# Light- and electric-field-induced first-order orientation transitions in a dendrimer-doped nematic liquid crystal

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Interaction of light and ac electric fields with a nematic liquid crystal (NLC) doped with nanosized second-generation carbosilane codendrimers containing terminal azobenzene fragments has been studied. A first-order Freedericksz transition in the linearly polarized light, accompanied by an intrinsic bistability in a wide region, was observed. An additional ac electric field decreases the light-induced Freedericksz transition threshold and narrows the bistability region. Light illumination transforms the second-order electric-field-induced Freedericksz transition to a first-order one. The width of the bistability region increases with the light wave intensity. The theory of the interaction of light and ac electric fields with the dendrimer-doped NLCs is developed taking into account an additional (with respect to the undoped nematic host) dependence of the optical torque on the angle between the director and the light field.

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## I. INTRODUCTION

The supramolecular structure of nematic liquid crystals (NLCs) is very sensitive to dc and ac electric and magnetic fields, as well as to the light field.

Magnetic- and electric-field-induced director deformations were first observed by Freedericksz and his colleagues as early as in the 1930s [1–3]. Nowadays, the phenomenon of the director reorientation under the action of a low-frequency electric field is the basis for most applications of NLCs.

When the unperturbed director $\mathbf{n}_0$ is normal to a field [in the case that the dielectric ($\Delta\varepsilon$) or magnetic ($\Delta\mu$) anisotropies are positive] or parallel to it ($\Delta\varepsilon<0$ or $\Delta\mu<0$), the director reorientation (Freedericksz transition) is of a threshold type [1–5].

The Freedericksz transition in the magnetic field $\mathbf{H}$ is always second order [4], its order parameter [6] being the angle $\psi$ of the director rotation. The symmetric phase corresponds to the uniform director field ($\psi=0$); the nonsymmetric one, to the deformed director field ($\psi\neq 0$). For the second-order transition, the angle $\psi$ is a continuous function of the field (in contrast to the first-order transition, in which case this angle is discontinuous at the threshold field).

The situation is more complex for electric field. The physical reason for this is the large dielectric anisotropy $\Delta\varepsilon\sim 1$ (six orders of magnitude higher than the magnetic anisotropy), which leads to reverse action of the director deformation on the electric field. Theoretical analysis [7] showed that in the case $\Delta\varepsilon>0$ the Freedericksz transition in the electric field $\mathbf{G}$ normal to the liquid-crystal layer (splay deformation in planar NLC cell) is always second order. At the same time, the transitions under electric field parallel to the layer (twist deformation in planar NLC cell and bend deformation in homeotropic NLC cell) can be first order at a sufficiently large dielectric and elastic (for bend) anisotropy [8–10].

Thus the first-order electric-field-induced Freedericksz transition can only occur when the electric field is parallel to aligning substrates. These transitions were observed in [9].

In [9,10], six possible geometries of the threshold director reorientation under simultaneously applied electric $\mathbf{G}$ and magnetic $\mathbf{H}$ fields were theoretically considered for NLCs with positive $\Delta\varepsilon$ and $\Delta\mu$ (one field induces the transition, the other tends to support the homogeneous director field $\mathbf{n}_0$). It was found that in some geometries the presence of stabilizing field can transform the second-order transition to the first-order, increasing at the same time the transition threshold. This is, e.g., the case for the electric-field-induced bend in a homeotropic NLC cell in a sufficiently strong magnetic field.

The interaction of the light field $\mathbf{E}$ with NLCs is even more complex. In this case, two coupled normal waves (the extraordinary and the ordinary ones) can be excited in an NLC, which greatly affects the light polarization inside NLC. In addition, the director field transformation depends on the light propagation direction.

Consider first the implementation of the geometry $\mathbf{E}\perp\mathbf{n}_0$ in the simplest case of the normal incidence of linearly polarized light on NLCs. In the homeotropic NLC cell, the second-order Freedericksz transition was observed [11–14]. It was predicted in [15,16] that the light-induced Freedericksz transition in this geometry can be first order at certain ratios of the dielectric tensor components (at light frequency) and elastic Frank constants. As in the case of ac electric field, the reason for the first-order transition is the reverse action of the director field deformation on the light field. However, as far as we know, this phenomenon was not observed experimentally. At the propagation of the ordinary wave in planar NLC, the director reorientation is practically suppressed (Maugin mode) [12,17,18].

Geometry $\mathbf{E}\perp\mathbf{n}_0$ is also possible in some other cases of light and NLC interaction. First, we note the normal inci-

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dence of a circularly (or elliptically) polarized light wave on the homeotropic NLC cell [19–25]. In this case, the interacting extraordinary and ordinary waves are excited, which strongly affects light polarization inside NLC. The first-order light-induced Freedericksz transitions and the bistability of the director polar angle $\theta$ (the angle between the director $\mathbf{n}$ and the normal to the cell plane, i.e., $\mathbf{n}_0$) were observed in [20,22]. One value of the angle $\theta$ corresponds to rising light intensity, the other sets in at falling intensity (i.e., the hysteresis loop is observed). The deformed state of the director field is however not stationary in this case: the director precession (change in the azimuthal angle) occurs due to the light angular momentum transfer to NLC. At the oblique incidence of the light wave polarized normally to the plane of incidence on a homeotropic NLC cell (the incidence of ordinary wave), periodic and stochastic director oscillations are generated (due to the excitation of extraordinary wave by the deformed director field); no bistability was observed [26–30].

Addition of an ac field to the light wave can give rise to the first-order orientation transitions even in the case of linearly polarized light (and excitation of single extraordinary wave in the NLC bulk). The first-order Freedericksz transitions in homeotropic NLC cells under normally incident linearly polarized light wave and stabilizing (magnetic or electric) field were studied theoretically in [31–34] and observed experimentally in [35–37]. The first-order electric-field-induced transitions were observed in the illuminated homeotropic [36] and planar [38] NLC cells (the theory of the latter effect was considered in [39]).

The relative width of the optical bistability region (hysteresis loop), defined as $\Delta_P=(P_{\mathrm{th},1}-P_{\mathrm{th},2})/P_{\mathrm{th},1}$ (where $P_{\mathrm{th},1}$ and $P_{\mathrm{th},2}$ are the transition thresholds at rising and falling light-beam power) was small. In [35], this width was about 0.03 (the hysteresis loop itself was smoothly varying); in [36,37], it was $\Delta_P=0.015$. In [36,38], the relative width of the bistability region observed at changing the electric field $\Delta_U=(U_{\mathrm{th},1}-U_{\mathrm{th},2})/U_{\mathrm{th},1}$ ($U_{\mathrm{th},1}$ and $U_{\mathrm{th},2}$ are the transition thresholds at rising and falling voltage) in illuminated homeotropic and planar NLC cells was 0.01 [36] and 0.05 [38], respectively. In the latter case, the discontinuous change in the measured phase shift was only observed at increasing voltage.

There are also a number of other experimental realizations of the first-order transitions and bistability in NLCs involving ac and optical fields. Among these are the electric-field-induced transition in thin homeotropic NLC cell with negative dielectric anisotropy near the nematic-smectic transition due to contribution of the anchoring forces [40], the transition in the simultaneously applied crossed destabilizing magnetic and electric fields [41], the transition in the field of two light waves propagating in different directions (and having different directions of linear polarization) [42], and the transition at rotation of the magnetic field [43]. First-order orientation transitions in two light waves and in a light beam of finite size were discussed in [44,45], respectively.

In the above consideration, the light-induced director reorientation occurred in transparent cells and was caused by the torque $\mathbf{\Gamma}_{\mathrm{tr}}$ due to light action on induced dipoles. At the same time, the orientational optical effects in NLCs are also possible due to excitation of low- [46–51] and high-molar-mass [52,53] dopants. The corresponding torque $\mathbf{\Gamma}_{\mathrm{abs}}$ [47,52,54–59] can exceed $\mathbf{\Gamma}_{\mathrm{tr}}$ by orders of magnitude. The first-order orientation transitions in dye-doped NLCs were observed in [60] at rotating NLC cell with respect to the light beam of constant power (similarly to [43]) and in [61] at simultaneous action of an obliquely incident linearly polarized laser beam and a uniform electric field on planar NLC cell. In the latter case, the transition was principally related to the difference in the light and ac electric fields directions and the nonuniformity of the transverse structure of the director field.

Thus, despite much interest in first-order Freedericksz transitions in NLCs, a purely optical transition in the field of a single linearly polarized wave was not observed. The reported implementations of the optical first-order transitions required an additional ac or light field, elliptical polarization of light wave, or rotation of an NLC cell with respect to the incident beam.

An approach to realize the optical first-order transition used in this paper is based on the fact that the factor $\eta$ of the enhancement of the light action [defined by relation $\mathbf{\Gamma}_{\mathrm{abs}}=\eta\mathbf{\Gamma}_{\mathrm{tr}}$, where $\mathbf{\Gamma}_{\mathrm{tr}}$ and $\mathbf{\Gamma}_{\mathrm{abs}}$ are the light-induced torques exerted on the undoped (transparent) and the dye-doped nematic host] can depend on the angle $\Psi$ between $\mathbf{n}$ and $\mathbf{E}$. Such dependence was observed experimentally for the dopants containing azobenzene chromophores [51–53,59,60,62–65].

The explanation of this phenomenon [62] is based on the existence of two isomers (*cis-* and *trans-*) of azomolecules and an assumption that these isomers are related to the torques of opposite signs: the trans-isomers give rise to the torque rotating the director away from the light field (negative $\eta$); the cis-isomers, to the torque rotating the director toward the light field (positive $\eta$). Initially, most chromophores are in the trans-state. Under light exposure the isomers are excited and can undergo conformational transitions. Since the order parameters of the isomers in nematic host are different (due to different shapes), the equilibrium concentrations of trans- and cis-isomers should depend on the angle $\Psi$: the concentration of trans-isomers is minimum at $\Psi=0$ and maximum at $\Psi=90°$ and vice versa for cis-isomers. Correspondingly, the enhancement factor $\eta$ should decrease with $\Psi$.

Previous experiments showed that the enhancement factor for azodopants can be either sign inversion (positive at $\Psi=0$ and negative at $\Psi=90°$) or negative irrespective of $\Psi$. In the former case, the normally incident light wave cannot rotate director both in the homeotropic ($\Psi=90°$) and planar ($\Psi=0$) NLC cells. In the latter case, the light-induced threshold transition should occur at the normal incidence of the light wave on the planar cell ($\Psi=0$). A sufficiently rapidly increasing function $|\eta(\Psi)|$ can provide a feedback channel transforming the transition order and leading to the director bistability.

A first-order transition in the linearly polarized wave in planar NLC doped with dendrimers containing azobenzene terminal groups was reported in short communication [66]. In this paper, we present the results of detailed experimental and theoretical study of the behavior of the dendrimer-doped nematic cell in the light and low-frequency electric fields.

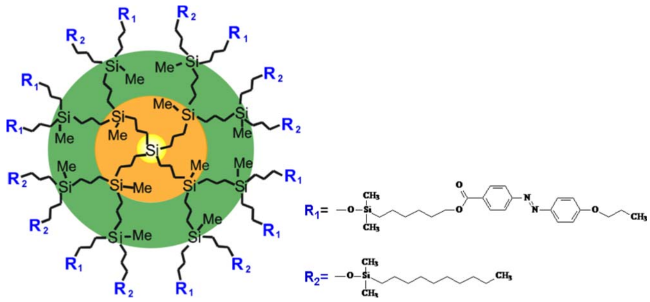

FIG. 1. (Color online) Structural formula of the second-generation carbosilane dendrimer with statistically distributed azobenzene ($R_1$) and aliphatic ($R_2$) terminal fragments. Me is the methyl group $CH_3$.

## II. EXPERIMENT

The substance under study was the nematic host ZhKM-1277 (mixture of biphenils and esters; NIOPIK, Russia) doped with 0.15% of the carbosilane dendrimer of the second generation (G2) with statistically distributed aliphatic and azobenzene terminal fragments (see structural formula in Fig. 1).

Previous studies of NLCs doped with the carbosilane homodendrimers [53,59] showed high values of the enhancement factor of this type of azocompounds and the influence of their structure (the generation number) on the type of the angular dependence $\eta(\Psi)$ (the sign-inversion or the negative). Our preliminary studies showed that enhancement factor due to the dendrimer G2 under study is negative independent of $\Psi$.

Nematic host material exhibits a nematic phase in a wide temperature range (from $-20\ ^\circ C$ to $+60\ ^\circ C$) and has a positive low-frequency dielectric anisotropy ($\Delta\varepsilon=12.1$ at $f=1$ kHz). The liquid-crystal cell (100 $\mu$m thick) was formed by two glass substrates (coated with transparent ITO electrodes) separated by Teflon spacers. Planar alignment of the cell was obtained by polyimide layers deposited on substrates by spin-coating, polymerized at high temperature and rubbed in antiparallel directions. The Freedericksz transition threshold in the ac field was 0.95 V. The absorption spectrum of the cell (Fig. 2) was measured by an MS-122 spectrophotometer (PROSCAN Special Instruments). In the visible range, the absorption of the cell decreases with wavelength.

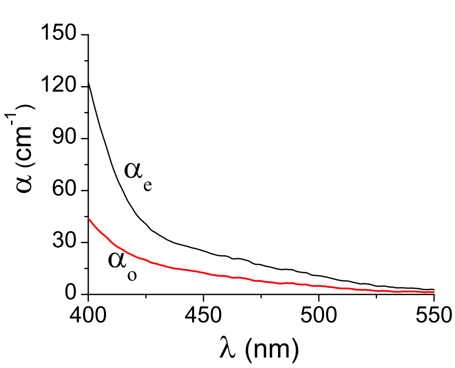

FIG. 2. (Color online) Absorption spectra of the ZhKM-1277 +0.15% G2 planar cell.

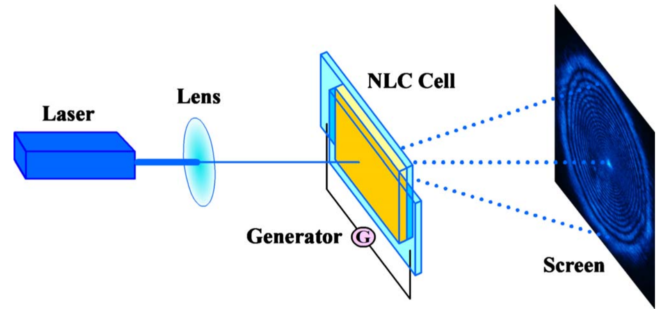


FIG. 3. (Color online) Schematic representation of the experimental setup.

The absorption coefficients of the extraordinary and ordinary waves are $\alpha_e=20\ cm^{-1}$ and $\alpha_o=10\ cm^{-1}$ at $\lambda=473$ nm.

The light-induced director reorientation was studied by the method of the aberrational self-action [67]. The change in the refractive index of the extraordinary wave due to the director deformation introduces a bell-like nonlinear phase shift into the light beam, which results in the formation of the ring-shaped aberrational pattern in the far field. The phase shift at the beam axis equals the number of rings multiplied by $2\pi$; the type of the self-action (self-focusing or self-defocusing) can be easily determined from the pattern intensity transformation upon the transverse cell displacement [63]. The self-focusing corresponds to director rotation to the light field **E**, an increase in the refractive index, and the positive phase shift; the self-defocusing corresponds to an opposite sense of director rotation, a decrease in the index, and the negative phase shift.

Schematic representation of the experimental setup is shown in Fig. 3. The horizontally polarized light beam of a solid-state LCS-DTL-364 laser (Laser-Export Co. Ltd, $\lambda=473$ nm) was focused into the NLC cell by a lens with the focal length of 18 cm. The plane of the liquid-crystal layer was vertical. The angle of the light incidence on the cell could be changed by rotating the cell about the vertical axis. The unperturbed director $\mathbf{n}_0$ was in the horizontal plane. An ac voltage from an MXG-9802A generator (METEX) could be applied to the cell. The aberration ring-shaped pattern was observed on the screen.

## III. RESULTS

At the normal incidence of a light beam on the cell and stepwise increase in the beam power $P$, no aberration pattern was observed up to the threshold value $P_{th,1}=37$ mW, at which the self-defocusing rings start to appear. The number $N$ of rings approached a limiting value of 34 in about 1 min (Fig. 4). Further increase in $P$ did not affect the pattern size significantly. Upon subsequent decrease in the beam power, the ring number $N$ first decreased smoothly and then, at $P_{th,2}=21.5$ mW, the pattern collapsed. The accuracy of the threshold measurements was 0.5 mW; at each point the measurement time was sufficient to assure the equilibrium number of aberration rings. Thus, a pronounced hysteresis of the $N(P)$ dependence is observed, the bistability region width being 15.5 mW. The relative value is $\Delta_P=0.42$, which significantly exceeds that found in [35–38].

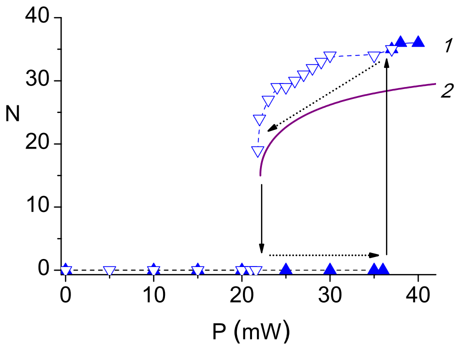


FIG. 4. (Color online) (1) The number $N$ of the self-defocusing rings at increasing (closed triangles) and decreasing (open triangles) power $P$ of the light beam ($\lambda=473$ nm) normally incident on planar ZhKM+0.15% G2 cell. (2) Theoretical dependence.

As the angle of the light beam incidence on the cell increases, the bistability region and transition thresholds decrease (Fig. 5). Thus, the bistability region width is 6 mW ($\Delta_P=0.3$) at $\alpha=20°$. Further increase in the incidence angle up to $\alpha=30°$ suppresses the bistability.

External ac electric field affects the light-induced Freedericksz transition in a similar way (Fig. 6). At the normal light incidence, the ac voltage $U=0.5$ V decreases the thresholds and narrows the bistability region to $P_{th,1}-P_{th,2}=7$ mW ($\Delta_P=0.37$). Further increase in the voltage leads to the smooth dependence $N(P)$ without hysteresis.

Thus, we observed the first-order light-induced Freedericksz transition. Changing the experimental geometry and application of ac field makes the transition second order.

The bistability manifests itself also in the dependence of the aberration ring number on the ac field. At a constant light beam power $P=22.5$ mW (this value is practically equal to the $P_{th,2}$ threshold of the purely optical transition, see curve 1 in Fig. 4), the dependence $N(U)$ [Fig. 7(a)] is of the threshold type and exhibits a hysteresis loop ($U_{th,1}=0.65$ V, $U_{th,2}=0.44$ V, and $\Delta_U=0.32$). At lower beam power $P=10$ mW, no threshold and hysteresis were observed. On the contrary, an increase in $P$ [Fig. 7(b)] widens the bistability region and decreases the threshold of the reverse transition $U_{th,2}$. So, at

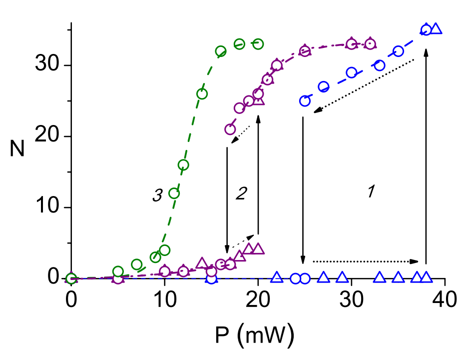


FIG. 5. (Color online) Dependences of the number $N$ of the self-defocusing rings on the power $P$ of the light beam ($\lambda=473$ nm) at various angles $\alpha$ of its incidence on planar ZhKM+0.15% G2 cell: $\alpha=(1)0°,(2)20°,(3)30°$. In dependences (2) and (3), the triangles and circles correspond to increasing and decreasing $P$, respectively.

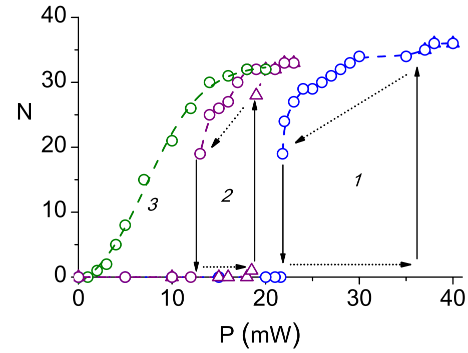


FIG. 6. (Color online) Dependences of the number $N$ of the self-defocusing rings on the power $P$ of the light beam ($\lambda=473$ nm) normally incident on the planar ZhKM+0.15% G2 cell for different ac voltages: $U=(1)0,(2)0.5,(3)0.7$ V. In dependences (1) and (2), the triangles and circles correspond to increasing and decreasing $P$, respectively.

$P=30$ mW, the relative bistability region width increases almost twice and comprises $\Delta_U=0.58$ ($U_{th,1}=0.6$ V and $U_{th,2}=0.25$ V). At $P=32.5$ mW, the threshold voltage $U_{th,2}$ vanishes and the director field does not relax to the homogeneous state even upon complete removal of the voltage.

These results imply that the influence of the light field transforms the second-order electric-field-induced Freedericksz transition into the first-order one.

## IV. DISCUSSION

Let us now discuss the reason for the optical bistability. The torque due to changing the intermolecular forces upon dye excitation is

$$\mathbf{\Gamma}_{abs}=\frac{\Delta\varepsilon_{eff}}{4\pi}(\mathbf{nE})[\mathbf{n}\times\mathbf{E}], \tag{1}$$

where $\Delta\varepsilon_{eff}=\eta\Delta\varepsilon$ is the effective optical anisotropy (the quantity replacing the optical anisotropy $\Delta\varepsilon$ in the expression for the torque

$$\mathbf{\Gamma}_{tr}=\frac{\Delta\varepsilon}{4\pi}(\mathbf{nE})[\mathbf{n}\times\mathbf{E}] \tag{2}$$

exerted on the director of transparent NLCs).

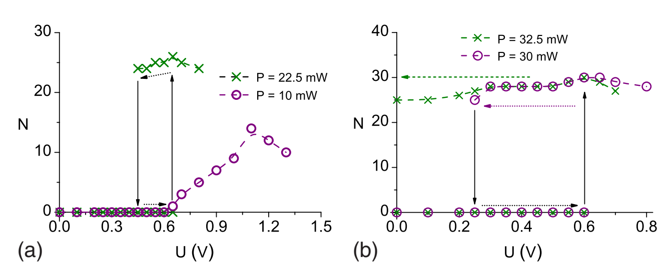


FIG. 7. (Color online) Dependences of the number $N$ of the self-defocusing rings on the applied ac voltage $U$ at the normal incidence of the light beam ($\lambda=473$ nm) on the planar ZhKM+0.15% G2 cell for various beam powers $P$: (a) $P=10$ and 22.5 mW; (b) $P=30$ and 32.5 mW.

If parameter $\Delta\varepsilon_{\text{eff}}$ is independent of the angle between $\mathbf{n}$ and $\mathbf{E}$, the description of the director reorientation is quite similar to the case of the low-frequency electric (with no allowance for the reverse influence of the director deformation) and magnetic fields. Then the light-induced Freedericksz transition is second order.

The situation can be changed if $\Delta\varepsilon_{\text{eff}}$ depends on $\Psi$. Indeed, if the function $\Delta\varepsilon_{\text{eff}}(\Psi)$ is negative and increases in magnitude with $\Psi$ sufficiently rapidly, an additional stable state $\Psi\neq 0$ will exist at the threshold light intensity [determined by the quantity $\Delta\varepsilon_{\text{eff}}(\Psi=0)$]. To put it differently, the director bistability appears and the Freedericksz transition becomes first order.

We represent the function $\Delta\varepsilon_{\text{eff}}(\Psi)$ in a simple form

$$\Delta\varepsilon_{\text{eff}} = -\Delta\varepsilon_{\text{eff}}^{(0)} - \Delta\varepsilon_{\text{eff}}^{(1)}\sin^2\Psi, \tag{3}$$

where $\Delta\varepsilon_{\text{eff}}^{(0)}$ and $\Delta\varepsilon_{\text{eff}}^{(1)}$ are positive parameters.

To justify relation (3) we note, that the rate of chromophore excitation (and with it the number of excited chromophores) is a function of $\cos^2\Psi$. Then the parameter $\Delta\varepsilon_{\text{eff}}$ should also depend on $\cos^2\Psi$ or, equivalently, on $\sin^2\Psi$.

Time evolution of the director field $\mathbf{n}(\mathbf{r},t)$ can be derived from the equilibrium of torques [4,5]

$$\mathbf{\Gamma}_{\text{visc}} + \mathbf{\Gamma}_{\text{elast}} + \mathbf{\Gamma}_{\text{elect}} + \mathbf{\Gamma}_{\text{opt}} = 0, \tag{4}$$

where

$$\mathbf{\Gamma}_{\text{visc}} = -\gamma_1\left[\mathbf{n}\times\frac{d\mathbf{n}}{dt}\right] \tag{5}$$

is the viscous torque ($\gamma_1$ is the viscosity coefficient),

$$\mathbf{\Gamma}_{\text{elast}} = K[\mathbf{n}\times\Delta\mathbf{n}] \tag{6}$$

is the elastic torque in the one-constant approximation ($K$ is the Frank elastic constant),

$$\mathbf{\Gamma}_{\text{elect}} = \frac{\Delta\varepsilon_{\text{low}}}{4\pi}(\mathbf{nG})[\mathbf{n}\times\mathbf{G}] \tag{7}$$

is the torque produced by the low-frequency field $\mathbf{G}$ ($\Delta\varepsilon_{\text{low}}$ is the dielectric anisotropy at the field frequency $\omega_{\text{low}}$), and

$$\mathbf{\Gamma}_{\text{opt}} = \mathbf{\Gamma}_{\text{abs}} + \mathbf{\Gamma}_{\text{tr}} \tag{8}$$

is the torque due to the action of light field, which, with regard to Eqs. (1)–(3), can be written as

$$\mathbf{\Gamma}_{\text{opt}} = \frac{\Delta\varepsilon^{(0)}(1+m\sin^2\Psi)}{4\pi}(\mathbf{nE})[\mathbf{n}\times\mathbf{E}], \tag{9}$$

where $\Delta\varepsilon^{(0)}=\Delta\varepsilon-\Delta\varepsilon_{\text{eff}}^{(0)}$ and $m=-\Delta\varepsilon_{\text{eff}}^{(1)}/\Delta\varepsilon^{(0)}$. The positive (at $\Delta\varepsilon_{\text{eff}}^{(0)}>\Delta\varepsilon$) parameter $m$ characterizes a feedback between the director rotation and the orienting action of light.

Let the $X$ axis of the Cartesian coordinate system be parallel to the NLC cell substrate (and the undeformed director $\mathbf{n}_0$), the $Y$ axis be normal to the substrates, the $Z$ axis be normal to the $XY$ plane (Fig. 8), and the unit vectors be denoted as $\mathbf{i},\mathbf{j},\mathbf{k}$. Then, the deformed director field can be represented in the form

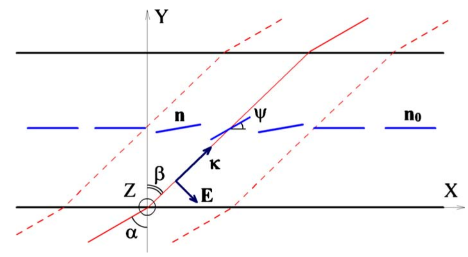


FIG. 8. (Color online) Reorientation of the director $\mathbf{n}$ in the field $\mathbf{E}$ of the light beam: $\mathbf{n}_0$ is the undeformed director, $\alpha$ is the angle of incidence, $\beta$ is the angle of refraction, $\boldsymbol{\kappa}$ is the wave vector, and $\psi$ is the angle of director rotation. Segments indicate the director orientation, solid line is the light beam axis, and the dashed lines are the conventional boundary of the beam.

$$\mathbf{n} = \mathbf{i}\cos\psi + \mathbf{j}\sin\psi, \tag{10}$$

where $\psi(y)$ is the director tilt in the $XY$ plane with respect to the $X$ axis. The low-frequency field is expressed in terms of the amplitude $U_0$ of the voltage applied to the NLC cell

$$\mathbf{G} = \mathbf{j}\frac{U_0\sin\omega_{\text{low}}t}{L}. \tag{11}$$

The light field is

$$\mathbf{E} = \frac{1}{2}\mathbf{e}Ae^{i(\kappa y-\omega t)} + \text{c.c.}, \tag{12}$$

where $A$, $\mathbf{e}=\mathbf{i}\cos\beta-\mathbf{j}\sin\beta$, $\boldsymbol{\kappa}$, $\omega$, and $\beta$ are, respectively, the amplitude, polarization unit vector, wave vector, light frequency, and the angle of refraction of the light wave.

Substituting Eqs. (10)–(12) into Eqs. (5)–(7) and (9) yields

$$\mathbf{\Gamma}_{\text{visc}} = -\mathbf{k}\gamma_1\frac{\partial\psi}{\partial t}, \tag{13}$$

$$\mathbf{\Gamma}_{\text{elast}} = \mathbf{k}K\frac{\partial^2\psi}{\partial y^2}, \tag{14}$$

$$\mathbf{\Gamma}_{\text{elect}} = \mathbf{k}\frac{\Delta\varepsilon_{\text{low}}}{8\pi}\frac{U_0^2}{L^2}\sin\psi\cos\psi, \tag{15}$$

$$\mathbf{\Gamma}_{\text{opt}} = -\mathbf{k}\frac{\Delta\varepsilon^{(0)}[1+m\sin^2(\psi+\beta)]|A|^2}{8\pi}\sin(\psi+\beta)\cos(\psi+\beta). \tag{16}$$

The torque $\mathbf{\Gamma}_{\text{elect}}$ was time averaged assuming that the inverse frequency $\omega_{\text{low}}^{-1}$ of the electric field is much lower than characteristic time of the director rotation. In Eqs. (12) and (16) we neglected the influence of the director field deformation on the light wave polarization and amplitude because the related correction does not change the character of the angular dependence of the $\mathbf{\Gamma}_{\text{opt}}$ torque.

From Eqs. (4) and (13)–(16), we finally obtain the equation for the angle $\psi$ of the director rotation

$$\frac{\partial\psi}{\partial\tau} = \frac{\partial^2\psi}{\partial\eta^2} + \delta_G \sin\psi\cos\psi + \delta[1 + m\sin^2(\psi+\beta)] \times\sin(\psi+\beta)\cos(\psi+\beta), \quad (17)$$

where $\eta=\pi y/L$, $\tau=t/\tau_0$ $(\tau_0=\gamma_1 L^2/\pi^2 K)$, $\delta_G=\Delta\varepsilon_{\rm low}U_0^2/8\pi^3K$, and $\delta=|\Delta\varepsilon^{(0)}||A|^2L^2/8\pi^3K$ are the dimensionless coordinate, time, square of the low-frequency field strength, and the light wave intensity, respectively.

The boundary conditions have the form

$$\psi|_{\eta=0} = \psi|_{\eta=\pi} = 0. \quad (18)$$

To solve Eq. (17), we approximate the spatial dependence of the function $\psi(\eta,\tau)$ by the first harmonic

$$\psi(\eta,\tau) = \psi_m(\tau)\sin\eta, \quad (19)$$

where $\psi_m$ is the director tilt angle at $y=L/2$. Substituting Eq. (19) into Eq. (17), multiplying by $\sin\eta$, and integrating over the segment $0\le\eta\le\pi$, we arrive at

$$\frac{d\psi_{\rm m}}{d\tau} = F(\psi_m,\delta,\delta_G), \quad (20)$$

where

$$F(\psi_m,\delta,\delta_G) = -\psi_m + \delta_G J_1(2\psi_m) + \delta\left\{\left(1+\frac{m}{2}\right)\times[\mathrm{J}_1(2\psi_{\rm m})\cos 2\beta + \mathbf{E}_1(2\psi_{\rm m})\sin 2\beta] - \frac{m}{4}[\mathrm{J}_1(4\psi_{\rm m})\cos 4\beta + \mathbf{E}_1(4\psi_{\rm m})\sin 4\beta]\right\}, \quad (21)$$

$J_1(x)$ and $\mathbf{E}_1(x)$ are the Bessel and Weber functions. The stationary states of the director field obey the equation

$$F(\psi_{\rm m},\delta,\delta_G,m) = 0. \quad (22)$$

Let us consider first the director field transformation at changing the parameter $\delta$ (i.e., the light beam power). For any value of the parameter $m$, Eq. (22) has the trivial solution $\psi_{\rm m}=0$. To analyze its stability, we linearize Eq. (20)

$$\frac{d\psi_m}{d\tau} = (\delta-1)\psi_m. \quad (23)$$

It follows immediately from Eq. (23) that the trivial solution is stable at $\delta<1$ and unstable at $\delta>1$. The nontrivial solutions $\psi_{\rm m}(\delta)$ constructed at $\delta_G=0$ and various values of the parameter $m$ are shown in Fig. 9. Solution stability analysis presented in Appendix A shows that the portions of the curves with the positive slope $(\partial\psi_{\rm m}/\partial\delta>0)$ represent stable solutions (solid-line portions) and those with the negative slope $(\partial\psi_{\rm m}/\partial\delta<0)$ correspond to the unstable solutions (dashed-line portions). At $m>0.8$, there appears a range of $\delta$ in which two stable states of the director (the spatially homogeneous and the deformed) exist.

As the light beam power (quantity $\delta$) increases, an abrupt transition from point $A$ to point $A'$ occurs at $\delta=1$ (see inset in Fig. 9). As the beam power decreases, a reverse transition occurs from $B'$ to $B$. Thus, the light-induced Freedericksz transition becomes first order and the director field bistability

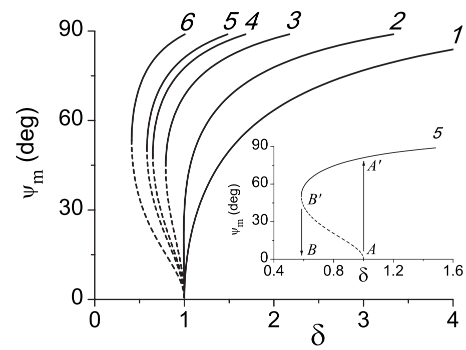


FIG. 9. Dependences of the director deformation angle $\psi_{\rm m}$ on the dimensionless light wave intensity $\delta$ for different values of the m parameter: $m=(1)0,(2)0.8,(3)2,(4)3,(5)3.6,(6)6$. Curve (5) corresponds to the experimental value of the relative bistability region width 0.42. Inset shows curve (5) on an enlarged scale; $AA'$ and $BB'$ show the director switching at rising and falling transitions.

(in the range $\delta_B<\delta<\delta_A$) appears. The experimental relative width of the bistability region $\Delta_P=0.42$ corresponds to $m=3.6$. The threshold beam powers $P_{\rm th,1}=37$ mW and $P_{\rm th,2}=21.5$ mW correspond to $\delta_{\rm th,1}=\delta_A=1$ and $\delta_{\rm th,2}=\delta_B=0.58$.

It is possible to roughly estimate the phenomenological parameters $\Delta\varepsilon_{\rm eff}^{(0)}$ and $\Delta\varepsilon_{\rm eff}^{(1)}$ involved in our model. Taking into account equation $\delta_{\rm th,1}=1$, we can write $P_{\rm th,1}\propto|A|^2_{\rm th,1}=8\pi^3K/|\Delta\varepsilon^{(0)}|L^2$. For the threshold power $P_{\rm th,tr}$ in transparent (homeotropic) NLC cell [67], $P_{\rm th,tr}\propto|A|^2_{\rm th,tr}=8\pi^3K/\Delta\varepsilon L^2$. Hence, $P_{\rm th,tr}/P_{\rm th,1}=(\Delta\varepsilon_{\rm eff}^{(0)}-\Delta\varepsilon)/\Delta\varepsilon$ and $\Delta\varepsilon_{\rm eff}^{(0)}=\Delta\varepsilon(1+P_{\rm th,tr}/P_{\rm th,1})$. For ZhKM-1277 the optical anisotropy is $\Delta\varepsilon=0.6$, the threshold power $P_{\rm th,tr}$ for the 100-$\mu$m NLC cell is approximately 100 mW. Then, with $P_{\rm th,1}=37$ mW, we obtain $\Delta\varepsilon_{\rm eff}^{(0)}\sim 2$ and $\Delta\varepsilon_{\rm eff}^{(1)}=(\Delta\varepsilon_{\rm eff}^{(0)}-\Delta\varepsilon)m\sim 5$.

The dependence of the aberration ring number on the light beam power calculated by formula (B6) (Appendix B) for the director field given by Eq. (22) according to curve 5 in Fig. 9 is shown in Fig. 4 (curve 2). It can be seen that the calculated number of rings is by about 20% less than the experimental one. This difference may be related to the spatial confinement of the beam and light wave decay in the NLC bulk (which was not taken into account in our consideration) as well as with the approximation of the director field by the sinusoid in Eq. (19).

Figure 10 shows the theoretical dependences of the director deformation angle on the intensity of the light wave at different angles of its incidence on the NLC cell. As in experiment, the bistability region width is maximum at the normal light incidence and decreases with rotating the cell (at $\alpha=0°$ and $10°$, $\Delta_P=0.42$ and 0.07, respectively). Calculation shows that the bistability disappears at $\alpha=15°$; in experiment however it is retained at $\alpha=20°$.

As follows from the results presented in Fig. 11, the optical bistability region width decreases at application of electric field. In a sufficiently high ($\delta_G=0.8$) field, the bistability disappears. The calculated bistability region width at $U=0.5$ V (which corresponds to $\delta_G=0.28$) comprises $\Delta_P=0.18$, which is two times smaller than in the experiment (Fig. 6). At $U=0.7$ V ($\delta_G=0.54$), calculation yields $\Delta_P=0.06$; in the experiment, no bistability occurs at this volt-

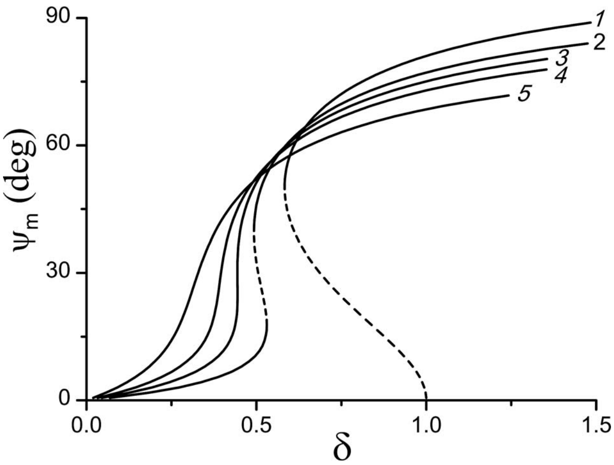


FIG. 10. Theoretical dependences of the angle $\psi_m$ of the director rotation on the dimensionless light wave intensity $\delta$ at $m=3.6$ and different incidence angles $\alpha=(1)0°,(2)10°,(3)15°,(4)20°,(5)30°$.

age. One can see, however, the developed theory correctly describes the character of the electric field influence on the light-induced transition.

Figure 12 shows the theoretical dependences of the director deformation angle on the dimensionless voltage square at different light intensities. It can be seen from Fig. 12 that the cell illumination decreases the threshold of the electric-field-induced transition. At $\delta > 0.2$, the hysteresis appears (Fig. 12, curves 3–5). If $\delta > 0.58$, the director field deformation (initially produced at a sufficiently high $\delta_G$) is retained even after switching off electric field. These calculated modes correspond to those observed in experiment (Fig. 7). However, in experiment the reversible hysteresis loop retains at larger $\delta$: at $P=30$ mW ($\delta=0.81$) the bistability ($\Delta_U=0.58$) was observed, while the calculation gives the irreversible deformation mode for this $\delta$.

Thus, the developed theory adequately describes the experimentally observed light-induced and electric-field-induced orientational transitions in NLC. Quantitative differences may be due to account of only the lowest longitudinal deformation mode, finiteness of the light beam size, light wave decay in the NLC bulk, etc.

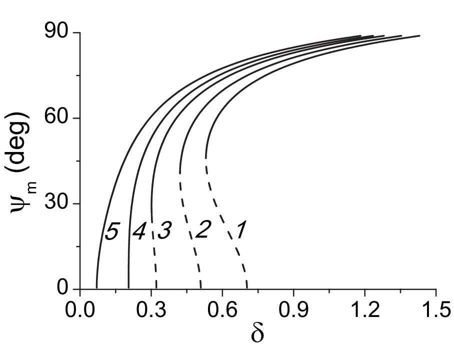


FIG. 11. Theoretical dependences of the director rotation angle $\psi_m$ on the dimensionless light wave intensity at $m=3.6$ and different values of the dimensionless square of the electric field voltage $\delta_G=(1)0,(2)0.28,(3)0.54,(4)0.8,(5)0.9$. Curves (2) and (3) correspond to $U=0.5$ and 0.7 V, respectively.

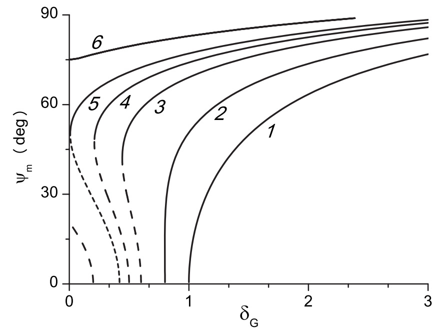


FIG. 12. Theoretical dependences of the director deformation angle $\psi_m$ on the dimensionless square $\delta_G$ of the electric voltage at $m=3.6$ and different values of the dimensionless light wave intensity $\delta=(1)0,(2)0.2,(3)0.4,(4)0.5,(5)0.58,(6)0.8$.

It should be noted that the first-order transitions and the bistability can in principle be expected for NLCs doped with conventional azodyes. However, the sign-inversion enhancement factor $\eta(\Psi)$ was reported for most azodyes studied to date; the negative $\eta$ was only reported for dye DR13 [64]. The data presented in [64] are insufficient to estimate the possibility to observe the first-order transition with this dye.

## V. CONCLUSIONS

The first-order Freedericksz transition in the linearly polarized light wave accompanied by intrinsic director bistability was observed. The transition occurred at a normal incidence of a light beam on a planar cell filled with nematic host doped with dendrimers. The light-induced Freedericksz transition can be controlled by additional ac voltage.

Light illumination transforms the second-order electric-field-induced Freedericksz transition into the first-order one. The bistability region increases with light beam power up to an irreversible deformation.

The relative bistability regions of the found transitions are by an order of magnitude higher than those observed previously for the first-order transitions in undoped NLCs under simultaneous action of light and low-frequency fields.

The first-order transition and bistability are due to the dependence of the enhancement factor (the ratio of the dendrimer-induced torque and the torque exerted on nematic host) on the angle between the director and the light field. The developed theory of the orientation transitions in dendrimer-doped NLC agrees with experimental data.

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## APPENDIX A

The stability of the nontrivial solution $\psi_m^{(0)}$ of Eq. (22) is controlled by the sign of the derivative $\partial F(\psi_m^{(0)},\delta,\delta_G,m)/\partial\psi_m$ (the solution is stable at the negative derivative and unstable in the opposite case). Let us derive expression for this derivative convenient to analyze dependences $\psi_m^{(0)}(\delta)$ (at constant $\delta_G$ and $m$) and $\psi_m^{(0)}(\delta_G)$ (at constant $\delta$ and m).

In the first case, we use the relation

$$\frac{\partial\psi_m}{\partial\delta} = -\frac{\partial F}{\partial\delta}\Big/\frac{\partial F}{\partial\psi_m}, \tag{A1}$$

following from the implicit function theory.

From Eq. (21) it follows that in equilibrium points $\psi_m^{(0)}$

$$\frac{\partial F}{\partial\delta} = \frac{\psi_m - \delta_G J_1(2\psi_m)}{\delta}. \tag{A2}$$

At $\psi_m < \pi/2$ and $\delta_G < 1$, the right-hand side of Eq. (A2) is positive. Then, the sign of the derivative $\partial F/\partial\psi_m$ is opposite to the sign of $\partial\psi_m/\partial\delta$, i.e., the portions of the curve $\psi_m(\delta)$ with positive slope are stable and those with the negative slope, unstable.

In similar lines, in analyzing the dependence $\psi_m^{(0)}(\delta_G)$, we use the relation

$$\frac{\partial\psi_m}{\partial\delta_G} = -\frac{\partial F}{\partial\delta_G}\Big/\frac{\partial F}{\partial\psi_m}. \tag{A3}$$

In equilibrium points $\psi_m^{(0)}$

$$\frac{\partial F}{\partial\delta} = J_1(2\psi_m). \tag{A4}$$

At $\psi_m < \pi/2$ the function $J_1(2\psi_m)$ is positive. Then, again, the portions of the curve $\psi_m(\delta_G)$ with positive slope are stable and those with the negative slope, unstable.

## APPENDIX B

The number of the aberration rings in the cross section of the beam passed through the liquid-crystal layer is [67]

$$N = \frac{|\Delta S_{NL}|}{2\pi}, \tag{B1}$$

where

$$\Delta S_{NL} = \frac{2\pi}{\lambda\cos\beta}\int_0^L [n_e^{(a)}(y) - n_e^{(p)}(y)]dy \tag{B2}$$

is the nonlinear phase shift at the beam axis and $n_e^{(a)}(y)$ and $n_e^{(p)}(y)$ are the refractive indices at the axis and the periphery of the light beam. These indices are expressed in terms of the angles $\Psi^{(a)}$ and $\Psi^{(p)}$ between the light field and the corresponding director

$$n_e^{(a,p)} = \frac{\sqrt{\varepsilon_\perp\varepsilon_\parallel}}{\sqrt{\varepsilon_\perp\cos^2\Psi^{(a,p)} + \varepsilon_\parallel\sin^2\Psi^{(a,p)}}}. \tag{B3}$$

On the assumption that the ratio $\Delta\varepsilon/\varepsilon_\perp$ is small, expression (B3) can be simplified to

$$n_e^{(a,p)} = \sqrt{\varepsilon_\parallel} - \delta n\sin^2\Psi^{(a,p)}, \tag{B4}$$

where $\delta n = \Delta\varepsilon\varepsilon_\parallel^{1/2}/2\varepsilon_\perp$.

Now we should specify the functions $\Psi^{(a,p)}(y)$. Let us consider the normal incidence of a light beam, assuming that the dependence of the director rotation angle on the $y$ coordinate is sinusoidal. Then

$$\Psi^{(a,p)}(y) = \psi_m^{(a,p)}\sin\frac{\pi y}{L}, \tag{B5}$$

where $\psi_m^{(a)} = \psi_m$ and $\psi_m^{(p)} = 0$. Substituting Eq. (B5) into Eq. (B4) and then into Eqs. (B1) and (B2) we arrive at

$$N = N_0\{1 - J_0[2\psi_m^{(a)}]\}, \tag{B6}$$

where $N_0 = \delta nL/2\lambda$.


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