Transformation of elliptical polarization of light wave into linear polarization in the isotropic phase of a nematic liquid crystal

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The self-precession of the polarization ellipse of a light pulse, whose duration is shorter than the isotropy relaxation time τ in the isotropic phase of a nematic liquid crystal, is accompanied by a degeneration: The wave becomes linearly polarized. Near the phase-transition temperature, an anomalously large increase in τ (by four orders of magnitude) is observed. This increase depends on the intensity of the light pulse.

Self-rotation of the polarization ellipse of a light wave is one of the classical effects of nonlinear optics.[1] A detailed study has been made of the steady state of this polarization self-effect, in which the eccentricity of the ellipse remains constant. In the time-varying case, in which the length of the light pulse, t_p, is comparable to the relaxation time of the nonlinear response of the medium, τ, the rotation of the polarization ellipse may be accompanied by a change in its eccentricity, according to numerical calculations.[2] In the present letter we are reporting the first experimental observation of these effects. It has been shown that if the condition $t_p \ll \tau$ holds, and if the light pulse has a sufficiently high energy, the polarization of the pulse degenerates to linear.

As the nonlinear medium for this study we selected the isotropic phase of a nematic liquid crystal, which has large cubic susceptibilities χ and which allows one to tune χ and τ by varying the temperature [as the temperature of the transition to the nematic phase, T^*, is approached, the values of χ and τ increase $\propto (T-T^*)^{-1}$; Ref. 3]. An elliptically polarized, collimated beam from a single-frequency, single-mode neodymium laser ($\lambda = 1.06\,\mu$m, pulse energy of 25 mJ, pulse length of 30 ns) passed through a cell (with a length $L = 3$ cm) filled with a nematic liquid crystal ("mixture A"[4]) in the isotropic phase (Fig. 1a). The size of the laser beam waist in the cell was $w_0 \approx 0.7$ mm. The degree of ellipticity of the polarization of the laser beam, e (i.e., the ratio of the minor semiaxis of the polarization ellipse to the major semiaxis), was determined from measurements of its normalized Stokes parameter ξ_2, i.e., from the expression $e = [1-(1-\xi_2^2)^{1/2}]/\xi_2$, before the beam entered the cell and after the beam left the cell. A Fresnel rhombus was placed in the path of the laser beam for these measurements of ξ_2. The rhombus was oriented in such a way that the light with the right-hand (left-hand) circular polarization was linearly polarized in the vertical (horizontal) plane after passing through the rhombus. The beam then passed through a Glan prism, which transmitted the light with the horizontal polarization in the

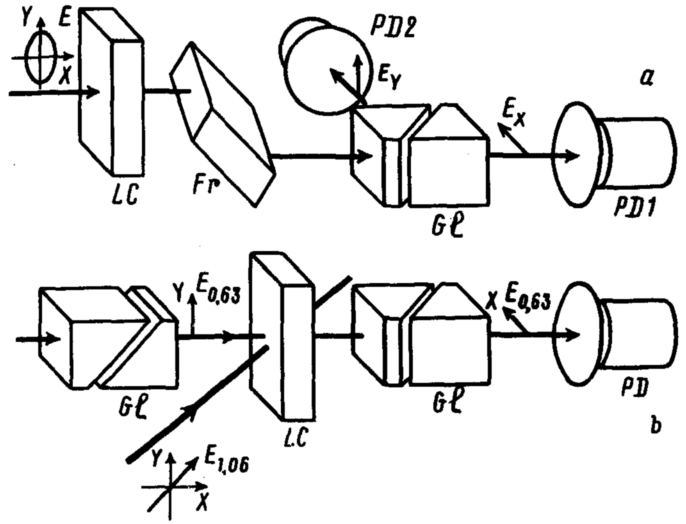

FIG. 1. Layout for measurements of (a) the Stokes parameter ξ_2 and (b) the anisotropy relaxation time τ. LC—Cell holding the nematic liquid crystal in the isotropic phase; Fr—Fresnel rhombus; Gl—Glan prism; PD—photocell.

forward direction, while deflecting the light with the vertical polarization. The value of ξ_2 in this case is equal to the difference between the intensities of the linearly polarized waves emerging from the Glan prism with vertical and horizontal polarization, divided by the sum of these intensities. In the experiments, the intensities of these wave were measured by two FK-19 photocells, which were connected to an S8-14 oscilloscope for a study of the time evolution of ξ_2.

Figure 2 shows oscilloscope traces and the corresponding time evolution of the Stokes parameter ξ_2 of the laser pulse transmitted through the layer of medium for various values of the temperature of the medium, T (the value of the Stokes parameter at the entrance to the medium was $\xi_2^0 = 0.78$, which corresponds to the value $e_0 = 0.32$). We see that the change in ξ_2 increases as T approaches the temperature of the transition to the nematic phase, $T^* = 74.5$ °C. At $T = 75.5$ °C the elliptical polarization degenerates into a linear polarization ($\xi_2 = 0$) and then converts back into a elliptical polarization, but with rotation in the opposite direction ($\xi_2 < 0$) and with a much smaller degree of ellipticity. At $T = 86$ °C the elliptical polarization, degenerating into a linear polarization, remains linear to the end of the pulse. A further increase in the temperature to $T = 97.5$ °C weakens this polarization self-effect.

In a theoretical analysis of the evolution of an elliptically polarized light wave, we describe the perturbations of the dielectric tensor $\delta\epsilon_{ij}$ of the isotropic phase of the liquid crystal by the following equation, as in Ref. 4:

$$\left(\tau\frac{\partial}{\partial t} + 1\right)\delta\epsilon_{ij} = 2\pi\chi\Big(E_iE_j^* + E_i^*E_j - 2\delta_{ij}\sum_k |E_k|^2/3\Big), \tag{1}$$

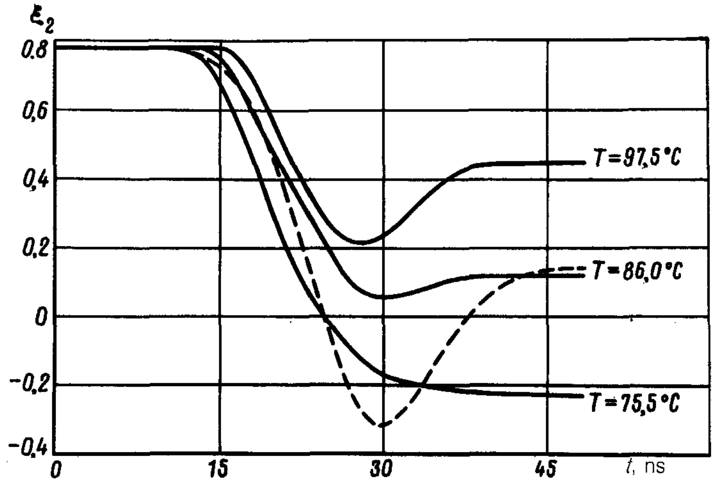

FIG. 2. Time evolution of the Stokes parameter ξ_2 of the laser pulse transmitted through the layer of nematic liquid crystal in the isotropic phase. The dashed line is theoretical and corresponds to $T = 75.5$ °C.

where $E_{i,j}$ are slowly varying components of the electric vector of the light wave. Assuming the light wave to be a plane wave propagating along a z axis, we find the following system of equations for the normalized Stokes parameters ξ_α $(\alpha = 1,2,3)$ from Maxwell's equations incorporating the perturbations $\delta\epsilon_{ij}$:

$$\frac{\partial \xi_\alpha}{\partial z} = e_{\alpha\beta\gamma}\epsilon'_\beta \xi_\gamma; \quad \left(\tau\frac{\partial}{\partial t} + 1\right)\epsilon'_{1,3} = gI\xi_{1,3}; \quad \epsilon'_2 = 0. \tag{2}$$

Here $I = cn(|E_x|^2 + |E_y|^2)/8\pi$, $g = (4\pi/cn)^2\omega\chi$ and $e_{\alpha\beta\gamma}$ is the Levi–Civita density. Equations (2) must be supplemented with boundary and initial conditions: $\xi_\alpha(0,t) = \xi_\alpha(z,0) = \xi^0_\alpha$; $\epsilon'_\alpha(0,t) = \epsilon'_\alpha(z,0) = 0$.

In the very unsteady case $(t_p \ll \tau)$, Eqs. (2) can be reduced to a system of ordinary differential equations through the use of the self-similar variable $\eta = gz\int_0^t I(t')dt'$. From this system of equations we find

$$\left(\frac{d^2}{d\eta^2} + \frac{1}{\eta}\frac{d}{d\eta} + \left[1 - \frac{2}{\eta^2}\int_0^{\eta}\int_0^{\eta'} \xi_2(\eta'')d\eta' d\eta''\right]\right)\xi_2 = 0. \tag{3}$$

For small values of ξ_2 $(|\xi_2| \ll 1)$, in the approximation linear in ξ_2, Eq. (3) becomes the differential Bessel equation. At large values of η we thus have a solution which describes damped oscillations of the degree of ellipticity: $\xi_2 \approx \xi^0_2\sqrt{2/\pi z}\cos(\eta + \pi/4)$. Figure 2 shows the results of a numerical solution of Eq. (3) for the boundary condition $\xi^0_2 = 0.78$, which corresponds to the experiments. The parameter value $\chi/\tau = 1.1 \times 10^{-3}$ cm·s/g was chosen to bring the zeros of the theoretical and experimental curves of ξ_2 versus t into coincidence. This value agrees in order of magnitude

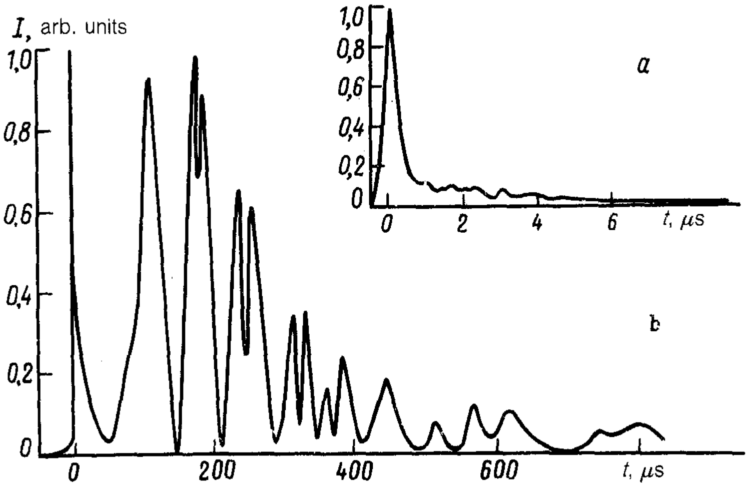

FIG. 3. Time evolution of the transmission of polarization shutter with a nematic liquid crystal in the isotropic phase at $T = 75.5$ °C. The energy density of the controlling pulse is (a) 0.1 or (b) 5 J/cm^2.

with $\chi/\tau = 1.8 \times 10^{-3}$ cm·s/g for MBBA and the ratio $\chi/\tau = 3.4 \times 10^{-3}$ cm·s/g for EBBA; the latter values were calculated from the results of Ref. 4.

Let us examine the possibility of an onset of self-focusing. Self-focusing is unimportant if the displacement of the rays of a Gaussian beam due to the nonlinear refraction is considerably smaller than the waist size w_0. In the thin-lens approximation, the ray deflection angle is $\vartheta = (\lambda/2\pi)(\partial\phi_{NL}/\partial r_\perp)$, where ϕ_{NL} is the nonlinear phase shift, and $r_\perp$ is the transverse coordinate. Assuming $(\partial\phi_{NL}/\partial r_\perp) \sim \phi_{NL}/w_0$ for an estimate, we find the condition under which there is no self-focusing: $\phi_{NL} \ll 2\pi w_0^2/(\lambda L)$ or $\phi_{NL} \ll 40\pi$. In these experiments, self-focusing can therefore be ignored.

Near the phase-transition temperature $T^* = 74.5$ °C, an anomalously large increase in τ (by four orders of magnitude; this increase depends on the intensity of the controlling pulse) was observed during measurements of τ in the isotropic phase of the liquid crystal. In these measurements, the cell with the nematic liquid crystal was placed between two crossed Glan prisms, which constituted a polarization shutter controlled by the laser pulse (Fig. 1b). As the energy density of the laser pulse was increased from 0.1 to 5 J/cm^2, the relaxation time increased from ≈ 100 ns to ≈ 1 ms. Figure 3 shows two curves of the time evolution of the intensity of the light from a probing He–Ne laser after passage through the polarization shutter. These curves were recorded at energy densities of (a) 0.1 and (b) 5 J/cm^2 of the controlling pulse from the neodymium laser. The anomalously large values of τ in the isotropic phase of nematic liquid crystal and the dependence of τ on the energy density of the controlling light pulse are not described by the theory of Ref. 3. The explanation may lie in a relatively pronounced ordering of molecules induced by the optical field. The electric field of the controlling pulse causes a pronounced change in the distributions of molecules with respect to orientation and angular velocity, thereby influencing the nature of the interaction of the molecules after the field is removed. We furthermore do not rule out the possibility that vortical hydrodynamic flows arise as a result of the application of the laser beam.

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