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Light-induced first-order orientational transitions in a nematic liquid crystal in the presence of an ordinary wave

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Abstract. **The effects of light-induced reorientation of the director of a nematic liquid crystal doped with dendrimer are investigated experimentally. The influence of light polarisation on the first-order orientational transition that occurs with changing the light beam power is studied. An irreversible first-order orientational transition when changing the polarisation of light is discovered and examined. A theoretical description of the observed effects is presented.**

Keywords: *nematic liquid crystals, dendrimers, light-induced reorientation, orientational transitions, optical bistability.*

1. Introduction

Nematic liquid crystals (NLCs) consist of rod-shaped molecules, mainly oriented in some direction determined by the unit vector, i.e., the director $\boldsymbol{n}$. The NLC director is very sensitive to external fields [1, 2]; its reorientation (Freedericksz transition) is at the heart of numerous applications of liquid crystals.

If the unperturbed director $\boldsymbol{n}_0$ is perpendicular to the field (in the case of positive anisotropy $\Delta\varepsilon$ or $\Delta\mu$) or parallel to it (in the case of negative anisotropy), the reorientation of the director has a threshold character. For magnetic and low-frequency electric fields the Freedericksz transition is a second-order orientational transition, i.e., the angle ψ of rotation of the director is a continuous function of the external field. The exception is the case when the electric field is parallel to a liquid crystal layer: in this situation the Freedericksz transition can be a first-order transition (angle ψ changes abruptly) [3, 4].

A light-induced Freedericksz transition in a transparent NLC [5–7] under the action of linearly polarised light is also a second-order transition. Since at optical frequencies $\Delta\varepsilon > 0$, the threshold transition occurs in the case of mutually perpendicular orientation of the light field $\boldsymbol{E}$ and the unperturbed director $\boldsymbol{n}_0$, which corresponds to normal incidence of light on a homeotropically oriented NLC.

The physical mechanism of optical orientation in transparent NLCs is associated with the action of light on induced dipoles; torque applied to the unit volume of the NLC has the form

$$\boldsymbol{\Gamma}_{\rm tr} = \frac{\Delta\varepsilon}{4\pi}(\boldsymbol{nE})[\boldsymbol{n}\times\boldsymbol{E}]. \quad (1)$$

In absorbing NLCs (usually a mixture of a transparent nematic matrix and a dye), another mechanism of optical orientation comes to the fore, associated with the change in intermolecular forces under excitation of the molecules [8]. Torque is then determined by the relation

$$\boldsymbol{\Gamma}_{\rm abs} = \eta\boldsymbol{\Gamma}_{\rm tr}, \quad (2)$$

where η is the enhancement factor of the orienting effect of light (η reaches values of $\sim10^2$ for the NLC absorption coefficient of $\sim10^2$ cm^{-1}).

Optical orientation caused by a change in intermolecular forces was observed in liquid crystal systems doped with dyes of different chemical structures [9–12]. In this case, the enhancement factor can be either positive (the director $\boldsymbol{n}$ is rotated parallel to the light field $\boldsymbol{E}$, as in the case of transparent NLCs) or negative ($\boldsymbol{n}$ rotates perpendicular to $\boldsymbol{E}$).

The strongest orientational effect was observed for a NLC doped with high-molecular azo compounds (i.e., compounds containing an azo-bridge N=N) [12]. Azo compounds are characterised by high orientational sensitivity to light, which is manifested in various media – liquids [13, 14], polymers [15–17] and Langmuir–Blodgett films [18]. An important feature of an azo molecule (azo fragment) is the presence of two (trans- and cis-) isomers; in this case, when light is absorbed, the conformational state of the molecule can be changed. The concentration ratio of the isomers in a nematic matrix in the field of an extraordinary wave will depend on the angle Ψ between the light field $\boldsymbol{E}$ and the director $\boldsymbol{n}$. This is explained as follows: due to a more elongated shape, a trans-isomer has a larger order parameter (the degree of ordering of the molecules with respect to the director). Therefore, the probability of its excitation depends more strongly on the angle Ψ than the probability of excitation of a cis-isomer. As a result, the proportion of cis-isomers will decrease with increasing Ψ. Because the isomers induce, in a nematic matrix, torques of opposite signs (trans-isomers cause the rotation of the director $\boldsymbol{n}$ perpendicular to the field

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E, and cis-isomers – parallel to ***E***), the enhancement factor η also depends on the angle Ψ (and may even change the sign) [19–21].

The dependence $\eta(\Psi)$ made it possible to realise first-order orientational transition in a NLC under the action of an extraordinary light wave [22–24]. Such a transition accompanied by bistability of the director was observed in NLCs doped with carbosilane dendrimers containing terminal azo fragments at normal incidence of light on a planar-oriented NLC ($\boldsymbol{n}_0$ was parallel to the walls of the cell). The enhancement factor η was negative and increased in absolute value with increasing angle Ψ (coincident with ψ in this geometry), which provided an additional positive feedback between the director rotation and the magnitude of torque.

The authors of [20, 21, 25] found that the enhancement factor η for a NLC doped with azo compounds depends on the polarisation of light. In the present work we study both experimentally and theoretically the orientational transitions in a NLC doped with dendrimer upon its exposure to a light beam of different polarisation.

2. Experimental conditions

The studies were performed at room temperature by using a nematic liquid crystal LCM-1277 (NIOPIK). We used a planar-oriented cell of thickness L = 100 mm. The inner walls of the glass substrates of the liquid-crystalline cell were coated with a conductive ITO layer.

The molecular structure of second generation (G2) carbosilane azo dendrimer is shown in Fig. 1. The weight concentration of dendrimer in the nematic matrix was 0.15%. The measured polarisation absorption spectra of the cell showed that the sample had a substantial dichroism. The absorption coefficients of the extraordinary and ordinary waves were as follows: $\alpha_e = 20$ cm^{-1}, $\alpha_o = 10$ cm^{-1} (λ = 473 nm).

Light-induced orientational transitions in the NLC were studied by the method of the aberrational self-action of a light beam [26, 27]. The orienting action of the light beam leads to deformation of the director field and to a change in the refractive index of the extraordinary wave. The front of the light wave passing through the liquid crystal layer is distorted, and a system of concentric rings is observed in the far zone as a result of diffraction. The number of aberration rings N is related with a change in the refractive index Δn by the expression

$$|\Delta n| = \lambda N/L, \tag{3}$$

where λ is the wavelength of light. The sign of self-action (self-focusing or defocusing) was determined by the transformation of the aberration pattern when the cell was quickly shifted perpendicular to the axis of the beam [21].

Figure 2a shows the experimental setup. A linearly polarised light beam from a 473-nm solid-state laser (LCS-DTL-364, Laser Export) (*1*) was focused by a lens (*3*) with focal length f = 18 cm and was incident normally on the NLC (*4*). The liquid crystal layer was located vertically (in the XZ plane), the unperturbed director $\boldsymbol{n}_0$ lied in the horizontal plane (parallel to the X axis). Under the action of light the director was rotated in the XY plane by the angle ψ (Fig. 2b). The plane of polarisation of the light beam was rotated by the angle φ with the help of a double Fresnel rhomb (*2*). In this case, ordinary and extraordinary waves were excited in the NLC [φ = 0 corresponds to the extraordinary wave (horizontal polarisation), and φ = 90° – the ordinary wave (vertical polarisation)]. The aberration pattern was observed on the screen (*5*).

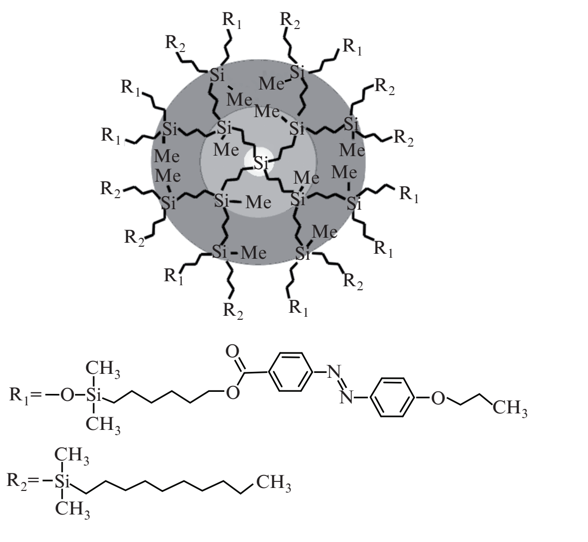

Figure 1. Chemical formula of the second generation (G2) carbosilane dendrimer with statistically distributed azo-benzene (R_1) and aliphatic (R_2) terminal fragments.

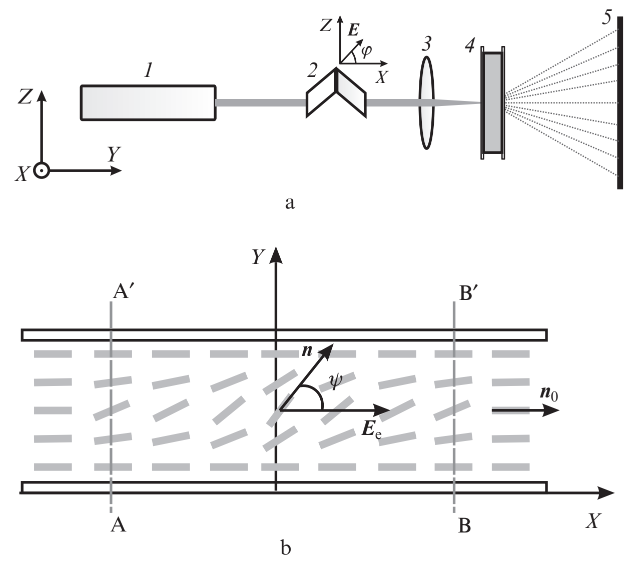

Figure 2. (a) Experimental setup [(*1*) solid-state laser (λ = 473 nm); (*2*) double Fresnel rhomb; (*3*) lens; (*4*) cell with the NLC; (*5*) screen] and (b) reorientation of the director ***n*** under the action of the light field ($\boldsymbol{n}_0$ is the unperturbed director; $\boldsymbol{E}_e$ is the field of the extraordinary wave; ψ is the angle of rotation of the director; lines AA′ and BB′ are the conventional boundaries of the beam).

3. Results of the experiment

In all the experiments regardless of the polarisation of light, we observed self-defocusing, i.e., the director was rotated from the direction of the light field. Figure 3 shows the dependences of the number of aberration rings N and the modulus of the average light-induced refractive index $|\Delta n|$ on the power of the extraordinary wave $P_e = P_0\cos^2\varphi$ (P_0 is the power of the beam incident on the liquid crystal cell), inducing reorientation (see Section 4), at different angles of rotation of the polarisation plane, φ. One can see that at $\varphi = 0$, 20° and 40° first-order orientational transitions are observed, accompanied by bistability with increasing and decreasing power P_e. For example, at $\varphi = 0$ in the range from $P_2 = 18$ mW to $P_1 = 27$ mW, there are two stable positions of the director. At $\varphi = 60°$ the first-order transition transforms into the second-order transition. Figure 3 also shows that with increasing angle φ, the reorientation threshold decreases, i.e., the enhancement factor η increases. In this case, the bistability region shrinks and disappears.

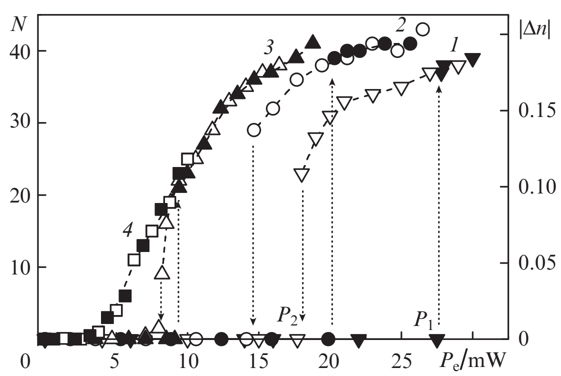

Figure 3. Dependences of the number of aberration rings N and the modulus of light-induced refractive index $|\Delta n|$, averaged over the thickness of the sample, on the power of the extraordinary wave $P_e = P_0\cos 2\varphi$ ($\lambda = 473$ nm) transmitted through the planar-oriented LCM-1277 + 0.15% G2 at normal incidence and angles of rotation of the polarisation plane $\varphi = 0$ (*1*), 20° (*2*), 40° (*3*) and 60° (*4*). Black points correspond to an increase in power P_e, light – to a decrease.

At a fixed value of the light power in the range $P_2 < P < P_1$ (Fig. 3) and with changing the angle φ we found first-order transitions, accompanied by the bistability of the director field (Fig. 4). Thus, when $P = 18.5$ mW and the polarisation plane is rotated from $\varphi = 0$ (the direction of the extraordinary wave) to $\varphi_1 = 35°$, the aberration pattern was not observed (the director field was not deformed). When $\varphi = \varphi_1$, 25 rings were formed. A further increase in φ leads to a monotonic decrease in N (suppression of reorientation). With decreasing angle φ from φ_1 to zero the number of rings increased to 35, i.e., the reverse transition was absent. Thus, in the range $0 < \varphi < 35°$ there are two stable states of the field director. Increasing the beam power led to a reduction of the region of the director field bistability (decrease in φ_1) and to an increase in the number of aberration rings that arise during the transition.

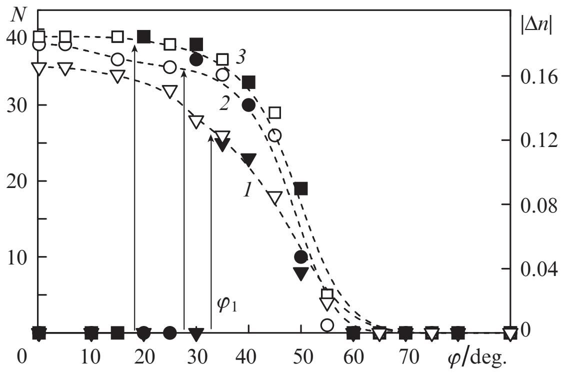

Figure 4. Dependences of the number of aberration rings N and the modulus of light-induced refractive index $|\Delta n|$, averaged over the thickness of the sample, on angle of rotation of the polarisation plane φ at powers of the light ($\lambda = 473$ nm) beam $P = 18.5$ (*1*), 20 (*2*) and 23 mW (*3*), incident on the planar-oriented LCM-1277 + 0.15% G2. Black points correspond to an increase in angle φ, light – to a decrease.

4. Discussion

Consider the orienting effect of superposition of the fields of the extraordinary ($\boldsymbol{E}_e$) and ordinary ($\boldsymbol{E}_o$) waves on the planar-oriented NLC. Field intensities of these waves can be written as

$$\boldsymbol{E}_e = \boldsymbol{i}A_e\exp[\mathrm{i}(\boldsymbol{k}_e\boldsymbol{r} - \omega t)] + \text{c. c.}, \tag{4}$$

$$\boldsymbol{E}_o = \boldsymbol{k}A_o\exp[\mathrm{i}(\boldsymbol{k}_o\boldsymbol{r} - \omega t)] + \text{c. c.}, \tag{5}$$

where A_e, A_o, $\boldsymbol{k}_e$, $\boldsymbol{k}_o$ are the amplitudes and wave vectors of the extraordinary and ordinary waves; ω is the frequency of the light field. Using (1) and (2), taking into account the mutual perpendicularity of the director $\boldsymbol{n} = \boldsymbol{i}\cos\psi + \boldsymbol{j}\sin\psi$ and the field $\boldsymbol{E}_o$, and neglecting the terms rapidly oscillating in time, we obtain an expression for the total torque:

$$\boldsymbol{\Gamma}_{\text{opt}} = \frac{\Delta\tilde{\varepsilon}}{8\pi}\boldsymbol{k}A_e^2\sin\psi\cos\psi$$

$$+\frac{\Delta\tilde{\varepsilon}}{8\pi}(\boldsymbol{i}\sin\psi - \boldsymbol{j}\cos\psi)A_eA_o\cos\psi\cos[(\boldsymbol{k}_e - \boldsymbol{k}_o)\boldsymbol{r}], \tag{6}$$

where $\Delta\tilde{\varepsilon} = \Delta\varepsilon + \Delta\varepsilon_{\text{eff}}$; $\Delta\varepsilon_{\text{eff}} = \eta\Delta\varepsilon$ is the effective anisotropy (value characterising the momentum of the intermolecular forces in absorbing NLCs). The second term in the right-hand side of (6) varies along the Y axis with a characteristic period $l = \lambda/(n_e - n_o) \approx 2.5$ μm, where n_e and n_o are the refractive indices of the extraordinary and ordinary waves. Because the elastic energy of the NLC is proportional to the square of the spatial derivative of the director field (i.e., inversely proportional to the square of the characteristic period of deformation) [1] and $l \ll L$, this term should not lead to a noticeable rotation of the director. Therefore, we assume that the reorientation of the director is due to the torque created by the extraordinary wave.

To describe the orientational transitions in the field of the extraordinary wave in the NLC doped with dendrimers, the authors of [23] suggested using the approximation

$$\Delta\varepsilon_{\text{eff}} = -\Delta\varepsilon_{\text{eff}}^{(0)} - \Delta\varepsilon_{\text{eff}}^{(1)}\sin^2\Psi \tag{7}$$

for effective anisotropy, where $\Delta\varepsilon_{\text{eff}}^{(0)}, \Delta\varepsilon_{\text{eff}}^{(1)}$ are the positive parameters.

When the extraordinary and ordinary waves propagate in the NLC, due to the appearance of the phase shift between them, polarisation of radiation will be elliptical, and will depend on the longitudinal and transverse coordinates. However, this change in polarisation will not affect light absorption by an ensemble of dye molecules (and hence, $\Delta\varepsilon_{\text{eff}}$), since the latter depends only on the direction of the optical axis (director $\boldsymbol{n}$) and the ratio of the intensities of the extraordinary and ordinary waves. Therefore, to generalise relation (7) to the case under study, it is sufficient to express the angle between the linearly polarised field $\boldsymbol{E}$ at the entrance to the crystal and the director $\boldsymbol{n}$ through the angle ψ of the director reorientation and through the angle φ of the polarisation plane. As a result, we obtain

$$\Delta\tilde{\varepsilon} = \Delta\varepsilon^{(0)}\left(1 + m\sin^2\psi + m\sin^2\varphi\cos^2\psi\right), \quad (8)$$

where $\Delta\varepsilon^{(0)} = \Delta\varepsilon - \Delta\varepsilon^{(0)}_{\rm eff}$; $m = -\Delta\varepsilon^{(1)}_{\rm eff}/\Delta\varepsilon^{(0)}$ is the parameter characterising an additional feedback between the angle Ψ and the optical torque.

The equation for the temporal change in the director field $\boldsymbol{n}(\boldsymbol{r}, t)$ can be obtained from the torque balance condition [1]. The director of the NLC is subjected to the viscous torques $\boldsymbol{\Gamma}_{\rm visc} = -\boldsymbol{k}\gamma_1\partial\psi/\partial t$ (γ_1 is the coefficient of viscosity), the elastic torque $\boldsymbol{\Gamma}_{\rm elast} = \boldsymbol{k}K\partial^2\psi/\partial y^2$ (K is the Frank elastic constant), and the optical torque $\boldsymbol{\Gamma}_{\rm opt} = [\Delta\tilde{\varepsilon}/(8\pi)]\boldsymbol{k}A_{\rm e}^2\sin\psi\cos\psi$. Equating to zero their sum, we obtain

$$\frac{\partial\psi}{\partial\tau} = \frac{\partial^2\psi}{\partial\zeta^2} + \delta_{\rm e}(1 + m\sin^2\psi + m\sin^2\varphi\cos^2\psi)\sin\psi\cos\psi, \quad (9)$$

where $\zeta = \pi y/L$ is the dimensionless coordinate; $\tau = t/\tau_0$ is the dimensionless time; $\tau_0 = \gamma_1 L^2/(\pi^2 K)$; $\delta_{\rm e} = |\Delta\varepsilon^{(0)}||A_{\rm e}|^2 L^2 \times (8\pi^3 K)^{-1}$is the dimensionless intensity of the extraordinary wave.

We approximate the distribution of the field director over the thickness of the crystal by the first harmonic

$$\psi(\zeta,\tau) = \psi_{\rm m}(\tau)\sin\zeta, \quad (10)$$

where $\psi_{\rm m}$ is the angle of rotation of the director in the middle of the crystal ($y = L/2$). Substituting (10) into (9), multiplying by $\sin\zeta$ and integrating in the range $0 \leqslant \zeta \leqslant \pi$, we obtain

$$\frac{{\rm d}\psi_{\rm m}}{{\rm d}\tau} = -\psi_{\rm m} + \delta_{\rm e}\left(A + B\sin^2\varphi\right), \quad (11)$$

where

$$A = J_1(2\psi_{\rm m}) + \frac{1}{2}mJ_1(2\psi_{\rm m}) - \frac{1}{4}mJ_1(4\psi_{\rm m}),$$

$$B = \frac{1}{2}mJ_1(2\psi_{\rm m}) + \frac{1}{4}mJ_1(4\psi_{\rm m}).$$

The trivial solution $\psi_{\rm m} = 0$ of equation (11) is stable at $\delta_{\rm e} < \delta_{\rm e,1}$, where

$$\delta_{\rm e,1} = (1 + m\sin^2\varphi)^{-1}. \quad (12)$$

The value of $\delta_{\rm e,1}$ corresponds to the threshold of the direct transition (for example, at $\varphi = 0$ – power P_1, see Fig. 3). Nontrivial stationary solutions of equation (11) at different angles φ are shown in Fig. 5. These solutions are stable at $\partial\delta_{\rm e}(\psi_{\rm m})/\partial\psi_{\rm m} > 0$ and unstable at $\partial\delta_{\rm e}(\psi_{\rm m})/\partial\psi_{\rm m} < 0$.

At $\varphi = 0$ [Fig. 5, curve (*1*)] there is a jump-like inverse transition at $\delta_{\rm e,2} < \delta_{\rm e,1}$ (at point A). Thus, in the region $\delta_{\rm e,2} < \delta < \delta_{\rm e,1}$ there exists bistability of the director field. The value of the parameter $m = 3.2$ used in the calculations corresponds to the experimental value of the relative width of the bistability region $\Delta = (P_1 - P_2)/P_1 = 0.38$ [Fig. 3, curve (*1*)].

One can see from Fig. 5 that an increase in φ leads to a decrease in the thresholds, to a reduction of the bistability region [curves (*1 – 3*)] and to a change in type of transition: at $\varphi = 60°$ [curve (*4*)] the second-order transition takes place.

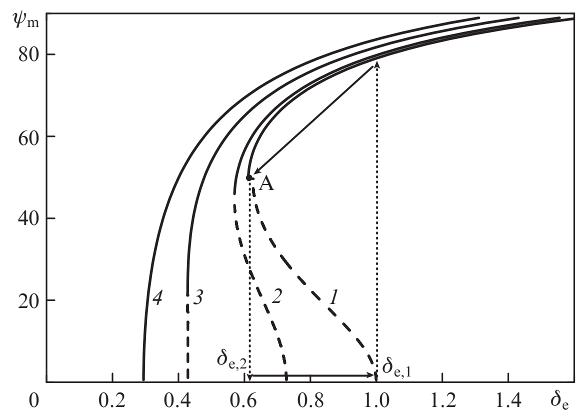

Figure 5. Theoretical dependences of the angle of rotation of the director $\psi_{\rm m}$ on the dimensionless intensity of the extraordinary wave $\delta_{\rm e}$ at angles of rotation of the polarisation plane $\varphi = 0$ (*1*), 20° (*2*), 40° (*3*) and 60° (*4*). Solid curves show the stable solutions, dashed – unstable.

Thus, the theoretical dependences are in qualitative agreement with experiment (Fig. 3).

Consider now the orientational transitions occurring at a fixed power of the light beam and under rotation of the polarisation plane. From equation (11), in the steady-state we obtain

$$\varphi_{\pm} = \frac{1}{2}\arccos\left(\frac{A \pm \sqrt{(A + B)^2 - 4B\psi_{\rm m}/\delta}}{B}\right), \quad (13)$$

where $\delta = \delta_{\rm e}/\cos^2\varphi$ is the dimensionless intensity of the light wave. The solution φ_- is stable at $\partial\varphi_-/\partial\psi_{\rm m} < 0$ and unstable at $\partial\varphi_-/\partial\psi_{\rm m} > 0$. The solution φ_+, for which $\partial\varphi_+/\partial\psi_{\rm m} < 0$ in the interval $0 < \psi_{\rm m} < \pi/2$, is always unstable.

Solutions $\psi_{\rm m}(\xi)$ ($\xi = \sin^2\varphi$) of equation (13) for different values of the dimensionless intensity in the interval $0.63 < \delta < 1$, where there is bistability at $\varphi = 0$ [Fig. 5, curve (*1*)], are shown in Fig. 6 . One can see that depending on the value of δ different regimes of light-induced reorientation are possible.

At $0.75 \leqslant \delta < 1$ [Fig. 6, curves (*1–3*)] an increase in ξ leads to the first-order orientational transition into the perturbed state (in the range $\xi_1 < \xi < \xi_2$ the trivial solution is unstable, and so the angle of reorientation, $\psi_{\rm m}$, changes abruptly when $\xi = \xi_1$). When ξ further increases, deformation of the director field is suppressed by reducing the extraordinary component of the light field. With a subsequent decrease in ξ, the angle $\psi_{\rm m}$ increases monotonically; the transition into the unperturbed state does not take place. Thus, the theoretical model is consistent with the experimental results (see Fig. 4). In a narrow region ($0.73 < \delta < 0.75$) the transition into the unperturbed state with increasing ξ and the corresponding reverse transition become abrupt and occur at different values of ξ. The possibility of observing such a regime requires a separate study.

In the range $0.63 < \delta < 0.73$ the trivial solution is stable at any ξ. At the same time there is a stable nontrivial solution [Fig. 6, curves (*4–5*)]. Transition into this state is possible, for example, by additionally subjecting the NLC to a destabilising low-frequency field, which has been verified experimentally.

The results obtained in this paper indicate the possibility of controlling the NLC state and parameters of optical bistability by changing the concentrations of the isomers of the dye

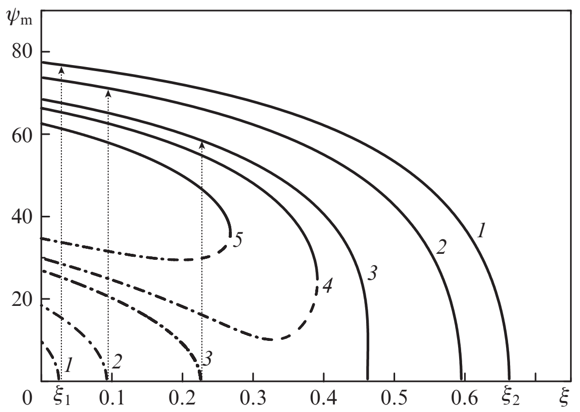

Figure 6. Theoretical dependences of the angle of rotation of the director ψ_m on the parameter $\xi = \sin^2\varphi$ at the dimensionless intensity $\delta = 0.95$ (*1*), 0.85 (*2*), 0.75 (*3*), 0.72 (*4*) and 0.65 (*5*). Solid curves show the stable solutions $\sin^2\varphi_-$, dashed – unstable solutions $\sin^2\varphi_-$, dot-and-dash – unstable solutions $\sin^2\varphi_+$.

molecule swith the help of radiation of ordinary polarisation. This radiation, in principle, can differ from the radiation of extraordinary polarisation that rotates the director of the NLC by the wavelength and direction of propagation, which may be useful for the implementation of schemes of optical switching and optical modulators.

5. Conclusions

We have studied experimentally and theoretically the orientational transitions in nematic liquid crystals doped with azo dendrimer. It is established that the rotation of the polarisation plane of light transforms the first-order transition, which occurs with changing the light wave intensity and is accompanied by bistability of the director of the NLC, into the second-order transition. We have discovered and investigated an irreversible first-order orientational transition when the angle of rotation of the polarisation plane changes and the power of the light beam remains constant. We have constructed a theoretical model of the interaction of the NLC with the superposition of the extraordinary and ordinary waves.

The results obtained indicate the possibility of controlling the type of the orientational transition and optical bistability of NLCs by using an ordinary wave.

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