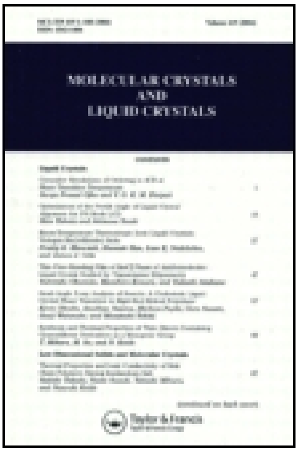

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Reorientation of Director of Nematic Liquid Crystals, Doped with Azodyes, under Light and Low-Frequency Fields

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Reorientation of Director of Nematic Liquid Crystals, Doped with Azodyes, under Light and Low-Frequency Fields

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An interaction of the director of NLCs, doped with mono- and diazodyes and showing the sign-inversion nonlinearity, with the simultaneously applied light and low-frequency electric fields has been studied using the technique of the aberrational self-action. Mathematical simulation of this interaction was performed. A fair fit to the experimental data was obtained.

Keywords nematic liquid crystal; dye; electric field; magnetic field

INTRODUCTION

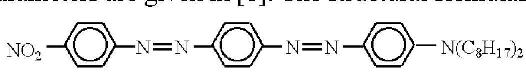
The study of NLCs under the simultaneosly applied light and low-frequency (electric or magnetic) fields started in [1]. This work showed experimentally that the low-frequency field, depending on the sign of low-frequency dielectric anisotropy $\Delta\varepsilon^{LF}$, can both increase ($\Delta\varepsilon^{LF} > 0$) and decrease ($\Delta\varepsilon^{LF} < 0$) the light induced Freedericksz transition (LIFT) threshold in homeotropically aligned NLC. For planarly aligned NLCs with $\Delta\varepsilon^{LF} > 0$, an application of the low-frequency field was accompanied by the aberrational self-focusing of normally incident light beam (absent without the field). Further studies [2-5] showed that the magnetic or electric fields, applied along the director of the homeotropically aligned NLC, can initiate the director field hysteresis observed at increase and subsequent decrease of the light beam power. In other words, external field transforms LIFT, being conventionally the second-order transition [6,7], into the first-order one.

In dye-doped NLCs, in which the efficiency of the light-induced director reorientation (LIDR) can be one or two orders higher than in the transparent NLCs, new features emerge in the interaction with the light and quasistatic electric fields. Thus in NLC with planar alignment, doped with conformationally active monoazodye DEANAB, the change in the LIDR sign (i.e., the nonlinearity sign) was observed under action of external low-frequency field.

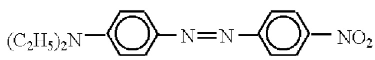
Below we report the results of the detailed study of the behaviour of NLCs, doped with the conformationally active dyes, under combined action of the light and low-frequency electric fields.

EXPERIMENTAL CONDITIONS

Experiments were performed on liquid-crystalline material ZhKM-1282, doped with diazodye KD-1 (0.025 wt.%), with homeotropic and planar alignment, and on ZhKM-1282, doped with monoazodye DEANAB (0.1 wt.%), with planar alignment. In addition, the nondoped homeotropic ZhKM-1277 and planar ZhKM-1282 were studied for comparison. The thickness of all the samples was $L \cong 100\ \mu m$. Nematic matrices ZhKM-1277 and ZhKM-1282 have positive low-frequency dielectric anisotropy and are the mixtures of cyanobiphenyls and esters; their parameters are given in [8]. The structural formulas of the dyes are:



KD-1



DEANAB

NLCs, doped with KD-1 or DEANAB, exhibit the sign-inversion nonlinearity [9]; the values of the critical angle between the director and the wave vector, at which the nonlinearity sign changes from the negative to positive, are $\beta_c = 15^o$ and 50^o, respectively.

The low-frequency electric field was applied to SnO_2 electrodes deposited on the interior side of the glass walls of the NLC cells.

A horizontally polarized light beam from argon laser (λ=515 nm) was focused into NLC by a lens (f = 18 cm). The cell plane was vertical.

The incidence angle α was changed by the cell rotation about the vertical axis (the angle α was taken positive for counterclockwise rotation and negative in the opposite case). The wave excited in NLCs was of the extraordinary polarization.

In narrow laser beams LIDR is accompanied by a light beam self-action: a system of aberrational rings can be observed on a screen placed behind a NLC. The number of rings N is related to the director rotation [10]. The sign of the self-action indicates to the director reorientation sense. If the director orients towards the light field (positive nonlinearity) the self-focusing is observed. In the opposite case, when the director orients perpendicularly to the light field (negative nonlinearity), the self-defocusing takes place. We determined the sign of the self-action by slightly shifting the NLC perpendicularly to the beam and observing the transformation of the aberrational pattern [9].

In measurements of the N(U) dependences, when coming to the next U value, the NLC illumination was interrupted by the time of about 30 s, being sufficient for the director to relax.

EXPERIMENTAL RESULTS AND DISCUSSION

Homeotropically Aligned Samples

Because the low-frequency field supports the homeotropic alignment, its increase results in monotonic (see Figure 1) decrease in the number of aberrational rings N. It is evident that the suppression of the self-focusing aberrational pattern therewith is more efficient for the dye-doped sample (the powers P of the light beam were chosen so that the N values for transparent and dye-doped NLCs coincided at U=0). This difference, as was found in an additional experiment, was not observed outside the dye absorption band. This fact and our theoretical estimates indicate that the dielectric and nonlinear optical properties of homeotropic ZhKM-1277 and ZhKM-1282 matrices are very close. The origin of the above difference can be explained as follows. For transparent NLC, the torque on the director **n** exerted by the electric light field **E** is proportional to the optical anisotropy $\Delta\varepsilon$, while for the doped NLC - to the effective optical anisotropy $\Delta\varepsilon_{eff}$ with the value and sign depending upon angle β between director **n** and wave vector **k** [9]. In the region of positive nonlinearity (in our experiment the refraction angle α_t exceeded β_c), $\Delta\varepsilon_{eff}$ decreases with β. The higher is U the less is β and hence the orienting effect of light beam (at beam powers unchanged) in doped NLC is less than that in the transparent one. Correspondingly, the N value for the doped NLC also is smaller.

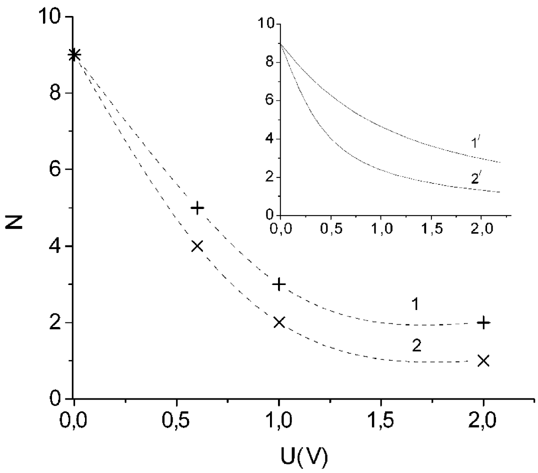


FIGURE 1. Experimental (1 and 2) and theoretical ($1'$ and $2'$) dependences of the number of the self-focusing rings N on low-frequency voltage U at oblique (α=52°) incidence of light beam on homeotropically aligned transparent ZhKM-1277 (✚, P=95 mW) and doped ZhKM-1282+0.025% KD-1 (✕, P=2 mW).

Planarly Aligned Samples

In NLC samples with planar alignment the dependences of N on U, as a rule, are nonmonotonic (Figures 2-4).

At oblique incidence, the dependences N(U) prove to be different for counterclockwise (α>0) and clockwise (α<0) NLC rotation (Figures 2b, 3b, и 4a). In NLC with DEANAB, at α>0 increasing U results in the change of the light beam self-action sign from positive to negative (self-focusing gives way to self-defocusing).

Consider first, following [1], the nonmonotonic character of the N(U) dependence using normal incidence (Figures 2a and 3a) as an example. The number of rings N is proportional to the difference in the e-wave refractive indices at the axis and the periphery of the light beam. At U=0 the director **n** is parallel to the light field **E** everywhere in NLC and, hence, the refractive index is uniform, which leads to N=0. At higher U, the effect of the low-frequency field substantially exceeds that of the light field; therefore, the director field is again uniform and N=0. At the intermediate U values, the effects of the light and the low-frequency fields are comparable and so the director rotation at the beam

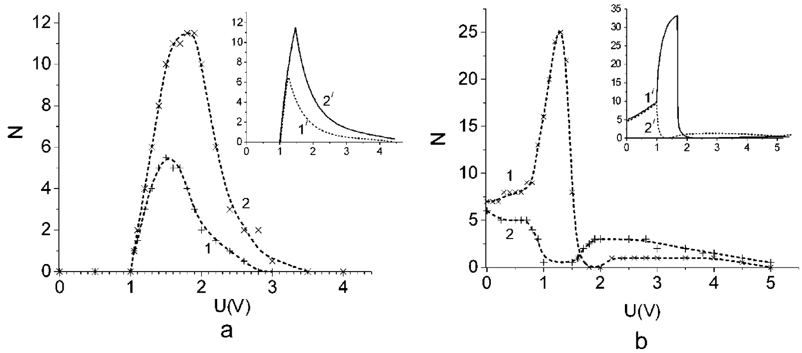


FIGURE 2. Experimental (1 and 2) and theoretical ($1^{/}$ and $2^{/}$) dependences of the number of self-focusing rings N on low-frequency voltage U for planarly aligned ZhKM-1282: (a) Normal incidence of light beam with power P = 35 mW (✚,$1^{/}$) and 70 mW (✕, $2^{/}$). (b) Oblique incidence of light beam with power P = 70 mW at angles $\alpha = +60^{o}$ (✕,$1^{/}$) and -60^{o} (✚,$2^{/}$).

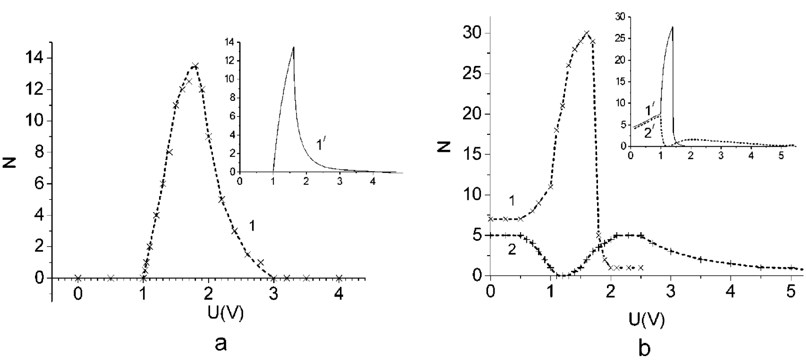


FIGURE 3. Experimental (1 and 2) and theoretical ($1^{/}$ and $2^{/}$) dependences of the number of self-focusing rings N on low-frequency voltage U for planarly aligned dye-doped ZhKM-1282+0.025% KD-1 (beam power P = 1 mW): (a) Normal incidence of the light beam (✕,$1^{/}$). (b) Oblique incidence of the light beam at angles $\alpha = +50^{o}$ (✕,$1^{/}$) and -50^{o} (✚,$2^{/}$).

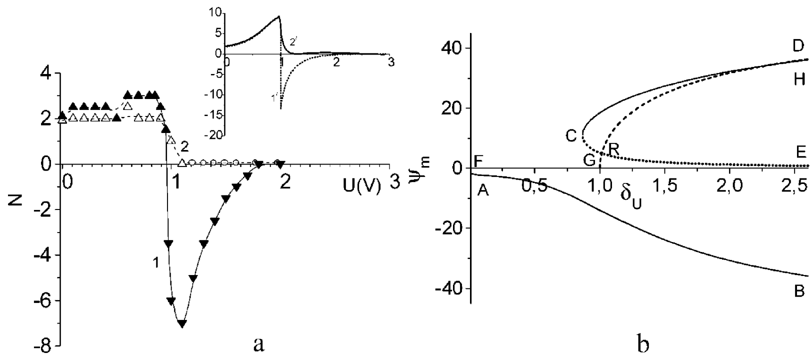


FIGURE 4. (a) Experimental (1 and 2; ▲ and Δ - self-focusing, ▼ - self-defocusing, O - the sign undefined) and theoretical ($1^{/}$ and $2^{/}$) dependences of the number of aberrational self-action rings N of light beam (P = 1.1 mW) incident at an angle $\alpha = +60^{o}$ (▲,▼,$1^{/}$) or α = - 60^{o} (Δ, O, $2^{/}$) on planar ZhKM-1282+0.1% DEANAB on low-frequency voltage U. (b) Theoretical dependence of the stationary director rotation angle ψ_m on scaled voltage $\delta_U = U/U_{th}$ (solid curve - stable states, dotted line (CE) - unstable states, dashed line (GH) - stable states without light).

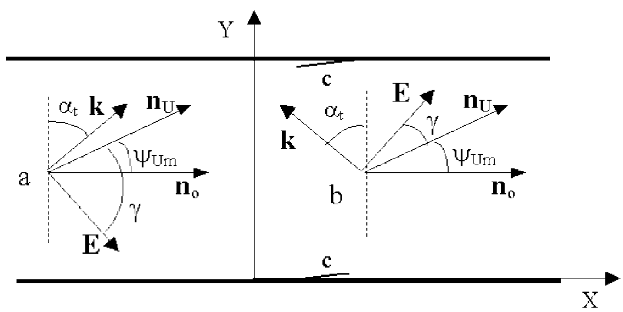


FIGURE 5. Interaction of a light beam with planarly aligned NLCs at $\alpha>0$ (a) and $\alpha<0$ (b). **E**, **k** are electric field and wave vector of light wave, $\mathbf{n}_o$ is unperturbed director, $\mathbf{n}_U$ is the director, rotated by an angle ψ_{Um} by low-frequency field prior to illumination, α_t is angle of refraction, γ is angle between $\mathbf{n}_U$ and **E**; c is a length indicating the pretilt.

axis (the both fields are applied) and the beam periphery (the low-frequency field is only applied) is different. Therefore the refractive index turns nonuniform thus producing the aberrational pattern.

To understand the effect of the light wave vector direction on the character of the N(U) dependences we notice that the sense of the director rotation by the low-frequency field, occurring prior to illumination (counterclockwise in Figure 5), is specified by the pretilt determined by the substrate rubbing direction. In this situation, the angle γ that the rotated director $\mathbf{n}_{Um}$ makes with the light field **E** in NLC center is different for the different directions of the light beam incidence on the NLC. Therefore the light-induced refractive index is also different. At $\alpha>0$, the angle γ increases with U, similar to the case of the normal incidence. Hence, the dependence N(U) is also similar to that for the normal incidence. At $\alpha<0$ and increasing U the angle γ approaches zero first, and then increases. This dependence is also reproduced for the number of rings. Decreasing N at higher U, as in the case of normal incidence, is due to the director field turning more uniform.

Finally, consider the sign-inversion dependence N(U) (Figure 4a, curve *1*) for NLC with DEANAB. Here, if the angle between **n** and **E** equals the critical value $\gamma_c = 90^o - \beta_c = 40^o$, the LIDR sign changes from positive to negative [9]. From Figure 5, it is evident that the change in the LIDR sign occurs at an angle $\psi_{Um} = \gamma_c - \alpha_t$ of the director rotation under low-frequency field. At $|\alpha|=60^o$ the angle of refraction is α_t~30°. Thus, as follows from Figure 5a, for $\alpha = +60^o$ the nonlinearity changes sign at ψ_{Um}~10°, which is observed experimentally. For $\alpha = -60^o$ (Figure 5b), the change in the sign must take place at ψ_{Um}~70°, which is attained at much higher U. In this case, the director field will be quite uniform, which is why the observation of the aberrational self-action is hardly possible (see Figure. 4a, curve *2*).

Calculation of the LIDR and the number of aberrational rings N.

The director reorientation equation can be derived from the balance of the torques produced by elastic and viscous forces and by the light and low-frequency fields. In the one-constant approximation in the reference frame shown in Figure 5 the equation has the form:

$$\dot{\psi} = \psi_{\eta\eta} - s\delta_U \sin\psi \cos\psi + sf\delta \sin(\psi + \alpha_t)\cos(\psi + \alpha_t)\,, \quad (1)$$

where ψ is the director rotation angle (counted counterclockwise),

$\tau = t/\tau_0$, $\eta = \pi y/L$, $\delta_U = U^2/U_{th}^2$, $\delta = S/S_{th}$, $\tau_0 = \gamma_1 L^2/\pi^2 K$, $f = \Delta\varepsilon_{eff}/\Delta\varepsilon$, U_{th} is Freedericksz transition threshold in low-frequency field, S_{th} is the threshold energy flux of the LIFT in homeotropic NLC, $\Delta\varepsilon$ is optical anisotropy, K is the Frank elastic constant, γ_1 is viscosity coefficient, and parameter s is +1 and -1 for homeotropic and planer NLCs, respectively. For transparent NLC, obviously, $f = 1$; for the doped ones we assume $f = f_0(\sin^2\beta - \sin^2\beta_c)$. Restricting our consideration to the first harmonic of the director deformation $\psi = \psi_m \sin(\pi y/L)$ we find

$$\dot{\psi}_m = -\psi_m - s\delta_U J_1(\psi_m) + \nu f_0 \delta[sV_{11}(1/2 - \sin^2\beta_c) - V_{12}/4] + (1-\nu)s\delta V_{11}, \quad (2)$$

where $V_{ij}(\psi_m, \beta) = J_i(2j\psi_m)\cos 2j\beta + \mathbf{E}_j(2j\psi_m)\sin 2j\beta$; ν=0 and 1 for transparent and dye-doped NLCs, respectively; and J_i and $\mathbf{E}_j$ are the Bessel and Weber functions, respectively.

With the relation $N = S_{NL}/2\pi$ between N and the nonlinear phase shift S_{NL} [10] we finally arrive at

$$N = -\frac{s\Delta\varepsilon\varepsilon_\perp^{1/2}}{8\lambda\cos\alpha_t}[(1+s) + (1-s)(\varepsilon_\parallel/\varepsilon_\perp)^{3/2}](V_{01}(\psi_m^C, \beta) - V_{01}(\psi_m^P, \beta)), \quad (3)$$

where ψ_m^C and ψ_m^P are the director rotation angles at the beam axis and the beam periphery, respectively.

The calculated dependences are shown in Figures 1-4. For the homeotropically aligned transparent and dye-doped NLCs (Figure 1) the values $\delta = 0.73$ and $\delta f_0 = 1.346$, respectively, were determined from the condition N(0)=9. For the planarly aligned nondoped NLC and the NLC with KD-1, the values $\delta = 0.47$ (at P=70 mW) and $\delta f_0 = 0.67$, respectively, were determined from the N_{max} values at normal incidence. For the planarly aligned NLC with DEANAB, the results obtained for $\delta f_0 = 0.74$ are presented. As is seen from Figures 1-4 the calculation provides a fair description of the experimental features.

For transparent planar NLC, the calculations showed that the voltage U_{max}, corresponding to the maximum number of the aberrational rings, increases by the factor of 1.15 at decreasing incidence angle α

from 50° to 20°, which is in qualitative agreement with experiment.

Consider the results for NLC with DEANAB at α>0, (where the nonlinearity changes its sign) in more detail. The corresponding solutions $\psi_m(\delta_U)$ of Eq. (2) are shown in Figure 4b: curves AB and DCE are obtained at simultaneous action of the light and low-frequency field ($\delta f_0 = 0.74$), while curve FGH corresponds to the absence of light field (that is, describes the director field at the beam periphery and at the beam axis prior to NLC illumination). Here, if the parameter δ_U is less than the value $\delta_R = 1.015$, equal to the abscissa of point R at which curves CE and GH intersects, then, upon starting the illumination, the transition from the curve FGH to curve AB occurs. If $\delta_U > \delta_R$, the transition to CD curve takes place. Therefore, at $\delta_U = \delta_R$ the sharp change in ψ_m^C and hence in N comes about accompanied by changing the self-action sign (resulting from increasing the angle γ).

Since curve AB in Figure 4b corresponds to the stable states of the director field, then in the different experimental situation - at the smooth increase of the voltage U and uninterrupted NLC illumination with the light beam - the transition from curve AB to curve CD should not take place. However, our additional experiments showed that this transition does take place and occurs at practically the same value of U as does the transition observed with the previously described technique. In a similar way, the reverse transition is observed at smoothly decreasing U. Here, at sufficiently low temperature $t = 11^{\circ}C$ ($\alpha = 45^{\circ}C$ and $P = 4$ mW), in a rather narrow range $\Delta U \approx 0.05$ B the bistability was observed.

The observed instability of the AB branch seems to be related to the spatial inhomogeneity of the light beam leading to an additional elastic force. This force is arisen due to the fact that at α>0 and smoothly increasing U after starting the NLC illumination, the sense of the director rotation is different at the beam axis (clockwise) and at the beam periphery (counterclockwise). The role of the beam width is confirmed by [11], where in planar NLC with the specially produced director pretilt, doped with anthraquinone dye, the formation of 180° walls was observed at increasing the low-frequency voltage in the case that the NLC was illuminated with the wide light beam. Therefore one should expect the much wider bistability range in the wider beams.

CONCLUSIONS

Thus, we found that the suppression by an low-frequency electric field of the director reorientation in the homeotropically aligned NLC doped with diazodye is more efficient as compared with the transparent matrix.

In planar samples the dependence of the director rotation angle on the external low-frequency field is nonmonotonic, its appearance depending upon the sign and the value of the angle of the light wave incidence onto NLC. In NLC doped with monoazodye one can observe the change of the director reorientation sign from the positive to negative one.

The calculations of the NLC director reorientation at simultaneous action of light and low-frequency fields are performed. Their results are in fair agreement with the main experimental dependences.

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