# Nematic director slippage: Role of the angular momentum of light

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We propose a theoretical model of the light-induced director slippage effect. In this effect the bulk director reorientation contributes to the surface director reorientation. It is found that the director and ellipticity profiles, obtained in the geometric optics approximation, are dependent on the ellipticity of the incident light wave. The director distribution is spatially modulated in linearly polarized light but grows monotonically in circularly polarized light. The surface director deviation has been examined, and comparison made with existing experimental data, which then permits the magnitude of the orientational nonlinearity coefficient to be calculated.

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## I. INTRODUCTION

Light-induced reorientation effects in liquid crystals are important both for fundamental reasons and because of the possibility of using these effects in applications [1]. In the majority of cases, the director reorientation occurs in the cell bulk and is imposed by the optical torque. This torque can originate either from: orientational nonlinearity [2], or from photorefractive effects [3,4], or as a result of photoinduced nonlinearity in dye-doped liquid crystals (LCs) [5–10].

Light-induced reorientation effects have usually been observed in liquid crystal cells with strong anchoring. By strong anchoring we imply that the director reorientation occurs in the bulk of liquid crystal and is negligible at the aligning surface. Development of new aligning materials providing weak anchoring [11–13] has allowed the observation of surface director reorientation.

An observation of such a reorientation, referred hereafter as a slippage effect, was reported by Marusii *et al.* [14] in a combined liquid crystal cell. One of the cell surfaces with strong planar anchoring provided homogeneous alignment of the nematic director parallel to the substrates. The other surface had rather small azimuthal anchoring. The liquid crystal was doped with azo-dye in order to increase the orientational nonlinearity coefficient, due to photoinduced trans-cis isomerization of azo-dye molecules [10,15]. A linearly polarized laser beam, propagating through the birefringent mixture, induced a bulk torque accompanied by director reorientation in the cell bulk and at the weak anchoring surface. The surface director reorientation was observed as a change in the polarization state of a probe laser beam propagating from the side of the strongly anchoring surface.

Recently Francescangeli *et al.* [16] found that the slippage effect competes with a the light-induced anchoring effect discovered earlier by Voloshchenko *et al.* [17]. At the same time, was noticed that this effect is sensitive to the polarization of the laser beam [18,19].

The key element of the present paper is to combine the physics of surface director reorientation with the use of light-induced bulk director reorientation. The reorientation occurs in a cell in which the director rotates in an elliptically polarized laser beam, subject to boundary conditions which constrain the surface director to remain in the plane of the cell. Light which propagates through a linear birefringent medium in this geometry changes its polarization state in a periodic way, with wavelength $\lambda/(n_e-n_o)$. In the course of one wavelength, the ellipticity of the light changes periodically between $-1$ and 1, as the light alternates between linear and circular polarizations. A liquid crystal, although no longer strictly a linear birefringent medium, in that there is a feedback on the director from the local fields, nevertheless conserves this property except for very intense light. As a consequence, the field-induced torque on the liquid crystal director is also spatially modulated and periodic. However, if the nonlinear feedback increases, the ellipticity spatial modulation is accompanied by a director reorientation. The reorientation of the director in the cell bulk immediately manifests itself as a director reorientation on the surface, if the surface anchoring is weak.

Since the director torque compensates the deposition of the angular momentum of light into the liquid crystal, the director reorientation depends significantly on the polarization state of the incident light beam. As a result, surface director reorientation is also sensitive to the polarization of the incident light beam.

The paper is organized as follows. In Sec. II we solve the self-consistent problem of propagation of monochromatic light wave in the planar liquid crystal cell. We use approximations in the spirit of geometric optics to obtain the director distribution in the cell. Then in Sec. III we compare existing experimental results to the theoretical predictions and estimate the photoinduced nonlinearity coefficient. Finally, in Sec. IV, we present some brief conclusions.

## II. DIRECTOR AND ELLIPTICITY PROFILES

Consider a nematic liquid crystal cell of thickness $L$ confined between the planes $z=0$ and $z=L$ of a cell uniform in the $xy$ plane. Let the director $\mathbf{n}=(\cos\varphi,\sin\varphi,0)$ describe the average molecular orientation in the cell. We suppose $\varphi=\varphi(z)$ and only consider the equilibrium orientation of the director, i.e., the angle $\varphi$ does not depend on time.

 

Let the elliptically polarized monochromatic light wave, with wave number $\mathbf{k}=k\mathbf{z}$, ellipticity $e_0$, and major ellipse axis making an angle $\alpha$ with the $x$ axis, be normally incident on the cell from the side of the surface with weak anchoring. Note that, because the light is propagating in an inhomogeneous anisotropic medium, the Poynting vector and the light polarization both vary in space throughout the sample.

The light illumination produces a bulk torque acting on the liquid crystal. The director distribution is determined by the balance between optical, elastic, and surface torques. To calculate the director configuration $\mathbf{n}(z)$, it is necessary to write down an effective free energy density $f$, which can then be minimized with respect to the director profile. This is

$$f=f_{\mathrm{el}}+f_{\mathrm{opt}}, \tag{1}$$

where $f_{\mathrm{el}}=\frac{1}{2}K_{22}(\partial\varphi/\partial z)^2$ is the nematic liquid crystal elastic energy and $K_{22}$ is the twist elastic constant.

To obtain an expression for the electromagnetic energy density $f_{\mathrm{opt}}$ we suppose the nematic liquid crystal to be a slowly varying uniaxial medium, so that $\varphi$ varies appreciably only over a length much greater than the optical wavelength $\lambda$. We also assume that the birefringence of the medium is small, i.e., $\Delta n=n_e-n_o\ll 1$. In this case the light polarization also varies slowly through the medium, and we can use the geometrical optics approximation (GOA) to solve Maxwell's equations for the field in the cell [20]. We shall also suppose the medium to be non-absorbing. In this case the beam intensity $I$, defined as the $z$ component of the average Poynting vector, remains constant. The approach we shall follow was developed by Santamato *et al.* [23,24]. We first remind the reader of the elements of this theory.

The total electromagnetic energy density $f_{\mathrm{opt}}$ of the light wave in a non-absorbing, nonmagnetic medium can be rewritten in terms of the light intensity $I$, ellipticity $e$, and major axis angle $\psi$. In the limit of low birefringence this takes the form

$$f_{\mathrm{opt}}=-\frac{In_0}{c}-\frac{I\Delta n}{2c}[1+(1-e^2)^{1/2}\cos 2(\psi-\varphi)], \tag{2}$$

where $I$ is the $z$ component of Poynting vector, or equivalently, the total intensity of the light wave.

The ellipticity $e$ and the major axis angle $\psi$ can be defined in terms of the Stokes parameters $\{S_i\}$, where

$$\begin{aligned} S_0&=|E_x|^2+|E_y|^2,\\ S_1&=|E_x|^2-|E_y|^2,\\ S_2&=2\,\mathrm{Re}(E_x^*E_y),\\ S_3&=2\,\mathrm{Im}(E_x^*E_y). \end{aligned} \tag{3}$$

Then the ellipticity, defined by $e=S_3/S_0$, is the ellipticity of the polarization ellipse of the light in the cell, and $2\psi=\arctan(S_2/S_1)$ is the angle which the major axis of the polarization ellipse forms with the $x$ axis.

Equations governing the polarization of light can be derived from the electromagnetic energy density $f_{\mathrm{opt}}$ by considering $f_{\mathrm{opt}}$ as a Hamiltonian function. Then $\psi$ is a generalized coordinate and $l_z=-(I/\omega)e$ is conjugate momentum, where $l_z$ is the average angular momentum carried by the optical beam along the propagation direction.

These equations are

$$\frac{\partial\psi}{\partial z}=-\frac{\omega}{2c}\Delta n\frac{e}{(1-e^2)^{1/2}}\cos 2(\psi-\varphi), \tag{4}$$

$$\frac{\partial e}{\partial z}=\frac{\omega}{c}\Delta n(1-e^2)^{1/2}\sin 2(\psi-\varphi). \tag{5}$$

The equilibrium orientation of the molecular director is given by the minimizer of the functional $\int_0^L f\{\varphi(z)\}dz$ subject to fixed field intensity $I$ and polarization state $(e,\psi)$.

Instead of deriving variational equations it is more useful to use the conserved quantities corresponding to the Lagrange density $f$. This does not depend explicitly on the $z$ coordinate. Noether's theorem then implies that there is a conserved integral analogous to the energy in classical dynamics. Thus the first integral for this problem can be obtained directly from the free energy density (1):

$$\frac{K_2}{2}\left(\frac{\partial\varphi}{\partial z}\right)^2+\frac{I\Delta n}{2c}(1-e^2)^{1/2}\cos 2(\psi-\varphi)=E. \tag{6}$$

There is also another conserved integral which can be derived from this free energy. This is the total angular momentum flux along the propagation direction, and is the sum of the elastic and optical angular momenta

$$K_2\frac{\partial\varphi}{\partial z}+\frac{I}{\omega}e=M. \tag{7}$$

Eliminating the director angle in Eq. (6), using Eqs. (4) and (7), yields the following equation for the polarization ellipse:

$$\left(\frac{\partial e}{\partial z}\right)^2=\left(\frac{\omega\Delta n}{c}\right)^2(1-e^2)-\left[E-\frac{1}{2K_2}\left(M-\frac{Ie}{\omega}\right)^2\right]^2\left(\frac{2\omega}{I}\right)^2. \tag{8}$$

This is the fundamental equation governing the distribution of ellipticity in the liquid crystal cell. Santamato *et al.* [23] then used it to discuss the optical Fredericksz transition in a planar cell. They observed, in addition, that this equation can in general be solved in terms of elliptic integrals.

We now turn to the application of the equation (8) to our geometry. We note that the physical structure of the general solutions strongly depends on the constants $M$, $E$, which in turn depend on boundary conditions in a rather complicated way. However, for the cell geometry in which the slippage effect has been observed, considerable simplifications are possible.

The boundary conditions involve a fixed director orientation at $z=L$, and a free but planar boundary at $z=0$:

$$\varphi|_{z=L}=0,\left.\frac{\partial\varphi}{\partial z}\right|_{z=0}=0. \tag{9}$$

Note that the director can slip over the surface with weak anchoring—no surface torque prevents it from the reorientation. As a result, the weak anchoring maximizes the response of the director to a bulk field.

The incident light wave is elliptically polarized and initially incident on the surface with zero anchoring:

$$e|_{z=0}=e_0,\psi|_{z=0}=\alpha. \tag{10}$$

In this geometry the constants $E$ and $M$ in Eq. (8) can be easily determined from the boundary conditions:

$$M=(I/\omega)e_0, \tag{11}$$

$$E=\frac{I}{c}\left[n_0+\frac{\Delta n}{2}[1+(1-e_0^2)^{1/2}\cos 2(\alpha-\varphi_0)]\right],$$

where $\varphi_0=\varphi(z=0)$. Then, Eq. (8) for the ellipticity and the director (7), simplifies to

$$\left(\frac{\partial e}{\partial s}\right)^2=1-e^2-[(1-e_0^2)^{1/2}\cos 2(\alpha-\varphi_0)-r(e_0-e)^2]^2, \tag{12}$$

$$\frac{\partial \varphi}{\partial s}=r(e_0-e), \tag{13}$$

where $s=(\omega\Delta n/c)z=2\pi\Delta n(z/\lambda)$. We have also introduced the dimensionless parameter $r$, which is proportional to the light intensity and defined by

$$r=Ic/(\Delta n K_{22}\omega^2). \tag{14}$$

We now consider the two distinct cases, of linearly and circularly polarized incident light waves.

### A. Linear polarization of the incident light

Zero ellipticity of the linearly polarized light implies that the light wave does not carry angular momentum, since the latter is proportional to the ellipticity.

The initial conditions for the polarization state $e$ read as

$$e_0=0,\ \ \psi\,|_{z=0}=\alpha, \tag{15}$$

and the equation for the ellipticity (12) simplifies to

$$\left(\frac{\partial e}{\partial s}\right)^2=1-e^2-[\cos 2(\alpha-\varphi_0)-re^2]^2. \tag{16}$$

Substitution of $y=re^2$ allows (16) the standard integral for the elliptic functions [25]. After integration, we obtain

$$e_{\rm lin}(s)=e_0\,{\rm sd}(\kappa s) \tag{17}$$

where $e_0=m'\sqrt{e_p/r}$, $\kappa=\sqrt{e_p r}/m$, $m'=\sqrt{1-m^2}$, $m^2=e_p/(e_p-e_n)$, $e_{p,n}$ are the positive and negative roots of the equation $1-e^2-[\cos 2(\alpha-\varphi_0)-re^2]^2=0$ correspondingly, ${\rm sd}(x)={\rm sn}(x)/{\rm dn}(x)$, where sn, cn, dn are Jacobi elliptic functions of index $m$.

The director distribution can be obtained by integrating (7) and has the form

$$\varphi_{\rm lin}(s)={\rm arccot}\left(\frac{m}{m'}{\rm cn}[\kappa s_L]\right)-{\rm arccot}\left(\frac{m}{m'}{\rm cn}[\kappa s]\right), \tag{18}$$

where $s_L=(\omega\Delta n/c)L$.

The estimation of the typical experimental values for $r$ will be done later. We now find the solution for $r\ll 1$.

As it is seen from the angular momentum conservation (7), to obtain the director distribution in linear order in the dimensionless light intensity $r$ we need to know the ellipticity $e$ in *zero* order in $r$. The latter can be found by solving Eq. (16) putting $r=0$:

$$e_{\rm lin}(s)=\sin(2\alpha)\sin(s). \tag{19}$$

Then the equation for the director (7) can be solved using elementary methods, yielding

$$\varphi_{\rm lin}(0)=2r\sin(2\alpha)\sin^2(s_L/2). \tag{20}$$

The ellipticity distribution and the director deviation given in Eqs. (19) and (20) might, in principle, have been obtained by assuming *a priori* that the ordinary and extraordinary light waves simply follow the distribution of the director. Equivalently, one can say that in the low $r$ limit the light beam propagates through the cell in the adiabatic or Mauguin regime [26]. In the adiabatic approach one can write the expression for the electric field components explicitly since the ordinary and extraordinary light waves follow the distribution of the director. Then this field is used as an external field which reorients the director. However, such an assumption is not theoretically satisfactory, and naive use of this approximation as an initial hypothesis can lead to internally inconsistent approximations [27].

Let us now summarize the important properties of the light propagation and the director distribution in the planar cell. Equation (19) yields that the polarization ellipse $e(z)$ is a periodic function with spatial period $l=\lambda/\Delta n$. In more general case (17) the spatial period depends on the light intensity, $l=4K(m)(\kappa\omega\Delta n/c)^{-1}$, where $K(m)$ is the complete elliptic integral of the first kind.

The liquid crystal director $\varphi(z)$ follows the distribution of the ellipticity with the same spatial period, and is shown in Fig. 1. It is seen that, in spite of the modulation, the average deviation of the director tends to be along the polarization of the incident light and increases with the light intensity.

We now make some comments on the director deviation $\varphi_0$ at $z=0$. First, Eq. (20) shows that the surface director is sensitive to the sign of the birefringence $\Delta n$. If $\Delta n>0$, the director tends to be parallel to the polarization direction. It reorients perpendicular to the polarization direction if $\Delta n<0$.

Second, in the limit of small intensities, $r\ll 0$, the liquid crystal response is maximal at $\alpha=\pi/4$, and does not occur for $\alpha=0,\pi/2$ since we are below the threshold of Freedericksz transition. However, in the general case, the value of $\alpha$ which provides maximal director response depends on the

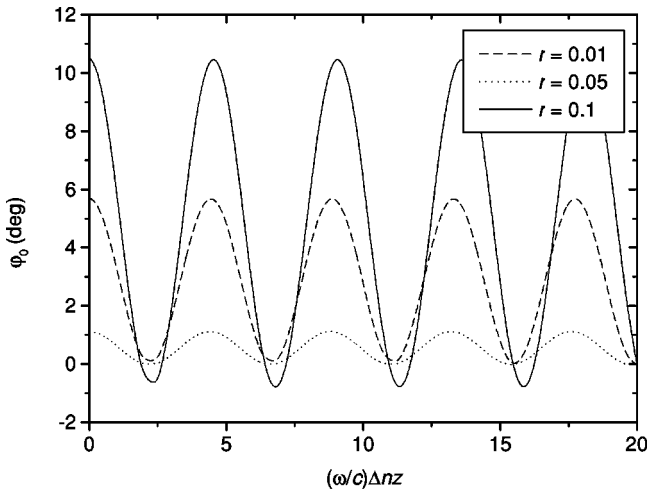


FIG. 1. Director distribution in the cell for different values of the dimensionless light intensity $r$. Linear polarization of the incident light. The director angle varies periodically through the cell thickness following the change in the light ellipticity.

light intensity. For bigger values of the parameter $r$ the maximal response of the director occurs at an angle $\alpha$ larger than $\pi/4$ (Fig. 2).

The most important comment is that the director deviation depends strongly on the cell thickness $L$ and refractive index difference $\Delta n$. Indeed, $\omega \Delta n L/(2c) = \pi \Delta n L/\lambda$, and for typical experimental conditions $L/\lambda \approx 100$. Thus a change in the refractive index $\Delta n \sim 10^{-2}$ leads to a considerable change in the director deviation $\varphi_0$. In practice this means that changes in the refractive index due to the laser-induced heating of the liquid crystal [21] or molecular phototransformations [22] influence the amplitude of the surface director deviation. This also means that the cell thickness should be tuned rather precisely in order for the surface director orientation to be observed. The optimal condition is that $\Delta n L/\lambda = N/2$, where $N$ is an integer.

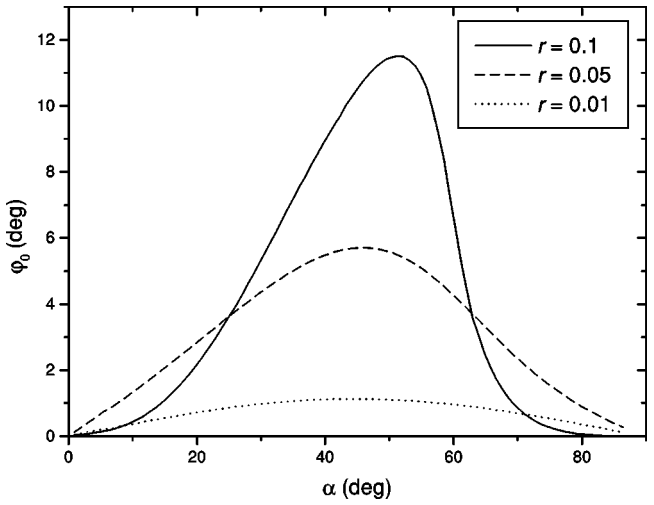


FIG. 2. Director deviation on the isotropic surface as a function of the angle between rubbing direction and polarization of the exciting light. The director response has a maximum at $\alpha = \pi/4$ and is absent at $\alpha = 0, \pi/2$ for small light intensities.

### B. Circular polarization of the incident light

Contrary to the case of linear polarization, circularly polarized light wave carries angular momentum along the propagation direction. The initial conditions for the polarization state $e$ are now

$$e_{z=0} = 1. \tag{21}$$

The boundary conditions for the director are the same as for the linearly polarized light (9).

The equation for the ellipticity then simplifies to

$$\left(\frac{\partial e}{\partial s}\right)^2 = 1 - e^2 - r^2[1-e]^4. \tag{22}$$

Substitution $y = 1 - e$ allows integration of this equation in terms of elliptical integrals [25]. After integration we get the distribution of the ellipticity in the cell:

$$e_{\mathrm{cir}}(s) = 1 - e_r \frac{q[1 - \mathrm{cn}(\kappa s)]}{p + q + (p-q)[1 - \mathrm{cn}(\kappa s)]} \tag{23}$$

where $e_r$ is the only real root of the equation $2 - e - r^2 e^3 = 0$, $p^2 = r^{-2} + 3e_r^2$, $q^2 = r^{-2} + e_r^2$, $\kappa = r\sqrt{pq}$. The polarization ellipse $e_{\mathrm{cir}}(s)$ has the same dependence on the spatial coordinate as we have in the case of linear polarization (17), i.e., is a periodic function with the same spatial period.

Now we turn to the director distribution. Integrating Eq. (7) we obtain,

$$\varphi(s) = \varphi_0 + \frac{e_r}{p-q}\left\{ 2 \arctan\left[ \sqrt{\frac{q}{p}} \tan\left(\frac{\kappa s}{2}\right)\right] - \sqrt{\frac{q}{p}} \kappa s \right\}. \tag{24}$$

To perform the integration we used an approximation $\mathrm{cn}(x) \simeq \cos(x)$, which is valid for small enough elliptic function indices or alternatively small enough values of $r$.

The expressions (23) and (24) in leading order in $r$ are

$$e_{\mathrm{cir}}(s) = \cos(s), \tag{25}$$

$$\varphi_{\mathrm{cir}}(0) = r[s_L - \sin(s_L)]. \tag{26}$$

Let us now analyze the ellipticity and the director profile in the cell. Equation (25) yields that polarization ellipse is exactly the same as for the linearly polarized light. The only change is that, in order to match the condition $e_{\mathrm{cir}}(0) = 1$ instead of $e_{\mathrm{lin}}(0) = 0$, we have to have cosine dependence on $s$ [cf. Eq. (19)].

However, the liquid crystal director is no longer a periodic function of the cell thickness. Even more, now it is monotonically growing function of $s$ (Fig. 3). This fact can be easily proved if one considers the angular momentum conservation (7). For the linear polarization it reads: $\partial\varphi/\partial s = -re$. Since $e(s)$ is a periodic function of $s$, $\varphi$ is also a periodic function of $s$ with the same spatial period. However, for circular polarization we have $\partial\varphi/\partial s = r(1-e) \geqslant 0$. Therefore, $\varphi$ grows monotonically with $s$. In fact, the director distribution is a superposition of monotonically growing

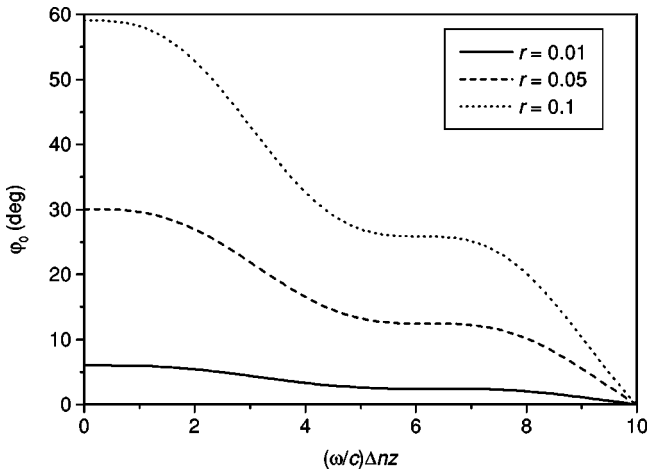


FIG. 3. Director distribution in the cell for different values of the dimensionless light intensity $r$. Circular polarization of the incident light. The director angle increases monotonically through the cell thickness.

and oscillating functions, since the ellipticity of the polarization ellipse is a periodic function of $s$.

It is also seen from Eq. (26) that the director deviation $\varphi_0$ at $z=0$ consists of two parts. The first part, $r\sin(s_L)$, originates from the ellipticity modulation. The other part, $-rs_L$, is due to the constant injection of light angular momentum into the cell bulk. For typical experimental conditions this contribution dominates, since $s_L=2\pi\Delta nL/\lambda \gg 1 > \sin(s_L)$. For the same reasons $\varphi_0^{\rm cir}\simeq 2\pi\Delta nL/\lambda\,\varphi_0^{\rm lin}$. Therefore, the surface director deviation in circularly polarized light is $2\pi\Delta nL/\lambda$ times bigger than in the linearly polarized light. There are even more interesting observations.

First, the resulting twist angle $\varphi_0$ is not sensitive to the variation of $\Delta n$ for typical experimental conditions ($r\ll 1$) and liquid crystal cell parameters ($s_L\gg 1$). This is very different from the situation with linearly polarized light wave. Second, $\varphi_0$ is proportional to the cell thickness $L$, i.e., in thick enough cells, its value can exceed $\pi$ and one can get supertwisted distribution of the director or even distribution that realizes in chiral nematics.

### C. Elliptically polarized light

Now we try to solve Eqs. (12) and (13) for the case of elliptically polarized light. Solving these equations for arbitrary value of $r$ is a complicated task. However, as one can see from the angular momentum conservation (7), to obtain the director distribution in linear order in the dimensionless light intensity $r$ we need to know the ellipticity $e(s)$ in zero order in $r$. Solving Eq. (12) in this limit we obtain that $e(s)$ is a superposition of the solutions $e_{\rm lin}(s)$ and $e_{\rm cir}(s)$:

$$e(s)=\sqrt{1-e_0^2}\sin(2\alpha)\sin s+e_0\cos s = \sqrt{1-e_0^2}e_{\rm lin}(s)+e_0e_{\rm cir}(s). \quad (27)$$

Correspondingly, in the leading order in $r$, the director deviation at $z=0$ is also a superposition of $\varphi_0^{\rm lin}$ and $\varphi_0^{\rm cir}$ and has the form

$$\varphi_0=r[\sqrt{1-e_0^2}\sin(2\alpha)(1-\cos s_L)+e_0(\sin s_L-s_L)] = \sqrt{1-e_0^2}\varphi_0^{\rm lin}+e_0\varphi_0^{\rm cir}. \quad (28)$$

It is seen from Eq. (28) that, for circularly polarized light wave, the director at the surface rotates according to the sign of the ellipticity: clockwise polarization leads to the clockwise reorientation of the director and vice versa.

## III. DISCUSSION

We now turn to the estimation of the parameter $r$ and the director deviation angles one can expect for typical experimental conditions. Quantitatively, in typical experiments, we have light intensities $I<10$ W/cm$^2$, and liquid crystal constants $\Delta n=0.1$, $K_{22}=3\times 10^{-7}$ dyn. With these fundamental parameter values, the dimensionless intensity $r<0.001 \ll 1$, which is fairly small. The maximal deviation of the director which can be observed under these circumstances is far less than $0.1^\circ$ for linearly polarized light and is less than $1^\circ$ for circularly polarized light. Such reorientations can hardly be detected real experiments.

However, the situation is different for the dye-doped liquid crystals. All previous experimental results [5,6] and theoretical predictions [8,30] are consistent with the following expression for the dye contribution to the free energy density,

$$f_{\rm dye}=\eta f_{\rm opt}, \quad (29)$$

where $f_{\rm opt}$ is the total electromagnetic energy density of the incident light wave (2) and the parameter $\eta$ characterizes the efficiency of the dye-induced director reorientation. The parameter $\eta$ is proportional to the dye concentration, but also depends on the molecular structures of both dye and liquid crystal.

Equation (29) implies that the effective torque imposed by the light wave increases proportionally to $\eta$. In typical experiments, we obtain a dye assisted nonlinearity coefficient $|\eta|\simeq 200$ [7]. This gives $r\simeq 0.1$ and typical director deviations about $10^\circ$, which can be measured experimentally without any difficulties.

Our theoretical framework also permits the evaluation of the photo-induced orientational nonlinearity coefficient $\eta$ from the existing experimental data. $\eta$ is the crucial phenomenological parameter which governs the interaction between light and the liquid crystal/azo-dye mixture. This quantity is an input parameter in our theory, and must either be measured or calculated using a microscopic theory [8,30].

Trying to estimate $\eta$ we are fully aware that the cell is absorbing, since the effect of the photoinduced nonlinearity is due to the absorption of light by the dye molecules. For the absorbing media, we are no longer able to apply the Lagrange approach, which involves energy and angular momentum conservation. Therefore, estimating coefficient $\eta$ we are neglecting effects related to the light absorption. For the linearly polarized incident light, the value of $\eta$ can be deter-

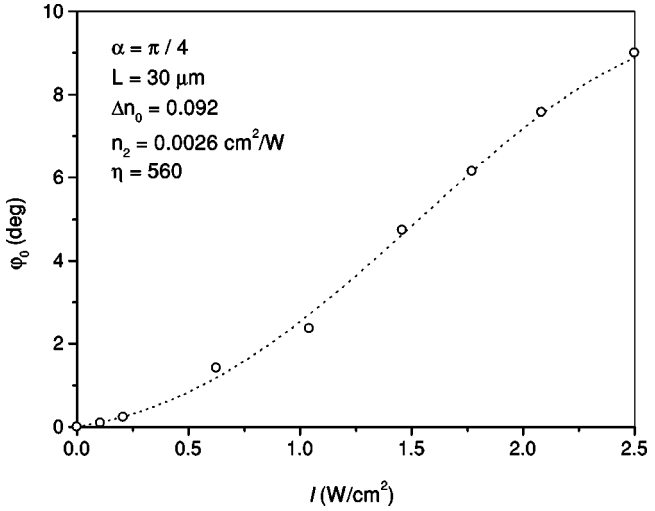


FIG. 4. Experimental data and the best fit of the director angle $\varphi_0$ as a function of the exciting laser beam intensity ($\alpha=\pi/4$). Fitting parameters: photoinduced nonlinearity coefficient $\eta$ and refractive index of the mixture $\Delta n_0$.

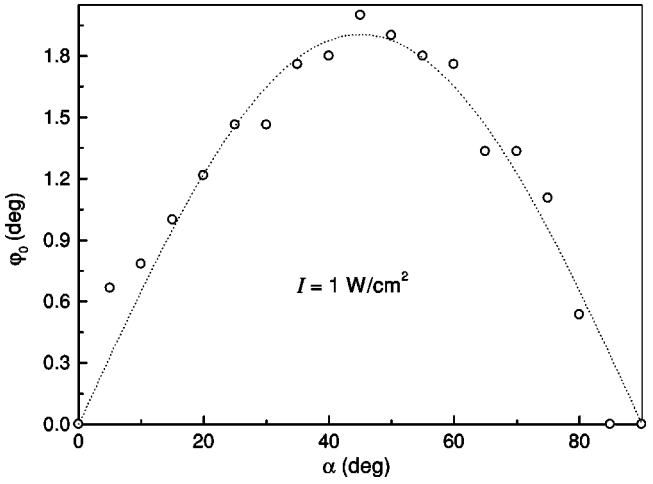


FIG. 5. Director deviation on the isotropic surface as a function of the angle between rubbing direction and polarization of the exciting light. The director response has a maximum at $\alpha=\pi/4$ and is absent at $\alpha=0,\pi/2$.

mined applying Eq. (20), which connects $\varphi_0$ to the dimensionless intensity $r$, to the experimental results presented in [14].

Typical experiments were performed in the cell with one isotropic surface and one surface consisting of a substrate covered with a rubbed polyimide film. The cell was filled with the nematic liquid crystal 4-*n*-pentyl-4-cyanobiphenyl (5CB) doped with the azo-dye methyl-red (MR) at weight concentration 0.1. The mixture was exposed from the side of the isotropic surface by a linearly polarized beam of a c.w. $Ar^+$ laser, irradiation wavelength of which $\lambda=488$ nm is near the maximum of the absorption band of MR dissolved in 5CB. Typical intensity of the $Ar^+$ laser was in the range 0.1–1 W/cm$^2$. Its polarization vector was set at 45° with respect to the initial director orientation. The cell response was observed with a probe He-Ne laser beam with wavelength $\lambda=638$ nm which is in the region of MR transparency.

Irradiation with a beam of a $Ar^+$ laser led to the director reorientation in the liquid crystal bulk and its slippage on the isotropic surface. The latter was detected as a rotation of the polarization of the probe beam. The experimental dependence of the rotation angle on the intensity of the incident light is presented in Fig. 4.

Equation (20) predicts that the surface director reorientation angle $\varphi_0$ will be a function of the laser intensity $I$ and refractive index anisotropy $\Delta n=n_e-n_o$. The birefringence of the dye-liquid crystal mixture can in principle also depend on the exciting light intensity indirectly both as a result of laser-induced heating of the liquid crystal [21], and because molecular phototransformation can change the dipole moment of the phototransformed molecules [22]. The intensity $I$ thus has a direct and an indirect effect on the reorientation of the director. In order to interpret the results correctly it is necessary to correct for the indirect effects. To do this we studied the dependence of the dye-liquid crystal birefringence $\Delta n$ on the exciting light intensity independently using a light-induced birefringence technique [22]. We found that in our case,

$$\Delta n=\Delta n_0+n_2 I, \tag{30}$$

where $n_2\simeq 0.0026\ (\mathrm{W/cm^2})^{-1}$.

Then, fitting of the data (Fig. 4) yields $\eta=-560$. The photo-induced orientational nonlinearity is considerably enhanced even with respect to the giant orientational nonlinearity of pure liquid crystals [2] and is consistent with what might be expected as a result of measurements in related systems [9,15] using the *z*-scan technique.

The theory developed in Eq. (20) also requires that the director response to the laser illumination light intensity be linear. In order to check whether this is the case, we measured the dependence $\varphi_0(\alpha)$. The results of measurements for $I=1$ W/cm$^2$ are presented in Fig. 5. We find, in agreement with Eq. (20), that the liquid crystal response is maximal at $\alpha=\pi/4$ and absent at $\alpha=0,\pi/2$.

As far as we are aware, there have been no direct observations of the slippage effect for circularly or elliptically polarized light. The effect has probably been implicitly observed in azo-dye doped liquid crystal cells [18,19]. In these experimetns it was found that when a cell is irradiated by circularly polarized light, an easy orientation axis is induced on an initially isotropic aligning surface. The direction of this axis correlated with the direction of the polarization rotation. The appearance of the easy axis can be explained as follows. First there is a light-induced surface director slippage. The director then freezes as a result of light-induced adsorption of the dye molecules on the aligning surface. By contrast, this reorientation cannot be explained if we use a model which depends only on the selective adsorption of the dye molecules on the aligning surface.

## IV. CONCLUSIONS

In this paper we have tried to interpret existing experimental facts on the surface director reorientation by assum-

ing the dye-induced torque to be proportional to the optical torque, and treating the nonlinear interaction between the light wave and the mixture in the geometrical optics approximation.

We now turn to a brief discussion of the implications of this work. We believe that the experimental technique we introduce in this paper can be used in further studies of the liquid crystal interface. Relatively straightforward extensions of the technique will, for example, permit measurements of fundamental surface parameters such as in-plane surface viscosities and weak anchoring coefficients.

There are also possible applications of the method involving the writing of the dynamic holographic polarization gratings. Furthermore, the present investigation, together with our results published earlier [16,17,28,29] have shown that it is possible to control the light-induced alignment memory effect by changing the concentration of azo-dye in the liquid crystal. There may also be scope for systematic investigations of surface memory effects, the basic mechanisms of which are still little understood.

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