \epsilon_\parallel - \epsilon_\perp$ is the anisotropy of the NLC, $\psi = \hat{\mathbf{k}\cdot\mathbf{m}}$, $1/\Gamma$ is the relaxation time of the

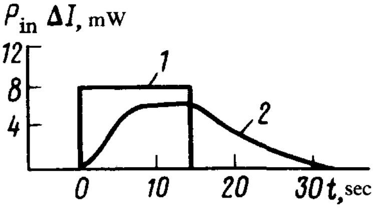

FIG. 4. Time scan for P_{in} (1) and ΔI (2) in presence of NOA; ΔI was obtained as a result of transmission of the probing beam through the MBBA layer placed between two polarizers (whose crossed angle is $\approx 30°$) in the presence of an Ar-laser pulse stimulus P_{in}.

medium, γ is the viscosity, $\hat{\mathbf{x}}$ and $\hat{\mathbf{z}}$ are unit vectors ($\hat{\mathbf{x}} || \mathbf{m}$), and $\epsilon_{air} = 1$.

A computer calculation using Eq. (1) gives the $\phi^{nl}\phi(P)$ dependence, which is qualitatively identical to that obtained in the experiment (Fig. 1). The saturation of ϕ^{nl} is due to the maximum orientation of the molecules (in the middle of the NLC sample) along **k**, which is achieved only at high powers. (The orientation of molecules at the surfaces adjacent to the NLC is specified and determined by the initial conditions.)$^{4)}$

Equation (1) explains the experimentally observed oscillations of ϕ^{nl} as a result of variation of ψ and d, as well as the absence of NOA for the "o" and "e" waves, for which SFL and DFL are possible. The experimentally obtained results make it possible to distinguish between their orientational and thermal mechanisms.$^{5)}$

Since n_o increases with temperature for MBBA and n_e decreases,[6] a thermal SFL appears for the "o" wave in the medium, and DFL appears for the "e" waves. Therefore, the SFL recorded for the "e" wave in the experiment in Fig. 1 has an orientational nature and the dependence in Fig. 2 is attributable to the thermal mechanism.$^{6)}$

Thus, we have recorded the NOA in the orientational nonlinearity (Fig. 1).

Since the NOA is accompanied by SFL at $\psi = 0$, its nature is also orientational ("transverse"). However, at large P and Δt the thermal effects play the main role, and heating of the medium results in opalescence.

4. The evolution dynamics of the NOA is shown in Fig. 4, where the shape of the input Ar-laser pulse P_{in} and the corresponding intensity variation ΔI of the probing beam at $P = 8$ mW are shown [the duration of P_{in} was determined by a shutter with $\Delta t = 15$ sec and by the response speed (duration of the wave fronts) ~5 nsec]. The asymmetry of the leading and trailing edges of the probing beam ΔI can be seen (in contrast to the symmetrical shape of P_{in}); this is due to relaxation of the medium. This, for example, produces a hysteresis loop in the ΔI vs. P_{in} dependence, which was measured at the same instant of time (see Ref. 7).

Thus, the NOA makes it possible to use a "single-transmission" polarization type of device with operating modes typical of an intra-cavity, nonlinear element.[7] Specifically, Fig. 4 corresponds to the power-limitation regime.

We thank N. V. Tabiryan for a useful discussion.

[1] In contrast to the isotropic media, in which this effect, called the rotation of the polarization ellipse,[3] was usually studied, it also appears here as a result of linear polarization of the incident light; we, therefore, call it NOA, although NOA is usually associated with spatial dispersion of a medium.[9]

[2] Strictly speaking, only a polarization induction, rather than true rotation of the molecules, can occur.

[3] A "longitudinal" reorientation in this case can also occur because of a possible orientation of the molecules at some angle **k** to the glass surfaces of the cell.

[4] Since these conditions can break down in the actual experiment, the experimentally obtained ϕ^{nl} values can exceed the ϕ^{nl} calculated from Eq. (1) because of the analogy of the structure, which appears in an NLC in the examined case, to the Mogen limit for a CLC.[6]

[5] This distinction is not always easy to draw directly from the transient time, since the characteristic constants for NLC vary within broad limits for different types of strain.[6]

[6] The orientation mechanism can also play a role here: the laser field removes the NLC molecules from the local equilibrium states and they "stick" along the rubbing direction. However, the picture in this case must be opposite to that which appears in the presence of a thermal stimulus (since $n_e > n_o$).

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