# Influence of External Electric Field on Time of Nonlinear Optical Reorientational Effect in Nematics

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Nonlinear reorientation phenomena in nematic liquid crystals causes an extremely large refractive index changes. However, this effect is relatively slow and determination the time necessary to appear or disappear the nonlinear effect is important issue. In this work we present measurements of the time of increasing the nematics reorientation induced by the light beam passing through the liquid crystalline layer. The influence of external low-frequency electric field suppressing reorientation is also reported. Obtained results describing relations between time and optical power of light as well as between time and external electric field intensity are in good agreement with theory.

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# 1. Introduction

Liquid crystals are well known anisotropic materials and their properties were analyzed in many books and papers [1-2]. In the last years the growing interest in new applications of liquid crystals is still observed. Nematic liquid crystals are also excellent media in nonlinear optics. They have the giant optical nonlinearity due to reorientational phenomenon, which is not observed in other types of materials. The main drawback in applications of reorientational nonlinearity in nematics is a relatively low time necessary to appear and disappear the nonlinear effect. In this paper the detailed measurements of the relaxation time of appearing the nonlinearity is presented.

The reorientation nonlinearity can be changed by external low-frequency electric field which modifies the value of nonlinear refractive index and also the time of appearing the nonlinear effect. In this work we present experimental results of influence of electric field on observed nonlinearity.

#### 2. Theory

The reorientation process is investigated in the nematic liquid crystalline cell with homeotropic alignment (figure 1). The director  $\mathbf{n}$ , defined the mean direction of molecules long axis, changes locally its direction due to both optical and electric external fields [1-2]. In analyzed configuration electric fields and director at boundaries



Fig. 1. Light propagation through the nematic film with applied electric field. Director  $\mathbf{n}$  defines the average direction of long axis of the liquid crystal molecules.

define one plane, that allows to introduce an angle  $\theta$ : **n**=(sin $\theta$ , 0, cos $\theta$ ), which describes the orientation of liquid crystals. The interactions between molecules as well as between molecules and external fields lead to appearing new equilibrium state. After minimization the total free energy from the Euler–Lagrange–Rayleigh equations the change of the orientation angle  $\theta$  is obtained in the form [1-2]:

$$K\frac{\partial^2\theta}{\partial x^2} + \left(\frac{\varepsilon_0\Delta\varepsilon}{4}E^2 - \frac{\varepsilon_0\Delta\varepsilon_e}{2}E_e^2\right)\sin 2\theta = \gamma\frac{\partial\theta}{\partial t},\qquad(1)$$

where  $\Delta \varepsilon = n_e^2 - n_o^2$  is the optical anisotropy described by extraordinary  $n_e$  and ordinary  $n_o$  indices,  $\Delta \varepsilon_e = \varepsilon_{\parallel} - \varepsilon_{\perp}$  is the electric anisotropy described by parallel  $\varepsilon_{\parallel}$  and perpendicular  $\varepsilon_{\perp}$  part of electric permittivity,  $\varepsilon_0$  is the permittivity of the free space, *K* is the elastic constants (in one-elastic constant approximation) resulted from the liquid crystalline elastic deformation, and  $\gamma$  is the rotational viscosity of liquid crystals. The electrical fields are denoted as *E* for optical field and  $E_e$  for low-frequency one.

Assuming small reorientation  $\sin \theta \approx \theta$  in homeotropic strong anchoring alignment  $\theta(x=0)=\theta(x=d)=0$ , one can find the simplified solution in the form:  $\theta(x,t) = \theta_0 \exp(t/\tau_0) \sin(\pi x/d)$ , where the relaxation time  $\tau_0$  is defined by:

$$\frac{\gamma}{\tau_0} = \frac{1}{2} \varepsilon_0 \Delta \varepsilon E^2 - \varepsilon_0 \Delta \varepsilon_e E_e^2 - \frac{\pi^2 K}{d^2}.$$
(2)

The reorientation begins for  $\tau_0 > 0$ , which requires that optical field is larger than the threshold value:  $E^2 \ge \frac{2\Delta\varepsilon_e}{\Delta\varepsilon} E_e^2 + \frac{2\pi^2 K}{\varepsilon_0 \Delta\varepsilon d^2}$ . It can be expected similar relation to this described by equation (2) if the incident light has finite size dimension. Generally, the laser sources have the Gaussian-shape beam with the electric field intensity distribution in the form  $E^2(r) = E^2 \exp(-2r^2/w_0^2)$ , where  $w_0$  is the beam waist. With small reorientation angle approximation, the steady-state distribution of the angle  $\theta$  is now described by the relation  $\theta(r, x, t) = R(r, t) \sin(\pi x/d)$ , which satisfies the strong anchoring conditions ( $\theta=0$  for x=0 and x=d). The transversal profile of reorientation in one-constant approximation takes the form [3-5]:

$$K\frac{\partial^2 R}{\partial r^2} + \frac{K}{r}\frac{\partial R}{\partial r} - \frac{K\pi^2}{d^2}R + \left[\frac{\varepsilon_0\Delta\varepsilon}{4}E^2\exp\left(-\frac{2r^2}{w_0^2}\right) - \frac{\varepsilon_0\Delta\varepsilon_e}{2}E_e^2\right](2R - R^3) = \gamma\frac{\partial R}{\partial t}, \quad (3)$$

where *r* is the radial coordinate. In a case of small reorientation ( $R^3 << R$ ) the amplitude of transversal profile *R* can be approximated by the Gaussian shape and the simplified solution in the form  $R = R_0 \exp(t/\tau)\exp(-r^2/w^2)$  can be assumed. The corresponding relaxation time  $\tau$  is respectively defined by [6]:

$$\frac{\gamma}{\tau} = \frac{1}{2} \varepsilon_0 \Delta \varepsilon E^2 \exp\left(-\frac{2r^2}{w_0^2}\right) - \varepsilon_0 \Delta \varepsilon_e E_e^2 - \frac{K\pi^2}{d^2} - \frac{4K}{w^2} \left(1 - \frac{r^2}{w^2}\right).$$
(4)

The relaxation time  $\tau$  corresponded to the finite size beam has a similar dependence on electric fields like relaxation time defined for plane wave  $\tau_0$ . Because the intensity of light beam is dependent on the transversal coordinate *r*, the time  $\tau$  is also changing across the light beam. Additionally, the relaxation time is dependent on the width *w* which is dependent on the light beam width  $w_0$ . That means, that the time of increasing and decreasing of nonlinear reorientation in the light beam center depends on the light beam width.

#### 3. Experimental

The theoretical prediction was checked in the experimental set-up presented in Figure 2a. As the light source we used semiconductor laser at wavelength 842 nm, which was pig-tailed with a single mode fiber. After focalizing the beam waist was equal to 5.3  $\mu$ m and the focus was placed in the half of the cell thickness. As a liquid crystal we used 4-trans-4'-n-hexyl-cyclohexyl-isothiocyanatobenzene (6CHBT). The



Fig. 2. Observation and detecting nonlinear reorientation time of response of nematic molecules caused by external electric and optical fields: experimental set-up a) and typical diffraction image with added black circle marking the position of detector b).



Fig. 3. Method of extrapolation the nonlinear reorientation time from experimental data.

behavior of the nematic layer of thickness  $60 \,\mu\text{m}$  was observed simultaneously by the CCD camera and the detector with 2 mm aperture set in the place of appearing minimum interference ring (see figure 2b). The low-frequency electric field was obtained from the square function generator.

In the figure 3 the response of the nematic layer on switching of the laser to the power of 51 mW for 27 seconds is presented. First, the optical field generated three minimum interference rings (compare with figure 4). Appearing the fourth interference ring was blocked by existing electric field at 8.3 kV/m and in the end of measuring gate it disappeared. The dynamics is especially important in the initial reorientational process, therefore we take into account only the period of time need to achieve the first maximum (zoomed frame in figure 3). The data in figure 3 was fitted by the exponential function  $y = -0.06 + 0.06 \exp(t/\tau)$ .



Fig. 4. Time dependence of output beam in diffraction image cross-section due to switching on the laser.



Fig. 5. Relation of inverted reorientation time vs. square electric field for different values of input light power.

The inverted time  $\tau$  dependence on square electric field for different values of input light power is shown in figure 5. The obtained inverse proportional relation of characteristics is fully agreed with the theoretical prediction made in the equation (4). The initial perturbations were a result of instabilities of generator, and they are not essential for making the trend of curve shape.



Fig. 6. Relation of inverted reorientation time vs. input light power for different values of square electric field.



Fig. 7. Influence small changes of input light power and value of applied electric field on observable appearing and disappearing of new minimum interference rings.

The square optical field can be easy transform in optical power, and the inverted time  $\tau$  should be proportional to the power. Results presented in figure 6 confirm this relation.

The influence of the small changes of input light power and value of the applied electric field on observable appearing and disappearing of new minimum interference rings is presented in the figure 7. The number of visible minimum rings decrease rapidly if we put strong enough electric field or too small the input laser power.

## 4. Conclusions

It should be noted, that we measured time of appearance the interference rings, which is not exactly the relaxation time described by equation (4). However, the existence of rings is determined by the phase difference between the center and the edge

of light beam out-coming from the nematic layer. This means, that the measured time is roughly equal to the relaxation time defined by equation (4) in the beam center  $\tau$ (*r*=0). Obtained results shows, that the measured time is dependent on the electric and optical field as defined by relation (4). By using these relations it is possible to measure material constants in nematic liquid crystals: elastic constant *K* and rotational viscosity  $\gamma$ . Measured values are in good agreement with measurements obtained by another methods. It seems, that the proposed method gives better results than the methods based on determination of the threshold power.

In our experiments the low-frequency electric field suppress the reorientation. As a result the increasing the applied field intensity causes increasing the time of appearance the nonlinear effect. However, if the low-frequency electric field support the reorientation the relaxation time will be reduced. Such method of increasing the speed of nonlinear effect can be applied for example in nematics with negative electrical anisotropy.

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