ISSN 1063-7761, Journal of Experimental and Theoretical Physics, 2015, Vol. 120, No. 5, pp. 905–911. © Pleiades Publishing, Inc., 2015.
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STATISTICAL, NONLINEAR, AND SOFT MATTER PHYSICS

Phase Diagrams of Orientational Transitions in Absorbing Nematic Liquid Crystals

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Received October 2, 2014

Abstract—A theory of orientational transitions in nematic liquid crystals (NLCs), which employs the expansion of optical torques acting on the NLC director with respect to the rotation angle, has been developed for NLCs with additives of conformationally active compounds under the action of optical and low-frequency electric and magnetic fields. Phase diagrams of NLCs are constructed as a function of the intensity and polarization of the light field, the strength of low-frequency electric field, and a parameter that characterizes the feedback between the rotation of the NLC director and optical torque. Conditions for the occurrence of first- and second-order transitions are determined. The proposed theory agrees with available experimental data.

DOI: 10.1134/S1063776115040196

1. INTRODUCTION

The action of external (magnetic, electric, or optical) fields on molecules of a nematic liquid crystal (NLC) leads to the rotation of its director **n**. For a mutually perpendicular or parallel orientation of director **n** and the external field, the director rotation has a threshold character (Fréedericksz transition) [1, 2]. The effect exhibits features of a phase transition, since the crystal changes its symmetry and certain critical phenomena are manifested near the threshold [3–6]. The role of the order parameter can be played by the angle of NLC director rotation in the central layer. The Fréedericksz transition usually exhibits features of a second-order transition, so that the director rotation angle is a continuous function of the applied field [2, 3, 7].

The fundamental possibility of a first-order transition in an NLC under the action of a linearly polarized light field followed from an analysis performed in [8–10]. These results showed that a first-order transition can take place in NLCs with a large anisotropy of dielectric permittivity. This effect was not observed experimentally. At the same time, it was shown theoretically and confirmed experimentally that first-order transitions are possible under the combined action of low-frequency and optical fields. A stabilizing low-frequency field can lead to a light-induced first-order transition, while the stabilizing effect of light can lead to a first-order transition induced by low-frequency fields [11–16].

Optical studies stimulated more detailed investigations of the interaction of NLCs with low-frequency fields. It was predicted [17, 18] and confirmed experimentally [18, 19] that a first-order transition takes place in a homeotropically oriented NLC under the action of an electric field applied parallel to the liquid-crystal (LC) layer. Frisken and Palffy-Muhoray [18, 20] theoretically studied the behavior of NLCs in combinations of electric and magnetic fields and showed that, similar to the case of a light field, the stabilizing action of one field can transform the second-order transition induced by another field into a first-order transition. The physical reason for the first-order transitions in all the aforementioned cases was a reverse effect of deformation of the NLC director field on the low-frequency electric field or the light field with extraordinary polarization.

It was also shown [21, 22] that first-order transitions in a circularly polarized light field could be induced by the energy exchange of extraordinary and ordinary waves in an NLC with light-induced nonplanar deformation of the director. In this case, the director field is nonstationary and exhibits precession caused by angular momentum transfer from light to the medium.

In absorbing NLCs, rotation of the director is related to modification of intermolecular forces upon absorption of light photons [23–25] rather than to the action of light on the induced dipoles (as it is in the aforementioned transparent NLCs).

First-order transitions under the action of light beam in the absence of low-frequency fields have been observed and studied in NLCs containing additives of absorbing azobenzene compounds [26–30]. As the power of a light beam with extraordinary polarization normally incident onto a planar-oriented NLC was increased to a certain value P_1, the director exhibited jumplike reorientation. During a subsequent decrease

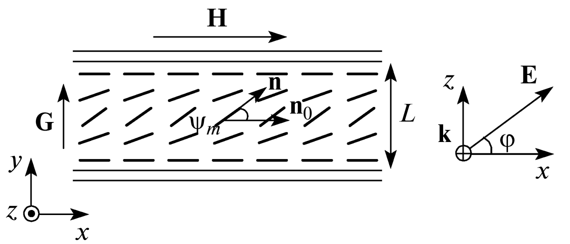

Fig. 1. Geometry of interaction of a light beam and planar NLC: $\mathbf{n}_0$ is the undisturbed director; $\mathbf{n}$ is the director rotated by external fields; ψ_m is the angle of director rotation in the central NLC layer; L is the LC layer thickness; $\mathbf{E}$ and $\mathbf{k}$ are, respectively, the electric field and wave vector of the light incident onto NLC; φ is the angle of rotation of the polarization plane of the incident light beam relative to the xy plane; $\mathbf{G}$ is the low-frequency electric field strength; and $\mathbf{H}$ is a constant magnetic field.

in beam power, the reverse transition to a homogeneous state of the director field took place at $P_2 < P_1$; that is, reorientation of the director field was accompanied by bistability. The relative width of the bistability region, $\Delta = (P_1 - P_2)/P_1$, exceeded by more than an order of magnitude the values known for the first-order optical transitions in transparent NLCs in the presence of an additional field. The application of a low-frequency electric field or the addition of an ordinary wave changed the order of the transition [26, 29].

The experimental results [26–30] were stipulated by additional feedback between the director rotation and optical torque, which appeared under the influence of the light field on the conformational composition of azobenzene compounds [31]. The theoretical description of orientational transitions [26, 29] was based on the exact solution of the torque balance equation for the NLC director.

At the same time, in the most studies, the orientational transitions in NLCs in external fields were theoretically described using the expansion of equations for the field of the NLC director or free energy with respect to the director rotation angle. For example, in [9, 13–15, 18–20], these transitions were treated using the Landau theory of phase transitions [32–34]. This approach yielded an adequate and clear description of the main properties of orientational transitions; in particular, it established conditions for achieving first- and second-order transitions and gave simple analytical expressions for the director rotation angle.

This work constructs a theory for orientational transitions in absorbing NLCs based on expansion of the torque balance equation with respect to the director rotation angle.

2. EQUATION OF DYNAMICS OF THE NLC DIRECTOR

Let us consider the interaction of a planar-oriented NLC with a normally incident linearly polarized light wave (Fig. 1) and introduce a Cartesian coordinate system with axis x parallel to undisturbed director $\mathbf{n}_0$ and axis y perpendicular to the LC layer and parallel to wave vector $\mathbf{k}$ of the light wave. The coordinates of the NLC director $\mathbf{n}(\mathbf{r})$ rotating in the xy plane can be expressed as

$$n_x = \cos\psi, \quad n_y = \sin\psi, \quad n_z = 0, \tag{1}$$

where $\psi(y)$ is the director rotation angle.

For the incident light with the polarization plane rotated by angle φ relative to the xy plane, the light field inside the NLC is a superposition of extraordinary and ordinary waves, which can be expressed as follows:

$$\mathbf{E} = \frac{1}{2}\{\mathbf{e}A_0\exp[i(k_o y - \omega t)] + \text{c.c.}\}, \tag{2}$$

where

$$\mathbf{e} = \mathbf{e}_e\cos\varphi\exp[i(k_e - k_o)y] + \mathbf{e}_o\sin\varphi \tag{3}$$

is the complex unit vector of light polarization; A_0 is the light field amplitude; $\mathbf{e}_e = \mathbf{e}_x$ and $\mathbf{e}_o = \mathbf{e}_z$ are the polarization vectors of the extraordinary and ordinary wave, respectively; k_e and k_o are the corresponding wave vectors; and ω is the light frequency.

The torque related to the modification of intermolecular forces in absorbing NLCs can be expressed as follows:

$$\mathbf{\Gamma}_{\text{abs}} = \eta\mathbf{\Gamma}_{\text{tr}}, \tag{4}$$

where

$$\mathbf{\Gamma}_{\text{tr}} = \frac{\Delta\varepsilon}{4\pi}(\mathbf{n}\cdot\mathbf{E})[\mathbf{n}\times\mathbf{E}] \tag{5}$$

is the optical torque for a transparent (undoped) NLC, $\Delta\varepsilon$ is the optical anisotropy, and η is the torque enhancement factor relative to that of the transparent NLC [31].

For NLCs doped with azobenzene compounds, the enhancement factor η is determined by concentrations c_t and c_c of the *trans* and *cis* isomers in azobenzene chromophores:

$$\eta = \eta_t c_t + \eta_c c_c, \tag{6}$$

where η_t and η_c are the parameters of the LC mixture. The *trans* isomers induce a negative torque in the nematic matrix ($\eta_t < 0$, the director rotates away from light field $\mathbf{E}$), and the *cis* isomers induce a positive torque ($\eta_c > 0$, the director tends to align parallel to light field $\mathbf{E}$). The light field induces conformational transitions of isomers and changes their concentrations [31]. In a sufficiently strong field (where the thermal *cis–trans* relaxation can be ignored), $c_c/c_t \sim \sigma_t/\sigma_c$, where σ_t and σ_c are the absorption cross sections of the corresponding isomers in the nematic matrix, which depend on the geometry of the light wave and the director interaction. This dependence is much more strongly manifested for the *trans* isomer, which has a more elongated shape and is characterized by a higher degree of ordering in the nematic matrix. As a result, the ratio of isomer concentrations (and, hence, the torque enhancement factor) also depends on the geometry of interaction of the light wave and NLC director.

According to the expression obtained in [31] for the torque enhancement factor η in an NLC under the action of an incident linearly polarized light wave, the influence of the geometry of interaction is manifested as the dependence of η on the variable $\sin^2\Psi = \sin^2\psi + \sin^2\varphi\cos^2\psi$ (where Ψ is the angle between the polarization vector of light incident on the NLC and the director $\mathbf{n}$). By expanding the function $\eta(\sin^2\Psi)$ in powers of $\sin^2\Psi$, we can write the torque enhancement factor in the following form:

$$\eta(\psi, \varphi) = \eta_0[1 + m_0(\sin^2\psi + \sin^2\varphi\cos^2\psi)], \quad (7)$$

where η_0 and m_0 are quantities dependent on the concentrations of azochromophores, η_t and η_c values, and parameters characterizing the absorption cross sections of isomers.

Using Eqs. (4), (5), and (7), we can express the total optical torque $\mathbf{\Gamma}_{\text{opt}} = \mathbf{\Gamma}_{\text{abs}} + \mathbf{\Gamma}_{\text{tr}}$ as follows:

$$\mathbf{\Gamma}_{\text{opt}} = (1 + \eta_0)[1 + m(\sin^2\psi + \sin^2\varphi\cos^2\psi)]\mathbf{\Gamma}_{\text{tr}}, \quad (8)$$

where $m = m_0\eta_0/(1 + \eta_0)$. For azo dopants leading to the first-order transition, we have $1 + \eta_0 < 0$ and the NLC director under the action of light rotates away from the light field direction. Parameter m (determined by the properties of the LC system) characterizes the depth of additional feedback between rotation of the director and optical torque $\mathbf{\Gamma}_{\text{opt}}$. The greater m, the higher the rate of torque increase during rotation of the director.

Let an additional low-frequency electric field $\mathbf{G} = \mathbf{e}_y G_0 \sin\Omega t$ perpendicular to the LC layer (where G_0 and Ω are the field amplitude and frequency, respectively) and constant magnetic field $\mathbf{H} = \mathbf{e}_x H$ parallel to the LC layer (see Fig. 1) act on the NLC together with the light field. The torques produced by these fields can be expressed as

$$\mathbf{\Gamma}_{\text{el}} = \frac{\Delta\varepsilon_{\text{lf}}}{4\pi}(\mathbf{n}\cdot\mathbf{G})[\mathbf{n}\times\mathbf{G}],$$

$$\mathbf{\Gamma}_{\text{magn}} = \frac{\Delta\mu}{4\pi}(\mathbf{n}\cdot\mathbf{H})[\mathbf{n}\times\mathbf{H}],$$

where $\Delta\varepsilon_{\text{lf}}$ is the anisotropy of the dielectric permittivity (at frequency Ω) and $\Delta\mu$ is the anisotropy of magnetic permeability. The NLC director is also influenced by the torques of elastic forces, $\mathbf{\Gamma}_{\text{elast}} = K[\mathbf{n}\times\Delta\mathbf{n}]$ (where K is the Frank elastic constant), and viscous forces, $\mathbf{\Gamma}_{\text{visc}} = -\gamma_1[\mathbf{n}\times\partial\mathbf{n}/\partial t]$ (where γ_1 is the viscosity coefficient).

By equating the sum of torques $\mathbf{\Gamma}_{\text{opt}}$, $\mathbf{\Gamma}_{\text{el}}$, $\mathbf{\Gamma}_{\text{magn}}$, $\mathbf{\Gamma}_{\text{elast}}$, and $\mathbf{\Gamma}_{\text{visc}}$ to zero and rejecting rapidly oscillating terms proportional to $\exp[\pm i(k_e - k_o)y]$, we obtain

$$\begin{aligned}\frac{\Delta\psi}{\partial\tau} &= \frac{\partial^2\psi}{\partial\xi^2} + \delta_e\sin\psi\cos\psi \\ &\times[1 + m(\sin^2\psi + \sin^2\varphi\cos^2\psi)] \\ &+ (\delta_G - \delta_H)\sin\psi\cos\psi,\end{aligned} \quad (9)$$

where $\xi = \pi y/L$ and $\tau = t/\tau_0$ are the dimensionless coordinate and time, respectively; $\tau_0 = \gamma_1 L^2/\pi^2 K$; and $\delta_e = |A_0|^2\cos^2\varphi/|A_{e,\text{th}}|^2$ and $\delta_H = H^2/H_{\text{th}}^2$ are the squared strength of the extraordinary wave field and magnetic field, respectively, normalized to the corresponding threshold values

$$|A_{e,\text{th}}|^2 = -\frac{8\pi^3 K}{(1+\eta_0)\Delta\varepsilon}, \quad H_{\text{th}}^2 = \frac{4\pi^3 K}{\Delta\mu L^2},$$

above which reorientation of the director begins. In the case of positive anisotropy $\Delta\varepsilon_{\text{lf}}$ of the permittivity, the value of $\delta_G = G_0^2/G_{0,\text{th}}^2$ (where $G_{0,\text{th}}^2 = 8\pi^3 K/|\Delta\varepsilon_{\text{lf}}|L^2$ has the meaning of the squared threshold field) can be represented as $\delta_G = U^2/U_{\text{th}}^2$, where U is the voltage applied to the NLC and U_{th} is the Fréedericksz threshold voltage. For $\Delta\varepsilon_{\text{lf}} < 0$, we have $\delta_G = -G_0^2/G_{0,\text{th}}^2$ and in this case the low-frequency field stabilizes the undisturbed director field.

By expanding the right-hand side of Eq. (9) into a power series in ψ up to ψ^5, approximating the longitudinal distribution of the director $\psi(\xi, \tau)$ with its lower spatial harmonic as $\psi(\xi, \tau) = \psi_m(\tau)\sin\xi$ (where ψ_m is the director rotation angle in the central plane of the LC-layer $y = L/2$), and using the Galerkin method, we can transform the equation in partial derivatives (9) into an ordinary differential equation:

$$\dot{\psi}_m = -a\psi_m - b\psi_m^3 - c\psi_m^5, \quad (10)$$

where

$$a = 1 - \delta_e(1 + m\sin^2\varphi) - \delta_G + \delta_H, \quad (11)$$

$$b = \frac{1}{2}\delta_e\left(1 - \frac{3}{2}m\right) + \frac{1}{2}(\delta_G - \delta_H) + \frac{5}{4}\delta_e m\sin^2\varphi, \quad (12)$$

$$c = -\frac{1}{12}(\delta_e + \delta_G - \delta_H) + \frac{1}{8}\delta_e m\left(5 - \frac{17}{3}\sin^2\varphi\right). \quad (13)$$

Note that Eq. (10) can also be written in the following form:

$$\dot{\psi}_m = -\frac{\partial F}{\partial\psi_m}, \quad (14)$$

where

$$F = \frac{a\psi_m^2}{2} + \frac{b\psi_m^4}{4} + \frac{c\psi_m^6}{6}.$$

Equation (14) is analogous to the equation that describes the order parameter dynamics in the Landau theory [33, 35], although function F is not a thermodynamic potential. Nevertheless, the formal analogy

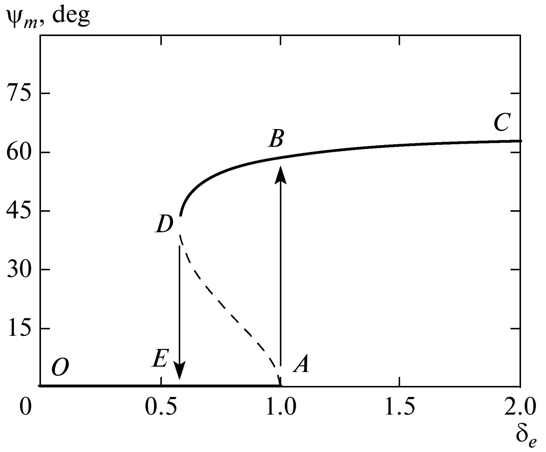

Fig. 2. Plot of the director rotation angle ψ_m vs. dimensionless intensity δ_e of the light beam for $m = 4.35$ (corresponding to the width $\Delta = 0.42$ of the bistability region [26]): (*OA*, *DC*) stable branches; (*AD*) unstable branch; arrows *AB* and *DE* indicate direct and reverse transitions.

of Eq. (14) to the equation of Landau theory allows us to use the latter for determining the order parameter and the type of transition. These quantities can also be determined directly from Eq. (10).

3. LIGHT-INDUCED FIRST-ORDER TRANSITION IN AN NLC

Let us first consider transitions under the action of an extraordinary light wave ($\varphi = 0$) in the absence of low-frequency electric and magnetic fields ($\delta_G, \delta_H = 0$). The transition threshold determined by the condition $a(\delta_e) = 0$ [32, 34] is $\delta_{e,1} = 1$, for which $b = (2 - 3m)/4$. If $b > 0$ ($m < 2/3$), then $\delta_e > 1$ corresponds to a second-order transition.

For $b < 0$ ($m > 2/3$), the threshold at $\delta_e = \delta_{e,1} = 1$ corresponds to a jumplike transition from a homogeneous state with $\psi_m = 0$ (point *A* in Fig. 2) to a deformed state with the amplitude

$$\psi_{m,1}^2 = \frac{-b + \sqrt{b^2 - 4ac}}{2c} \tag{15}$$

(point *B*). Solution (15) (curve *CD* in Fig. 2) exists provided that $b^2 - 4ac \geq 0$. The condition

$$b^2 - 4ac = 0 \tag{16}$$

or in the given case

$$\delta_e = \delta_{e,2} = \frac{8(15m - 2)}{27m^2 + 84m - 4} \tag{17}$$

determines the lower boundary of the bistability region. As δ_e decreases to $\delta_{e,2}$, the system exhibits a reverse jumplike transition from a deformed state (point *D*) to a homogeneous state (point *E*). The width of the bistability region is $\Delta = 1 - \delta_{e,2}$. In addition to solution (15), there exists another solution

$$\psi_{m,2}^2 = \frac{-b - \sqrt{b^2 - 4ac}}{2c},$$

which is depicted by the dashed line in Fig. 2, but this solution is unstable.

The value of $m = 4.35$, for which the plot of $\psi_m(\delta_e)$ was constructed in Fig. 2, corresponds to a width of $\Delta = 0.42$ of the bistability region for the first-order transition observed in a ZhKM-1277 nematic matrix doped with a 0.15% G2 (second-generation) dendrimer [26].

The results are graphically illustrated by the (δ_e, m) phase diagram in Fig. 3a, where curves *1* and *2* correspond to the direct ($\delta_{e,1} = 1$) and reverse (Eq. (17)) transitions, respectively. The director field is homogeneous in region I, homogeneous or deformed (depending on the prehistory) in region II, and deformed in region III. Tricritical point *T*, in which the type of the orientational transition exhibits a change, is determined by the conditions

$$a = 0, \quad b = 0 \tag{18}$$

and has the coordinates ($\delta_{e,T} = 1$, $m_T = 2/3$). Line *AA*' corresponds to $m = 4.35$ determined from experimental data [26], for which the curve $\psi_m(\delta_e)$ is plotted in Fig. 2. The length of segment A_2A_1 is equal to a width of the bistability region $\Delta = 0.42$.

It should be noted that *m* determined [26] from the exact solution to the torque balance equation for the experimental value of $\Delta = 0.42$ was 3.6. The difference from $m = 4.35$ obtained in this work from Eq. (17) is related to the use of expansion with respect to angle ψ.

The values of the width of the bistability region for orientational transitions in the extraordinary light wave can vary significantly for different LC systems. For example, the value for an NLC doped with the G3 (third-generation) dendrimer is $\Delta = 0.22$ [27], which corresponds to $m = 2.32$. For an NLC doped with comblike homopolymers having different numbers of side fragments (14 and 29) [28], the related values were $\Delta = 0.23$ and 0.05 [28], which correspond to $m = 2.40$ and 1.16, respectively. A very large width $\Delta = 0.77$ of the bistability region observed in [30] for an NLC doped with a statistical comblike copolymer corresponded to $m = 16.1$.

4. ABSORBING NLCs IN A COMBINATION OF OPTICAL AND LOW-FREQUENCY ELECTRIC FIELDS

In the presence of a low-frequency electric field ($\delta_H = 0$, $\delta_G \neq 0$) for extraordinary light wave ($\varphi = 0$), the coordinates of a tricritical point on the (δ_e, m) phase diagram determined using Eqs. (11)–(13) and (18) are as follows:

$$\delta_{e,T} = 1 - \delta_G, \quad m_T = \frac{2}{3(1 - \delta_G)}. \tag{19}$$

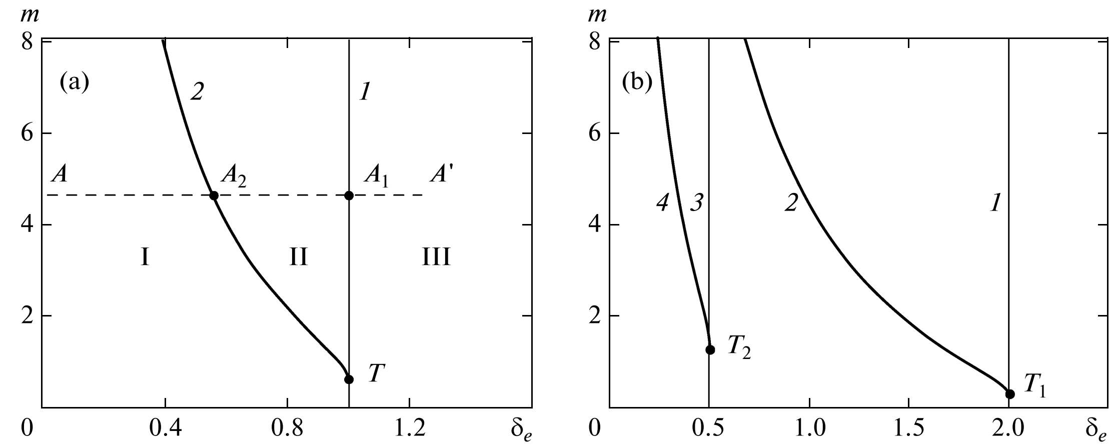

Fig. 3. Phase diagram of orientational transitions plotted in (δ_e, m) coordinates at $\delta_H = 0$, $\varphi = 0$ and $\delta_G = 0$ (a) and $\delta_G = 0.5$ and -1.0 (b). Curves *1* (a) and *1*, *3* (b) correspond to the threshold intensity for transitions into a disturbed state; curves *2* (a) and *2*, *4* (b) correspond to the threshold intensity for reverse transitions; T, T_1, and T_2 are tricritical points. Region I corresponds only to undisturbed state of the NLC director, region II admits the existence of both disturbed and undisturbed states, and region III corresponds only to disturbed states of the director field.

As can be seen from these formulas, the effect of a low-frequency electric field on the light-induced transition depends on the sign of δ_G, i.e., on the sign of the anisotropy of dielectric permittivity $\Delta\varepsilon_{lf}$. For $\Delta\varepsilon_{lf} > 0$ ($\delta_G > 0$), the low-frequency field reduces the transition threshold and increases the m value necessary for the first-order transition. In contrast, for $\Delta\varepsilon_{lf} < 0$ ($\delta_G < 0$), the threshold increases so that the first-order transition takes place at a smaller m. Expressions (11)–(13) show that a constant magnetic field exhibits a stabilizing influence on the light-induced transition, which is analogous to the effect of electric field in the case of $\Delta\varepsilon_{lf} < 0$. Note that, as was pointed out in the Introduction, an analogous effect—the improvement of conditions for the first-order transition in transparent NLCs—was observed in the presence of additional stabilizing fields.

The effect of a low-frequency electric field on the light-induced transition is also manifested in the (δ_e, m) phase diagram in Fig. 3b for $\delta_G = 0.5$ and -1.0. Here, the curves of direct transitions (*1* and *3*) determined from the condition for $a(\delta_e, \delta_G) = 0$ are represented by vertical lines $\delta_{e,1} = 1 - \delta_G$. The curves of reverse transitions (*2* and *4*) were calculated using the relation

$$m = \frac{2\left(10 - 7(\delta_{e,2} + \delta_G) + \sqrt{52(\delta_{e,2} + \delta_G)^2 - 152(\delta_{e,2} + \delta_G) + 100}\right)}{9\delta_{e,2}}, \tag{20}$$

which follows from condition (16).

Figure 4 shows a phase diagram on the (δ_e, δ_G) plane for the experimental conditions [26] ($\Delta\varepsilon_{lf} > 0$), which is plotted using the relation $\delta_{e,1} + \delta_{G,1} = 1$ and a solution of Eq. (16):

$$\delta_{G,2} = 2 - \delta_{e,2} + \frac{21}{2}m\delta_{e,2} - \sqrt{117m^2\delta_{e,2}^2 + 12m\delta_{e,2} + 4}. \tag{21}$$

The coordinates of a tricritical point on this phase diagram are expressed by the following formulas:

$$\delta_{e,T} = \frac{2}{3m}, \quad \delta_{G,T} = 1 - \frac{2}{3m} \tag{22}$$

For the conditions studied in [26], these formulas yield $\delta_{e,T} = 0.15$ and $\delta_{G,T} = 0.85$.

For $\delta_G < \delta_{G,T}$, the light-induced transition is of the first order (line AA' in Fig. 4). For $\delta_G > \delta_{G,T}$ a second-order transition takes place (line BB'). In experiments reported in [26], the Fréedericksz threshold voltage was ($U_{th} = 0.95$ V) and the first-order transition took place at $U = 0$ and $U = 0.5$ V ($\delta_G = 0$ and $\delta_G = 0.28$, respectively), while the second-order transition took place at $U = 0.7$ V ($\delta_G = 0.54$). The latter value is significantly smaller than $\delta_{G,T} = 0.85$, which is probably related to the existence of a pretilt of the director on the LC cell substrates. The pretilt leads to rotation of the director in a subthreshold region, which increases the influence of a low-frequency field and results in a more rapid change in the transition order.

The (δ_e, δ_G) phase diagram in Fig. 4 also illustrates the case of transitions caused by the variation of a low-frequency field in the presence of illumination (vertical lines CC', DD', and EE'). As can be seen, an increase in intensity of the light field must lead sequentially to a second-order transition (CC', $\delta_e < \delta_{e,T}$), first-order transition (DD', $\delta_{e,T} < \delta_e < \delta_{e,2}$), and irreversible first-order

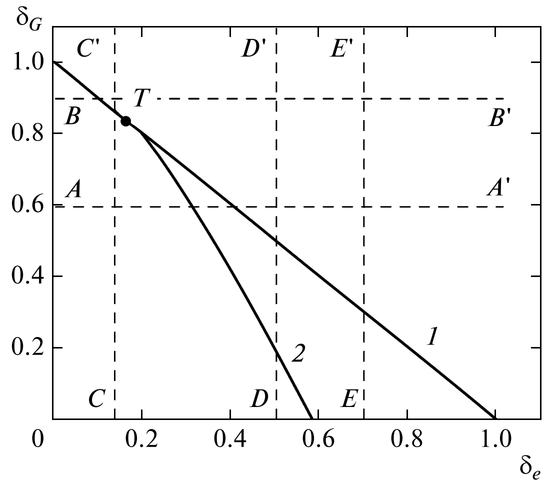

Fig. 4. Phase diagram of orientational transitions plotted in (δ_e, δ_G) coordinates for (*1*) direct transitions at a threshold power from the homogeneous to deformed state and (*2*) reverse first order transitions; *T* is the tricritical point; lines *AA*', *BB*', *CC*', *DD*', an *EE*' correspond to various light-induced and low-frequency electric-field-induced transitions.

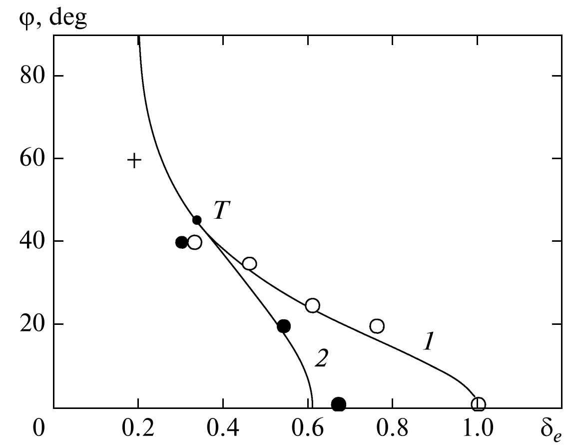

Fig. 5. Phase diagram of light-induced orientational transitions plotted in (δ_e, φ) coordinates at $m = 3.95$ for (*1*) direct and (*2*) reverse transitions in comparison to experimental points for (○) direct and (●) reverse first-order transitions; point (+) refers to a second-order transition [29]. *T* is the tricritical point.

transition (*EE*', $\delta_{e,2} < \delta_e$). In the latter case, an increase in the low-frequency field leads to a jumplike transition from the homogeneous to the deformed state at the point of intersection of lines *EE*' and *1*. The reverse transition to a homogeneous state does not take place even if δ_G decreases to zero (and the deformation is maintained by the light field). All these types of transitions have been experimentally observed [26] at a light beam power of $P = 10$ mW ($\delta_e = 0.27$), $P = 22.5$ and 30 mW ($\delta_e = 0.61$ and 0.81), and $P = 32.5$ mW ($\delta = 0.88$), respectively. However, the light beam powers at which these changes in transition regimes took place are higher than the theoretically calculated values. These discrepancies are apparently explained by the same factors as those in the case of light-induced transitions.

5. EFFECT OF POLARIZATION ON THE LIGHT-INDUCED ORIENTATIONAL TRANSITION IN ABSORBING NLCs

Let us consider the influence of the light polarization on the states of NLC, assuming for the sake of simplicity that low-frequency electric and magnetic fields are absent. Then, the coordinates of a tricritical point are determined by the following relations:

$$\delta_{e,T} = \frac{5}{3(m+1)}, \quad \varphi_T = \arcsin\sqrt{\frac{3m-2}{5m}}. \tag{23}$$

The thresholds of the direct and reverse transitions are given by the formulas

$$\delta_{e,1} = \frac{1}{1 + m\sin^2\varphi}, \tag{24}$$

$$\delta_{e,2} = \frac{8(15m - 17m\sin^2\varphi - 2)}{3(2 - 3m + 5m\sin^2\varphi)^2 + 8(1 + m\sin^2\varphi)(15m - 17m\sin^2\varphi - 2)}. \tag{25}$$

As the angle of rotation of the director φ increases up to the φ_T value, the first-order transition caused by the variation of δ_e changes to the second-order transition. This change is related to weakening of the feedback between director rotation angle ψ and optical torque Γ_{opt} with increasing φ, which follows directly from Eq. (8). At $\delta_e = \delta_{e,T}$, there is a change in the order of a transition induced by the variation of angle φ.

Figure 5 presents the phase diagram of light-induced orientational transitions plotted in (δ_e, φ) coordinates for $m = 3.95$ in comparison to experimental points for the thresholds of first- and second-order transitions [29]. The *m* value was determined by averaging over values calculated using Eq. (24) for the experimental points of direct transitions at various $\varphi \neq 0$ and the width of bistability region at $\varphi = 0$. The data in Fig. 5 show good coincidence of the theory and experiment.

6. CONCLUSIONS

We have constructed a theory of orientational transitions in NLCs doped with light-absorbing conformationally active additives, which employs the expansion of torques acting on the NLC director with respect to the angle of rotation. Transitions induced by variation of the intensity of an extraordinary light wave, light polarization, and low-frequency electric and magnetic fields have been described.

Phase diagrams of NLCs have been calculated as functions of the intensity and polarization of the light field, low-frequency electric field strength, and parameter *m* that characterizes additional feedback between the NLC director rotation and optical torque. Conditions for the occurrence of first- and second-order transitions have been considered. The results of calculations adequately describe the first- and second-order transitions, including a change in the transition type upon application of an additional fields.

NLCs under the action of light and low-frequency electric and magnetic fields provide a good experimental model for studying fluctuational phenomena during first- and second-order phase transitions, including those in the vicinity of tricritical points.

ACKNOWLEDGMENTS

This work was supported by the Russian Science Foundation, project no. 14-12-00784.

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Translated by P. Pozdeev