PHYSICAL REVIEW E **90**, 062503 (2014)

Director reorientation in a nematic liquid crystal with a photosensitive layer

Laura O. Palomares,[1,2] J. Adrián Reyes,[2] and István Jánossy[1]
[1] *Wigner Research Centre for Physics, Hungarian Academy of Sciences, P.O. Box 49, H-1525 Budapest, Hungary*
[2] *Física Química, Instituto de Física, Universidad Nacional Autónoma de México, Apartado Postal 20–364, C.P. 01000, México Distrito Federal, Mexico*

(Received 29 September 2014; published 4 December 2014)

We use a molecular-motor model previously proposed for a nematic cell with an azo-dye monolayer to calculate the director orientation when light is normally impinged on the cell. We consider an initial planar configuration for which one of the surrounding plates, which we call the reference plate, is submitted to a hard-anchoring boundary condition. The other confining plate has a coating monolayer of azo-dye molecules such that the change of the orientation of azo-dye isomers, due to light, causes changes in the nematic director. The boundary conditions on both plates along with the optical field determine the director configuration in the bulk. The existence of periodic solutions for the density of isomers in trans and cis states, corresponding to weak optical fields, has been discussed in the literature. Using a similar approach, we find an approximate expression for the density of isomers, written in terms of the director angle, which allows us to close the equation for the director configuration on the boundary having a photosensitive plate. We decouple the director's angle and the isomer densities by assuming extremely different temporal time scales between them. We show for a given sample that switching times inversely depend on the trans-cis transition rate of photoexcitation whereas relaxation times do not depend on it. On the other hand, switching and relaxation times linearly depend on effective surface viscosity values. Our model allows us to estimate surface viscosity values.

DOI: 10.1103/PhysRevE.90.062503 PACS number(s): 61.30.Gd, 42.79.Kr

I. INTRODUCTION

Photoinduced reorientation in liquid crystals has been studied for several years. There are two fundamental types of optical reorientations: First, the reorientation can be caused by bulk effects [1]; second, the reorientation can be a result of photogenerated modifications in the anchoring conditions at the liquid-crystal boundaries [2]. An irreversible scheme of photoalignment uses polarized light to achieve direction-selective cross-linking or breaking of polymer chains. For reversible realignment most often azo dyes are used, which show trans-cis photoisomerization [2]. The dye molecules are either covalently attached to the substrate surface or incorporated into a polymer matrix. Initially the azo-dye molecules are in a trans configuration and are randomly aligned. In the in-plane reorientation a polarized pump beam excites the trans dyes in an orientationally selective manner. The dyes, which are aligned parallel to the polarization direction, are converted to the cis form; therefore, the orientational distribution of the trans molecules becomes anisotropic, with a peak along the direction perpendicular to the polarization. As the elongated trans molecules align more effectively the liquid-crystal molecules than the V-shaped cis ones, this direction becomes also the surface alignment of the director on the azo-dye-containing plate. In this way, polarized light reorients the surface director perpendicular to its polarization direction [3,4].

In a standard setup for in-plane reorientation the liquid crystal is sandwiched between a reference plate that is covered with a traditional rubbed polyimide layer and a dye-coated plate. In an initially planar cell, when the orienting light beam enters the cell from the dye-coated side, the liquid-crystal layer becomes twisted; the twist angle is determined by the polarization direction of the exciting beam. This procedure was applied by Gibbons *et al.* to write, with the help of polarized light, grayscale images in liquid-crystal cells [5]. Fedorenko *et al.* [6] have studied the effect of light-induced gliding of the easy axis of dye-doped nematic liquid crystal on an aligning polymer surface. A drift of the easy axis over tens of degrees has been observed.

It was shown recently that when the light enters the cell from the reference plate and its polarization is parallel to the director alignment at the entrance face, instability of the planar alignment can occur [7]. There are several models to describe the anisotropic distribution induced by polarized light in a layer of azo-dye molecules [8,9]. In most of these models the azo-containing layer is considered as an isolated unit. To describe the photoinduced reorientation in a standard cell, however, the interaction between the azo dyes at the interface and the nematic liquid crystal has to be considered too. A model taking into account this interaction was proposed by Jánossy *et al.* [10]. Later, the same idea was used by Palffy-Muhoray *et al.* [11]. In their paper they pointed out an analogy between their model of photoalignment and the so-called molecular motors. This model was extended later on by Kinderlehrer and Kowalczyk [12].

In the present paper we utilize the molecular motor description. We are concerned about the temporal evolution of the orientation in a nematic cell with an azo-dye monolayer under photoexcitation. In order to study this evolution we solve the Fokker-Plank equations, which involves the rotational current of isomers that is mainly a diffusive process, the minimization of free energy of the nematic, and the balance of torques. Moreover, in these equations the isomerization due to thermal and photoexcitation processes is taken into account along with the interaction energy between the azo dye and nematic.

This paper is organized as follows. In Sec. II we give a brief description of the physical system and introduce the Fokker-Plank equations and the mathematical approach and methods we used to solve the equations. Section III provides the results of nematic dynamic reorientation when the cell is illuminated from the azo-dye monolayer with light of different intensities and polarizations, the temporal evolution of the twist director configuration in the cell, and the optical path difference. A summary is presented in Sec. IV.

II. PHYSICAL SYSTEM AND FOKKER-PLANCK EQUATIONS

We consider a nematic cell sandwiched between one mechanically buffed polyimide reference plate and one photosensitive alignment layer that consist in an azo-dye monolayer. This cell is illuminated with polarized light. The light polarization is along the initially uniform director field (horizontal polarization), which is parallel to the buffing direction (initial planar configuration). Under these conditions the director field develops a photoinduced twist deformation.

The model proposed by Palffy-Muhoray *et al.* to describe the time evolution both the director and the dye orientational distribution is given by the equations [11,12]

$$\frac{\partial \varrho_t}{\partial t} = \nabla_x(D_t\nabla_x\varrho_t + D_t\varrho_t\nabla_x\psi_1/kT) - \varrho_t f_t + \varrho_c f_c, \quad (1)$$

$$\frac{\partial \varrho_c}{\partial t} = \nabla_x(D_c\nabla_x\varrho_c + D_c\varrho_c\nabla_x\psi_2/kT) - \varrho_c f_c + \varrho_t f_t, \quad (2)$$

$$\gamma\frac{\partial\theta}{\partial t} = \frac{K}{L^2}\frac{\partial^2\theta}{\partial y^2}, \quad (3)$$

$$\gamma_s\frac{\partial\theta}{\partial t} = \frac{K}{L}\frac{\partial\theta}{\partial y} - \sum_{i=1,2}\int_\Omega dx\,\psi_{ix}\varrho_i, \quad y=0, \quad (4)$$

$$\theta = \theta_1 \quad \text{at} \quad y = 1, \quad (5)$$

where $1/D_t$ and $1/D_c$ are the diffusion times for the trans and cis isomers, $\theta = \theta(y,t)$ is the orientation of the director, γ is the orientational viscosity in the bulk, γ_s is the surface orientational viscosity on the layer, L is the sample thickness, K is the liquid-crystal elastic constant, and k is the Boltzmann constant. Here f_t and f_c are transition rates, which take into account changes in the population of cis-trans isomers due to thermal fluctuations. Equations (1) and (2) mainly represent a diffusion process where the last terms are due to the annihilation and creation of isomers.

As in Ref. [12], a geometrically simplified situation is assumed, where the liquid crystal is between two plates at $y = 0$ and 1, with the dye in a thin layer at $y = 0$. Strong anchoring prevails at $y = 1$ (at the reference plate), which translates to a Dirichlet condition for our problem. The dye orientation will be assumed to lie on a circle parametrized by x, $0 \leqslant x \leqslant 2\pi$. We set $n = [\cos\theta(y,t), \sin\theta(y,t)]$, $y \in D = (0,1)$, the trans-isomer and cis-isomer densities per unit of area are $\varrho_t = \varrho_1(x,t)$, $\varrho_c = \varrho_2(x,t)$, where $x \in \Omega$, with $\Omega = \{0 \leqslant x \leqslant 2\pi\}$, and time is positive $t > 0$. All these considerations yield

$$\frac{\partial\rho_i}{\partial\bar t} = \frac{\partial}{\partial x}\left(\frac{\partial\rho_i}{\partial x} + \psi_{ix}\rho_i\right) + \sum_{j=1,2} a_{ij}\rho_j, \quad (6)$$

$$\gamma D\frac{\partial\theta}{\partial\bar t} = \frac{K}{L^2}\frac{\partial^2\theta}{\partial y^2}, \quad (7)$$

$$\gamma_s D\frac{\partial\theta}{\partial\bar t} = \frac{K}{L}\frac{\partial\theta}{\partial y} - \frac{U_{01}}{L^2}\sum_{i=1,2}\int_\Omega dx\,\psi_{ix}\rho_i, \quad y=0, \quad (8)$$

$$\theta = \theta_1 \quad \text{at} \quad y = 1, \quad (9)$$

where the repeated indices do not imply an implicit sum and we have assumed $D_t = D_c = D$. Here $\varrho_i = \rho_i/L^2$ $(i = 1,2)$ are the surface fraction of isomers, $\bar t = tD$, $\sigma = kT/U_{01}$ is a parameter measuring the ratio between the equilibrium thermal energy and the strength of the interaction energy of trans cis isomers, and $\psi_{1\,(2)} = \cos^2(\theta - x)/\sigma$ is the interaction energy of the dye in the trans (cis) state and the nematic, which depends on the angle between the nematic and the dye molecules, where x is the direction of the dye director with respect to the direction of the initial planar configuration. Here the coefficients a_{ij} are dimensionless transition rates, the interaction energy between the azo dye and nematic, and photoexcitation, given by the expressions

$$-a_{11} = a_{21} = f_t/D = \nu_{10}e^{\psi_1} + \mu_t(\mathbf{E}\cdot\mathbf{I})^2, \quad (10)$$

$$-a_{22} = a_{12} = f_c/D = \nu_{20}e^{U_{02}\psi_1/U_{01}} + \mu_c(\mathbf{E}\cdot\mathbf{I})^2, \quad (11)$$

where $\mathbf{I} = (\cos x, \sin x)$ is the orientation of the dye isomers, ν_{i0} are constants with $\nu_{20} + \nu_{10} = 1$, ν_{10} is a small constant, μ measures the trans-cis transition rate due to photoexcitation, and $\mathbf{E} = \mathbf{e}/E_0$ is the dimensionless electric field of light, where its polarization in the most general case is time dependent $\mathbf{e} = [E_1(t), E_2(t)]$ and E_0 is a reference field.

A. Set of equations for weak optical fields

Here we shall consider the particular case in which the interaction energy of the dye in the cis state and the nematic is null, $\psi_2 = 0$, $\nu_{10} = 0$, $\nu_{20} = 1$, $\mu_c = 0$, $\mu_t = \mu$, and $\mathbf{E} = (E,0)$ (horizontally polarized light). These parameters correspond to a moderately strong interaction between the trans isomer and the nematic field, but no interaction between the cis isomer and the nematic field. This gives

$$-a_{11} = a_{21} = \mu(\mathbf{E}\cdot\mathbf{I})^2 = \mu E^2\cos^2 x, \quad (12)$$

$$-a_{22} = a_{12} = \nu_{20}. \quad (13)$$

Let us consider that the relaxation for the dye is much faster than that of the nematic, so we can approximate the set of equations by assuming that ρ_i have relaxed to a state in which they follow instantaneously the dynamics of θ. This amounts to assuming that $\partial\rho_i/\partial\bar t = 0$ by keeping a nonvanishing value of $\frac{\partial\theta}{\partial t}$. In other words, the densities of the isomers ρ_i are slaved by θ since the former have a response time faster than θ. Hence, if we define the variable $w_1(x) = e^{\psi_1}\rho_1$ and insert it into Eq. (6) we can solve this equation system for this variable, for values of $\mu \simeq 0$ (which corresponds to a slow cis-trans transition rate due to photoexcitation), to obtain [12]

$$w_1(x) = 1 + \mu W^1 + O(\mu^2), \quad (14)$$

where W^1 is given by the expression

$$\begin{aligned} W^1(x) = {} & W^1(0) + \beta_1 \int_0^x e^{\cos^2(\theta-\bar{t})/\sigma} dt \\ & + \frac{E^2}{2\sigma} \int_0^x e^{\cos^2(\theta-\bar{t})/\sigma} \left(\int_0^{\bar{t}} e^{\lambda s} e^{-\cos^2(\theta-s)/\sigma} \cos^2 s \, ds \, e^{-\lambda \bar{t}} - \int_{\bar{t}}^{2\pi} e^{-\lambda s} e^{-\cos^2(\theta-s)/\sigma} \cos^2 s \, ds \, e^{\lambda \bar{t}} \right) d\bar{t} \\ & - \frac{1}{2} \int_0^x e^{\cos^2(\theta-\bar{t})/\sigma} \left(e^{\lambda \bar{t}} \frac{K_1}{e^{2\pi\lambda}-1} - e^{-\lambda \bar{t}} \frac{K_2}{e^{2\pi\lambda}-1} \right) d\bar{t}. \end{aligned} \tag{15}$$

We can obtain β_1 from (15) by taking into account periodic solutions $W^1(2\pi) = W^1(0)$,

$$\begin{aligned} \beta_1 = {} & \left[\frac{1}{2} \int_0^{2\pi} e^{U_{10} \cos^2(\theta-\bar{t})/\sigma} \left(e^{\lambda t} \frac{K_1}{e^{2\pi\lambda}-1} - e^{-\lambda \bar{t}} \frac{K_2}{e^{2\pi\lambda}-1} \right) d\bar{t} - \frac{E^2}{2\sigma} \int_0^{2\pi} e^{U_{10} \cos^2(\theta-\bar{t})/\sigma} \right. \\ & \left. \times \left(\int_0^t e^{\lambda s} e^{-U_{10} \cos^2(\theta-s)/\sigma} \cos^2 s \, ds \, e^{-\lambda \bar{t}} - \int_t^{2\pi} e^{-\lambda s} e^{-U_{10} \cos^2(\theta-s)/\sigma} \cos^2 s \, ds \, e^{\lambda \bar{t}} \right) \right] \Big/ \int_0^{2\pi} e^{U_{10} \cos^2(\theta-\bar{t})/\sigma} d\bar{t}, \end{aligned} \tag{16}$$

where $\lambda = \sqrt{\nu_{20}/\sigma}$ and

$$K_1 = \frac{E^2}{\sigma} \int_0^{2\pi} ds \, e^{-\lambda s - U_{10} \cos^2(s-\theta)/\sigma} \cos^2 s, \tag{17}$$

$$K_2 = \frac{E^2}{\sigma} \int_0^{2\pi} ds \, e^{\lambda s - U_{10} \cos^2(s-\theta)/\sigma} \cos^2 s. \tag{18}$$

The torque exerted by the dye on the liquid crystal results from the last term of Eq. (8), which we can calculate from $w_{1x}(x) = \frac{dw_1(x)}{dx}$ to obtain

$$\rho_1 \psi_{1x} = w_{1x}(x) e^{-\psi_1} - \rho_{1x}. \tag{19}$$

Thus, the surface torque density reduces to the expression

$$\tau = \frac{U_{01}}{L^2} \sum_{i=1,2} \int_\Omega dx \, \psi_{ix} \rho_i = \frac{U_{01}}{L^2} \int_0^{2\pi} w_{1x}(x) e^{-\psi_1} dx \tag{20}$$

if we restrict our treatment to periodic conditions $\rho_1(2\pi) = \rho_1(0)$. Inserting Eq. (15) in Eq. (14), we obtain $w_{1x}(x)$ to first order in μ, $w_{1x}(x) = \mu W_x^1$. Thus, by solving the integral of Eq. (20) we obtain the next expression for the torque

$$\tau = \mu \frac{U_{01}}{L^2} \beta_1 2\pi, \tag{21}$$

where $\beta_1 = \beta_1(\theta(y,t))$. Inserting the expression of the torque $\tau = \tau(\theta)$ into Eq. (8), we get

$$\gamma_s D \frac{\partial \theta}{\partial \bar{t}} = \frac{K}{L} \frac{\partial \theta}{\partial y} - \tau(\theta). \tag{22}$$

This first-order partial equation for the orientation is only valid at the dye layer ($y = 0$). Hence, it plays the role of a boundary condition for Eq. (7). To find the value $\frac{\partial \theta}{\partial y}$ at the layer border, it is convenient to obtain the solution of Eq. (7), which can be expressed in terms of the boundary condition [13]

$$\theta(y,\bar{t}) = \frac{2K}{\gamma D L^2} \sum_{n=1}^{\infty} n \sin n\pi y \int_0^{\bar{t}} \exp\left(-\frac{K n^2 \pi^2}{D L^2 \gamma} (\bar{t} - t') \right) \times \theta(0,t') dt'. \tag{23}$$

From this expression we calculate straight forwardly $\frac{\partial \theta(0,\bar{t})}{\partial y}$ at $y = 0$.

$$\frac{\partial \theta(0,\bar{t})}{\partial y} = \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{K}{\gamma D L^2} n^2 \pi^2 \int_0^{\bar{t}} \exp\left(-\frac{K n^2 \pi^2}{D L^2 \gamma} (\bar{t} - t') \right) \times \theta(0,t') dt'. \tag{24}$$

We consider the discrete variable $z_n = \sqrt{K/\gamma D} n\pi/L$. Thus, we rewrite the former equation as

$$\frac{\partial \theta(0,\bar{t})}{\partial y} = \frac{1}{\pi} \int_0^{\bar{t}} \sum_{n=1}^{\infty} z_n^2 e^{-z_n^2(\bar{t}-t')} \theta(0,t') dt'. \tag{25}$$

If the distance $\Delta z = z_{n+1} - z_n = \sqrt{K/\gamma D}\pi/L = 1/\sqrt{t} \ll 1$, then the sum can be approximated by an integral

$$\sum_{n=1}^{\infty} z_n^2 e^{-z_n^2(\bar{t}-t')} \rightarrow \int_0^{\infty} z^2 e^{-z^2(\bar{t}-t')} dz = \frac{1}{4} \sqrt{\frac{\pi}{(\bar{t}-t')^3}}. \tag{26}$$

If we consider $L = 10\ \mu$m, $K = 10^{-11}$ N, and $\gamma = 10^{-1}$ Pa s, we obtain a characteristic time $t = L^2 \gamma / K \sim 1$ s and then, for times shorter than t, the sum can be approximated by the integral. However, this time interval is still very short for our relaxation process, which has been observed in the experiments to be of the order of minutes. Indeed, the only way to reduce Δz is by widening the sample corresponding to a thicker sample. Thus, this approach is not adequate for our purposes.

A complementary approximation can be developed for times larger than t, the exponential with $n = 1$ decays rapidly, and the following exponential terms with $n = 2$ and 3 decay with a decay constant 4 and 9 times larger, respectively, so we calculate only the first term of the sum, i.e.,

$$\frac{\partial \theta(0,\bar{t})}{\partial y} = \frac{2}{\pi} \frac{K}{\gamma D L^2} \pi^2 \int_0^{\bar{t}} \exp\left(-\frac{K \pi^2}{L^2 \gamma D} (\bar{t} - t') \right) \theta(0,t') dt'. \tag{27}$$

Substituting into Eq. (22) for the boundary condition of the azo-dye monolayer, we obtain

$$\gamma_s D\frac{\partial\theta}{\partial\bar{t}} = \frac{K}{L}\frac{2}{\pi}\frac{K}{\gamma DL^2}\pi^2\int_0^{\bar{t}}\exp\left(-\frac{K\pi^2}{L^2\gamma D}(\bar{t}-t')\right) \times\theta(0,t')dt' - \tau(\theta), \tag{28}$$

which describes the orientation of the nematic at the dye layer induced by the optical reorientation of the dye. If we rewrite this equation in terms of the dimensionless variable $\hat{\tau} = \frac{K}{\gamma DL^2}t = \frac{1}{\tau}\bar{t}$, where τ is the relaxation time and L is the cell thickness ($L \sim 10$ μm); we considered $L = 10$ μm. We get, after direct substitution of $\hat{\tau}$ in Eq. (28),

$$\frac{K\gamma_s}{\gamma L^2}\frac{\partial\theta}{\partial\hat{\tau}} = \frac{K}{L}\frac{2}{\pi}\pi^2\int_0^{\hat{\tau}} e^{-\pi^2(\hat{\tau}-t')}\theta(0,t')dt' - \tau(\theta), \tag{29}$$

which become

$$\frac{\partial\theta(0,\hat{\tau})}{\partial,\hat{\tau}} = \frac{2\gamma L}{\gamma_s}\pi\int_0^{\hat{\tau}} e^{-\pi^2(\hat{\tau}-t')}\theta(0,t')dt' - \frac{\gamma L^2}{K\gamma_s}\tau(\theta). \tag{30}$$

Note that this is an equation valid only on the surface on the azo-dye monolayer; then bulk effects are not taken into account. Next, we shall solve the latter equation for the director orientation at the boundary with the azo-dye monolayer by using the numeric method that we explain in detail in the next section.

B. Numeric formulation of the integro-differential equation

To discretize Eq. (30), we substitute the derivative by its discrete formula, that is,

$$\frac{\partial\theta(0,\hat{\tau})}{\partial\hat{\tau}} = \frac{\theta(0,\hat{\tau}_{j+1}) - \theta(0,\hat{\tau}_j)}{\Delta\hat{\tau}}, \tag{31}$$

and then the integral can be expressed as

$$\int_0^{\hat{\tau}} e^{-\pi^2(t-\hat{\tau})}\theta(0,\hat{\tau})d\hat{\tau} = \sum_{i=0}^{j}\theta(0,\hat{\tau}_j)\exp(\hat{\tau}_i - \hat{\tau}_j)\Delta\hat{\tau}. \tag{32}$$

Thus, inserting both expressions in the integro-differential equation (30) we get

$$\frac{\theta(0,\hat{\tau}_{j+1}) - \theta(0,\hat{\tau}_j)}{\Delta\hat{\tau}} = \frac{2\gamma L}{\gamma_s}\pi\sum_{i=0}^{j}\theta(0,\hat{\tau}_j)\exp(\hat{\tau}_i - \hat{\tau}_j)\Delta\hat{\tau} - \frac{\gamma U_{01}}{\gamma_s K}2\pi\mu\beta(\theta(0,\hat{\tau}_j)). \tag{33}$$

Numerical calculation for β_1 shows that it can be suitably fitted by the function

$$\beta_1(\theta) = [A + B\sin 2\theta(\hat{\tau})], \tag{34}$$

where A and B are constants. We can express the discrete version of the differential equation as the matrix given by

$$\begin{bmatrix} -1 & 1 & 0 & 0 & 0 & \cdots \\ 0 & -1 & 1 & 0 & 0 & \cdots \\ 0 & 0 & -1 & 1 & 0 & \cdots \\ 0 & 0 & 0 & -1 & 1 & \cdots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \ddots \end{bmatrix}\begin{bmatrix} \theta(\hat{\tau}_1) \\ \theta(\hat{\tau}_2) \\ \theta(\hat{\tau}_3) \\ \theta(\hat{\tau}_4) \\ \vdots \end{bmatrix} + \frac{2\gamma L}{\gamma_s}\pi\Delta\hat{\tau}^2\begin{bmatrix} \exp(\hat{\tau}_2-\hat{\tau}_1) & 0 & 0 & 0 & 0 & \cdots \\ \exp(\hat{\tau}_3-\hat{\tau}_2) & \exp(\hat{\tau}_3-\hat{\tau}_1) & 0 & 0 & 0 & \cdots \\ \exp(\hat{\tau}_4-\hat{\tau}_3) & \exp(\hat{\tau}_4-\hat{\tau}_2) & \exp(\hat{\tau}_4-\hat{\tau}_1) & 0 & 0 & \cdots \\ \exp(\hat{\tau}_5-\hat{\tau}_4) & \exp(\hat{\tau}_5-\hat{\tau}_3) & \exp(\hat{\tau}_5-\hat{\tau}_2) & \exp(\hat{\tau}_5-\hat{\tau}_1) & 0 & \cdots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \ddots \end{bmatrix}\begin{bmatrix} \theta(\hat{\tau}_1) \\ \theta(\hat{\tau}_2) \\ \theta(\hat{\tau}_3) \\ \theta(\hat{\tau}_4) \\ \vdots \end{bmatrix} = \frac{U_{01}\gamma}{\gamma_s K}2\pi\mu\Delta\hat{\tau}\begin{bmatrix} \beta_1(\theta(\hat{\tau}_1)) \\ \beta_1(\theta(\hat{\tau}_2)) \\ \beta_1(\theta(\hat{\tau}_3)) \\ \beta_1(\theta(\hat{\tau}_4)) \\ \vdots \end{bmatrix}.$$

It is interesting to note that the matrix of the second term being triangular permits us to solve consecutively the linear equation system established by this expression.

III. RESULTS AND DISCUSSION

Photoinduced twist for different polarizations, transition rates of photoexcitation, and viscosities

Figure 1 depicts the director orientation at the boundary with the azo-dye monolayer when the nematic cell is illuminated with horizontally polarized light for different trans-cis transition rates of photoexcitation. The initial condition is a planar nematic cell with $\theta(y,0) = 0^\circ$. These plots show that under photoexcitation, the director direction evolves to a maximum twist angle θ_m (value reached after a switching time of around 65 s for $\mu_1 = 8 \times 10^3$ m^2/V^2 s, considering

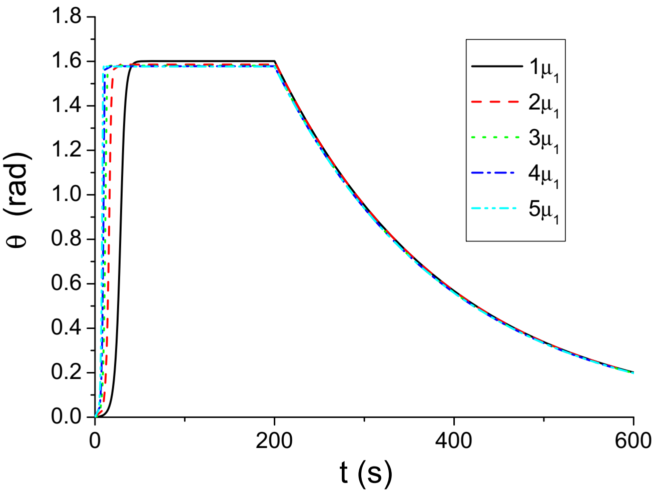

FIG. 1. (Color online) Director orientation when the nematic cell is illuminated with horizontally polarized light (polarization in the direction of the initial planar configuration) for different transition rates of photoexcitation. Light is switched off after 200 s. The initial condition is a planar nematic cell with $\theta_0 = 0^\circ$. The following parameters were used: $\psi_2 = 0$, $\nu_{10} = 0$, $\nu_{20} = 1$, $\mu_c = 0$, $\gamma = 0.08$ Pa s (5CB), $\gamma_e = 564$ nm, $\sigma = kT/U_{01} = 2$, $U_{02} = 0$, $L = 10^{-5}$ m, $K = 10^{-11}$ N, and $\mu_1 = 8 \times 10^3$ m^2/V^2 s.

a typical irradiance value for a He-Ne laser beam waist), and remains at that value until the light is switched off and the relaxation process starts. Notice that for lower values of light intensity, θ_m is reached after a longer time. Unless stated otherwise, the light is switched off after 200 s. In some graphs there is an initial maximum value for a short time that suddenly reduces to a steady value that we take as θ_m. Here we define the switching time as the time to increase the director angle from the initial planar configuration $\theta = 0$ to 90% of θ_m upon photoexcitation; in contrast, the relaxation time is the time to relax the director configuration from θ_m to 10% of θ_m when the light is switched off.

For each value of the trans-cis transition rate of photoexcitation there are two possible solutions for Eq. (33): One is clockwise and the other is counterclockwise for the director direction; they are mirror solutions of each other, as shown in Fig. 2. In experiments the twist direction can be biased by adding a small quantity of chiral molecules to the nematic cell.

Since polarized light tends to reorient the azo-dye molecules in the direction perpendicular to polarization, there is almost no effect on a planar cell sample upon being illuminated with vertical polarized light and a similar behavior occurs when an initially twisted sample at $\pi/2$ is illuminated with horizontally polarized light. In contrast, when the twisted sample at $\pi/2$ is illuminated with different trans-cis transition rates of photoexcitation for vertically polarized light (perpendicular to the initial planar configuration), two different solutions are found, as shown in Fig. 3: One increases the twist angle from $\pi/2$ to approximately π and the other one takes the cell to its initial configuration. When the light is switched off, the cell evolves to its initial planar configuration in both cases. Thus, the relaxation time for the second solution is null, but in the switched off case the decay rate is faster.

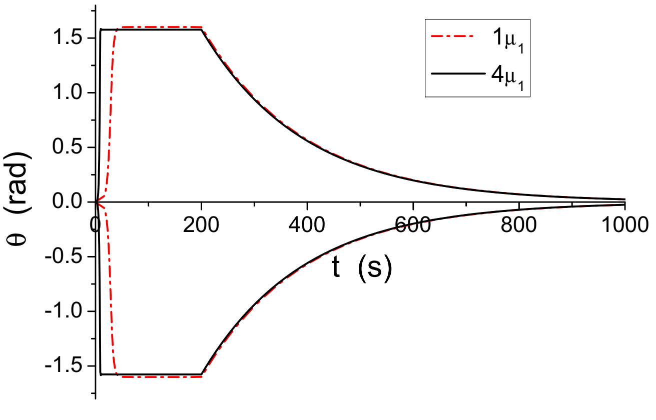

FIG. 2. (Color online) Director orientation when the nematic cell is illuminated with horizontally polarized light for certain trans-cis transition rates of photoexcitation. The initial conditions are those of a planar cell. There are two possible director orientations, one clockwise and the other counterclockwise. The parameter values are the same as in Fig. 1.

Figure 4 presents a comparison between the twist angle when the cell is illuminated with vertically and horizontally polarized light. In this figure the scale for the vertically polarized light curve is translated by $\pi/2$ in order to compare closely with the horizontally polarized light curve. Relaxation times have similar values when the sample is illuminated with vertically and horizontally polarized light of a given intensity and the corresponding switching times are similar too. We observe that θ_m is slightly larger for vertically polarized light. Note that the vertically polarized light curve without photoexcitation and translation of $\pi/2$ goes from π to 0 rad, whereas the horizontally polarized light curve starts in $\pi/2$

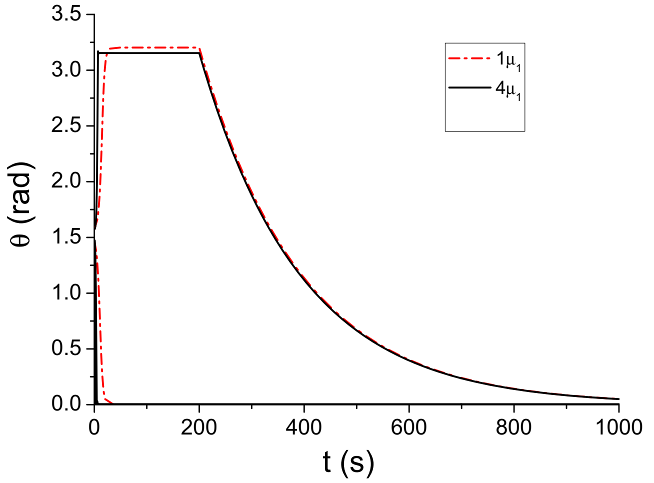

FIG. 3. (Color online) Director orientation when the nematic cell is illuminated with vertically polarized light for two different trans-cis transition rates for light intensity. The initial condition is a twisted nematic cell of $\pi/2$. There are two possible director orientations under photoexcitation: clockwise, which increases the twist at around π, and counterclockwise, which evolves to the initial planar configuration. Light is switched off after 200 s.

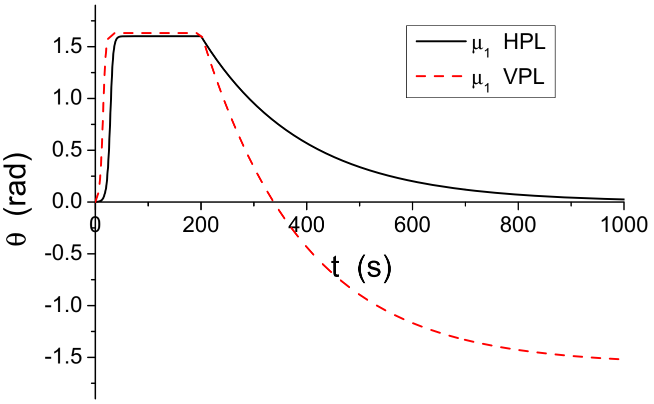

FIG. 4. (Color online) Director orientations when the nematic cell is illuminated with horizontally and vertically polarized light for two different trans-cis transition rates for the light intensity. The initial condition is a planar cell for horizontally polarized light (HPL) and a twist cell at $\pi/2$ for vertically polarized light (VPL).

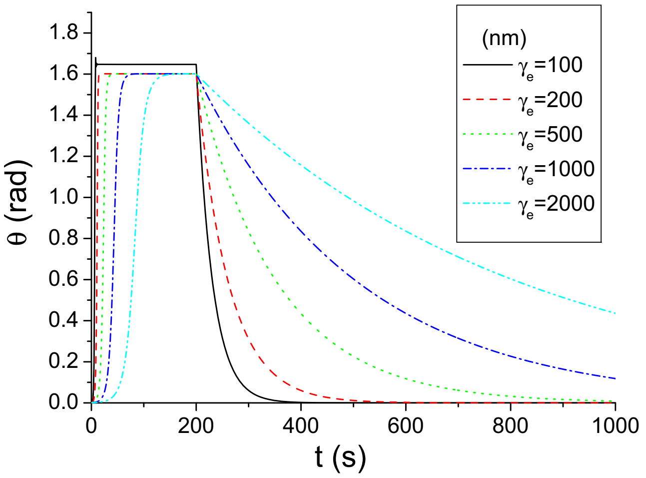

FIG. 6. (Color online) Director direction θ vs time for different values of effective surface viscosity (rate between surface viscosity and bulk viscosity) γ_e and the same parameters as in Fig. 1, with $\mu = \mu_1$ and horizontally polarized light. The light is switched off after 200 s.

and diminishes to 0 rad; nevertheless, they display similar relaxation times.

Figure 5 exhibits switching times t_s vs the trans-cis transition rate of photoexcitation μ, showing an inversely proportional behavior. For higher transition rates, switching times are faster compared with those for lower intensities.

Figure 6 displays various plots of the director reorientation due to photoexcitation for different effective surface viscosity values. Here we see that the switching and relaxation times depend on the effective surface viscosity $\gamma_e = \frac{\gamma_s}{\gamma}$. We use the same parameter values as in Fig. 1 as well as $\mu = \mu_1$.

Figures 7 and 8 show the switching and relaxation times as a function of the effective surface viscosity together with a very good linear fit adjusted to the data calculated. Here the switching and relaxation times tend to zero for a vanishing value of the effective surface viscosity, however, the numerical solution becomes oscillating by considering values of the effective surface viscosity lower than 100 nm.

Figure 9 depicts the director configuration in the cell against time and the distance inside the cell. Our results exhibit the expected behavior. That is, for $y = 0$ we have assumed the strong anchoring condition $\theta_0 = 0\,\forall t > 0$, whereas for $y = 1$ we have obtained the boundary condition from Eqs. (6)–(9). We have considered the particular case for $\mu = \mu_1$, which is shown in Fig. 1. Finally, the director configuration in the whole bulk, corresponding to values of y in the interval [0,1], is uniformly twisted. Then, from Eq. (21) we have

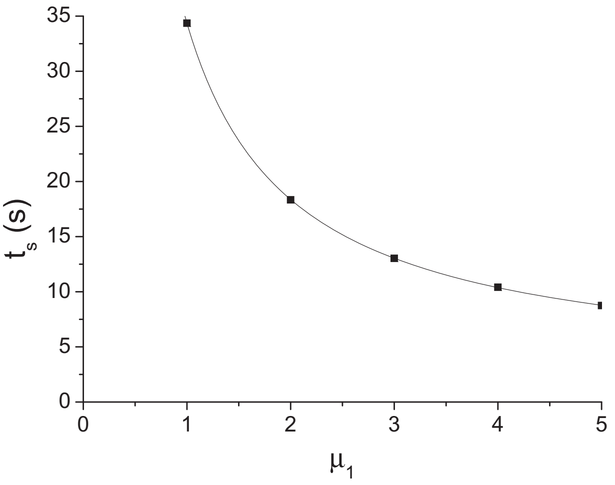

FIG. 5. Switching time t_s vs trans-cis transition rates for light intensity μ. They are inversely proportional. Horizontally polarized illuminating light with initial planar configuration is considered. The squares correspond to numerical calculations obtained by using this model while the solid line is a fitted curve, for the same parameter values as in Fig. 1.

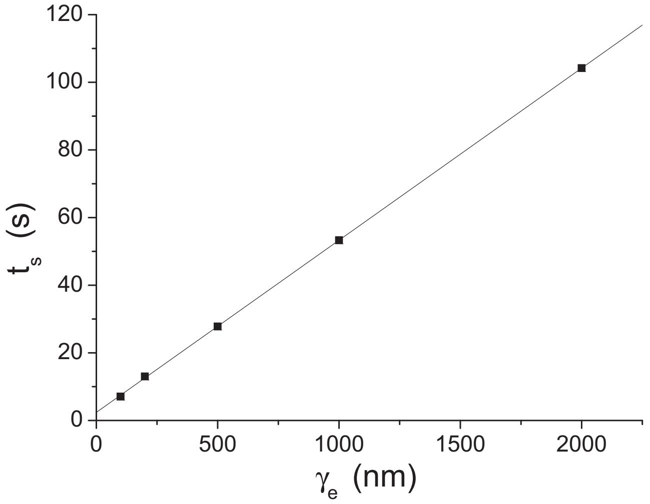

FIG. 7. Switching time t_s vs effective surface viscosity γ_e. The squares correspond to numerical calculations obtained by using this model while the solid line curve is a fitted line, for the same parameters as in Fig. 1, with $\mu = \mu_1$ and horizontally polarized light.

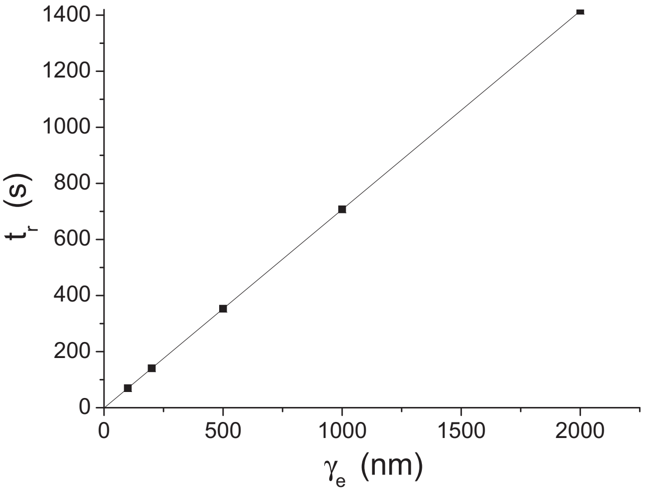

FIG. 8. Relaxation time t_r vs γ_e. The relaxation time is the required time to reorient the cell from the maximum twist to 10% of θ_m, after turning off the light. The squares correspond to numerical calculations and the solid line is a fitted line, for the same parameters as in Fig. 1, with $\mu = \mu_1$ and horizontally polarized light.

calculated the temporal evolution of the torque $\tau(\theta(t))$, which is displayed in Fig. 10. The graph shows that the torque presents a very pronounced minimum for a value around 28 s, for which the angular acceleration is maximum. Indeed, the torque exhibits its largest variations for the switching time interval; after this interval the torque is small and almost constant, which is consistent with an asymptotically constant angle.

Note that the switching and relaxation times for the director direction are calculated for the azo-dye monolayer, which is the boundary of the system. They are given in s; indeed, t_r and t_s are expected to be of the order of seconds or minutes for the whole cell. In our computer simulations we have found no oscillating solutions of the twist angle only for values of surface viscosities $\gamma_e \geqslant 100$ nm when $\mu = \mu_1$. It is worth mentioning that our calculated effective surface viscosity values are of the order of those experimentally obtained for a nematic cell with a photoaligning poly-(vinyl-cinnamate) layer [14]. Here we consider the assumption of a time-scale difference, for which the relaxation time for the dye is much faster than that of the nematic. Under this hypothesis the only time scale involved in the dynamics is that of the nematic orientation since that corresponding to the dye was completely neglected. In this sense the dye time scale would contribute with a delay between the dye and the nematic's director, which can be accumulated in each thin film forming the bulk in order to give rise to a whole delay that would be quite different in order of magnitude from that of the single layer at the boundary.

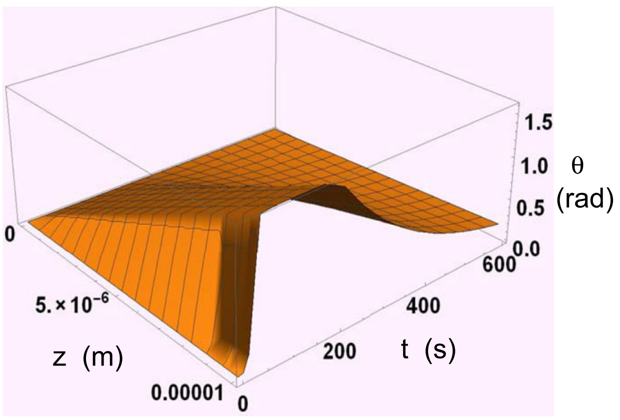

FIG. 9. (Color online) Director orientation in the nematic cell versus the normal distance and time. The light is switched off after 200 s.

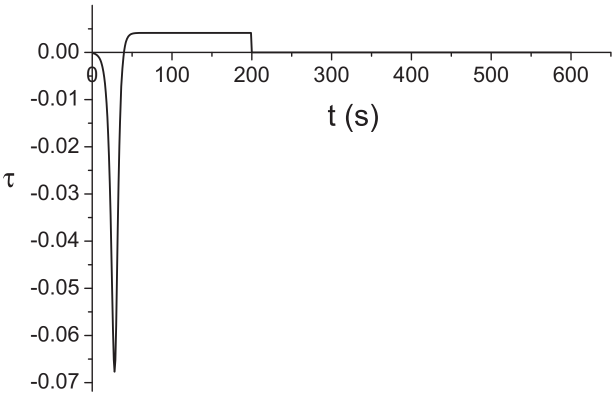

FIG. 10. Torque exerted on the nematic for the azo-dye molecules when the horizontal polarized light normally impinges on the nematic cell, for the same parameters as in Fig. 1, with $\mu = \mu_1$.

IV. CONCLUSION

In summary, we have established and solved the integro-differential equation governing the director orientation in a nematic's cell boundary in which there is also an azo dye, along with the equation of the bulk director vector by using a perturbative approach similar to that given in Ref. [12]. Here we have utilized a Fourier series expansion to relate the values of the azo-dye orientation and surface nematic orientation to those of nematic bulk. Finally, we have proposed a certain discretization method to solve the resulting integro-differential equation found for our model.

We have found in our numerical simulation that neither the maximum angles nor the relation times depend on the trans-cis transition rate of photoexcitation. However, whereas switching times inversely depend on the trans-cis transition rates of photoexcitation, relaxation times practically do not depend on it for a specific nematic cell with an azo-dye monolayer. On the other hand, switching and relaxation times linearly depend on the effective surface viscosity values of the nematic. We have found that the maximum twist angle in the cell does not depend on the trans-cis transition rates and effective surface viscosities. Therefore, it is possible to control switching times by means of light intensity for the same parameters of the sample, whereas, switching and relaxation times can be controlled by choosing a nematic cell with azo-dye monolayer having a suitable effective surface viscosity value. Nevertheless, we have found that only for effective surface viscosity values larger than or equal to 100 nm

($\gamma_e \geqslant 100$ nm) does the solution of Eq. (30) (the time evolution of the director direction on an azo-dye monolayer plate) exhibit a nonoscillating solution, when $\mu = \mu_1$. Here the interaction energy strength of trans isomers is considered to be of the order of 0.5 times the thermal activation energy $U_t = 0.5kT$. Finally, it is important to stress that by using our model it is possible to estimate surface viscosity values of the nematic by fitting the parameter values for the integro-differential equations.

ACKNOWLEDGMENTS

This work was supported by the Hungarian National Science Foundation OTKA under Contract No. K 81250 and partially supported by DGAPA Grant No. UNAM IN110012-3. L.O.P. acknowledges the postdoctoral scholarship provided by "Consejo Nacional para la Ciencia y Tecnología" (CONACYT), Mexico, to carry out this research.

[1] L. Marrucci, Liquid Cryst. Today **11**, 6 (2002).

[2] K. Ichimura, Chem. Rev. **100**, 1847 (2000).

[3] A. G. Dyadyusha, T. Marusii, Y. Reznikov, A. Khiznyak, and V. Reshetnyak, JETP Lett. **56**, 17 (1992).

[4] W. M. Gibbons, P. J. Shannon, S. T. Sun, and B. J. Swetlin, Nature (London) **351**, 49 (1991).

[5] W. M. Gibbons, T. Kosa, P. Palffy-Muhoray, P. J. Shannon, and S. T. Sun, Nature (London) **377**, 43 (1995).

[6] D. Fedorenko, K. Slyusarenko, E. Ouskova, V. Reshetnyak, K. R. Ha, R. Karapinar, and Y. Reznikov, Phys. Rev. E **77**, 061705 (2008).

[7] I. Jánossy, K. Fodor-Csorba, A. Vajda, and L. Palomares, Appl. Phys. Lett. **99**, 111103 (2011).

[8] M. Dumont, in *Photoactive Organic Materials, Science and Applications*, edited by F. Krajzar, V. M. Agranovich, and C. Y. Lee, NATO Advanced Studies Institute, Series: High Technology (Kluwer, Dordrecht, 1996), Vol. 9, p. 501.

[9] Alexei D. Kiselev, V. G. Chigrinov, S. V. Pasechnik, and A. V. Dubtsov, Phys. Rev. E **86**, 011706 (2012).

[10] I. Jánossy, A. Vajda, T. Paksi, and T. Kosa, Mol. Cryst. Liq. Cryst. **359**, 157 (2001).

[11] P. Palffy-Muhoray, T. Kosa, and E. Weinan, Appl. Phys. A **75**, 293 (2002).

[12] D. Kinderlehrer and M. Kowalczyk, Discrete Contin. Dyn. Syst. B **11**, 153 (2009).

[13] H. S. Carslaw and J. C. Jaeger, *Conduction of Heat in Solids*, 2nd ed. (Oxford University Press, Oxford, 1959), Sec. 3.5.

[14] M. Vilfan, I. D. Olenik, A. Mertelj, and M. Copic, Phys. Rev. E **63**, 061709 (2001).