Optically induced twist Fréedericksz transitions in planar-aligned nematic liquid crystals

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The optical-field-induced Fréedericksz transition for a twist deformation by a normally incident laser beam in a planar-aligned nematic liquid crystal is studied. The Euler equation for the molecular director and the equations describing the evolution of the beam polarization in the birefringent medium are solved simultaneously in the small-perturbation limit. The stability of the undistorted state is investigated. An alternate series of stable and unstable bifurcations is found. This phenomenon has no analog in the Fréedericksz transition induced by dc electric and magnetic external fields.

In recent years, nonlinear interaction between a normally incident linearly polarized light beam and a homeotropically oriented nematic liquid-crystal film has received a great deal of attention.[1] The existence of a characteristic threshold intensity for induced molecular reorientation was experimentally well demonstrated.[2] The underlying physical mechanism for such an effect, known as the optical Fréedericksz transition, is essentially the same as in the corresponding dc Fréedericksz transition.[3] The geometry dictates that the polarization of the light beam remains unchanged in traversing the cell, even with molecular reorientation. There are a number of other dc Fréedericksz transitions with different geometries to which one may also find optical analogue. In most cases, however, the underlying physical processes of the dc- and optical-field-induced transitions are very different because the beam polarization varies in propagating through the medium. In this paper, we consider optical Fréedericksz transition in a planar-aligned cell induced by a light beam linearly polarized in a direction perpendicular to the molecular alignment. The induced molecular reorientation yields a change of birefringence seen by the beam, and consequently, the beam polarization changes continuously as it propagates through the cell. We must consider, simultaneously, the local action of the *elliptically polarized* light beam on the liquid-crystal molecules and the change of the beam polarization due to birefringence arising from molecular reorientation. It turns out that even the threshold behavior for the induced transition is characteristically unique. This is what we would like to focus on in the present paper. The transition is second order. The present case shows little resemblance to the corresponding dc case. The characteristics of our case are intimately related to the exchange of angular momentum between the light beam and the liquid-crystal medium.

Let us consider a planar nematic cell of thickness L, with the molecular alignment originally along $\hat{\mathbf{x}}$. A light beam, linearly polarized along $\hat{\mathbf{y}}$, is normally incident on the cell along $\hat{\mathbf{z}}$. If the optical field is sufficiently strong to reorient the liquid-crystal molecules, then the local molecular orientation is described by the unit director $\hat{\mathbf{n}}=(\cos\phi,\sin\phi,0)$, and the local beam polarization is described by the Stokes unit vector $\hat{\mathbf{s}}=(s_1,s_2,s_3)$ on the Poincaré sphere. Here, both $\hat{\mathbf{n}}$ and $\hat{\mathbf{s}}$ are functions of z and time t. The quantity s_3 denotes the polarization ellipticity: $s_3=(I_R-I_L)/(I_R+I_L)$, I_R and I_L being the intensities of the right- and left-handed circular components of the beam, respectively.

It has been shown that the evolution of the beam polarization of an optical wave propagating in an inhomogeneous uniaxial medium is approximately governed by the precession equation[4,5]

$$\partial\hat{\mathbf{s}}/\partial z=\mathbf{\Omega}\times\hat{\mathbf{s}}\ , \tag{1}$$

where, in the present case,

$$\mathbf{\Omega}=(2\pi/\lambda)\Delta n\ (\cos(2\phi),\sin(2\phi),0)\ ,$$

λ is the optical wavelength and $\Delta n=n_e-n_0$ is the difference between the extraordinary and the ordinary refractive indices of the medium. Equation (1) can be derived from Maxwell's equations in the slowly-varying envelope approximation in the limit of low birefringence $\Delta n/n$. For higher birefringence, one needs a more rigorous approach, in which the beam polarization should be described by a set of pseudo-Stokes parameters.[5] For conventional liquid crystals, $\Delta n/n\simeq 0.15$, and Eq. (1) is sufficiently accurate.

According to the angular momentum conservation law, the angular momentum lost by the light beam per unit time in traversing the medium must be equal to the optical torque exerted on the liquid-crystal molecules. The angular momentum carried by a beam with intensity I and ellipticity S_3 is $(IS_3/\hbar\omega)\hbar=(I\lambda/2\pi c)S_3$ per unit area and unit time. Then, the angular momentum per unit volume and unit time lost by the light beam between planes z and $z+dz$ is $(I\lambda/2\pi c)\partial S_3/\partial z$, which is also the optical torque per unit volume acting on the

molecules. With the inertia term neglected, the equation of motion for the molecular reorientation is obtained by equating the optical torque to the viscous and elastic torques:

$$-\gamma_1(\partial\phi/\partial t)+k_{22}(\partial^2\phi/\partial z^2)+(I/c)(\lambda/2\pi)(\partial S_3/\partial z)=0 , \tag{2}$$

where γ_1 is the viscosity coefficient and k_{22} the elastic constant for twist.

Equations (1) and (2) form a set of coupled nonlinear partial differential equations for the quantities $\hat{\mathbf{s}}$ and ϕ, which must be solved with the initial and boundary conditions

$$\begin{aligned}&\phi(z,0)=0 ,\\ &\phi(0,t)=\phi(L,t)=0 ,\\ &\hat{\mathbf{s}}(0,t)=\hat{\mathbf{s}}_0 ,\end{aligned} \tag{3}$$

where $\hat{\mathbf{s}}_0$ is the Stokes vector for the input light. For linear polarization along the y axis, we have $\hat{\mathbf{s}}_0=(-1,0,0)$. It is easily seen that $\hat{\mathbf{s}}=\hat{\mathbf{s}}_0=(-1,0,0)$ and $\phi=0$ is a solution of Eqs. (1) and (2).[6] This corresponds to the propagation of an ordinary wave in the undistorted sample. This solution, however, becomes unstable when a critical intensity threshold $I_{\rm th}$ is reached. The bifurcation point can be found by using Lyapunov's first method for test of stability extended to continuous media as follows.

Equation (1) is linearized by taking ϕ, S_2, and S_3 as small quantities. With the introduction of the following normalized quantities:

$$\begin{aligned}&u=z/L ,\\ &\tau=t(k_{22}/\gamma_1 L^2) ,\\ &\tilde{I}=(I/ck_{22}\Delta n)(\lambda/2\pi)^2 ,\\ &\tilde{L}=2\pi\,\Delta n\, L/\lambda ,\end{aligned} \tag{4}$$

the *linearized* set of equations becomes, for $0\le u\le 1$,

$$\begin{aligned}&-\partial\phi/\partial\tau+(\partial^2\phi/\partial u^2)+\tilde{I}\tilde{L}(\partial S_3/\partial u)=0 ,\\ &\partial S_1/\partial u=0 ,\\ &\partial S_2/\partial u=-\tilde{L}S_3 ,\\ &\partial S_3/\partial u=\tilde{L}(S_2+2\phi) ,\end{aligned} \tag{5}$$

with $S_1\simeq S_1(0,t)=-1$. Although the set of Eq. (5) is not self-adjoint, we can still take a trial solution of the form

$$\begin{aligned}&\phi(u,\tau)=\psi(u)\exp(\alpha\tau) ,\\ &S_{2,3}=\sigma_{2,3}(u)\exp(\alpha\tau) .\end{aligned} \tag{6}$$

Insertion of Eq. (6) into Eq. (5) yields a set of equations that can be solved by the standard method for eigenvalue problems. We report here only the results of the calculation. The dimensionless eigenvalue α is obtained from the transcendental equation

$$[(x_{22}-\tilde{L}^2)/(x_1^2-\tilde{L}^2)]\sin(x_2)/x_2=\sin(x_1)/x_1 , \tag{7}$$

where $x_1(\alpha)$ and $x_2(\alpha)$ are the two roots of the biquadratic algebraic equation

$$(x^2+\alpha)(x^2-\tilde{L}^2)-2\tilde{I}\tilde{L}^2x^2=0 . \tag{8}$$

The results show that α always appears to be real instead of complex. A plot of α as a function of $\tilde{I}$ for $\tilde{L}=5$ is presented in Fig. 1. At a given intensity I, there exists a discrete set of eigenvalues α_n $(n=0,1,2,\ldots)$ with $\alpha_0\ge\alpha_1\ge\alpha_2\ge\cdots$.

According to Lyapunov's criterion for stability, the solution $\hat{\mathbf{s}}=(-1,0,0)$, $\phi=0$ is stable if, and only if, all α_n have a negative real part. This is certainly not the case here. Let $\tilde{I}_m$ $(m=0,1,2,\ldots)$ be the zeroes of $\alpha(I)$, as shown in Fig. 1. We have $\tilde{I}=\tilde{I}_m$ (m even) correspond to the points where the solution $\hat{\mathbf{s}}=(-1,0,0)$, $\phi=0$ becomes unstable with further increase of $\tilde{I}$. The lowest threshold for instability is given by $\tilde{I}_0$. As seen in Fig. 1, there is a series of alternate stable and unstable intervals as the light intensity increases. In the regions $\tilde{I}<\tilde{I}_0$ and $\tilde{I}_m^*\le\tilde{I}\le\tilde{I}_{m+1}$ (m odd) the undistorted state $\phi=0$ is stable, such that any spontaneous fluctuations will be damped out in time. For $\tilde{I}_m\le\tilde{I}\le\tilde{I}_{m+1}$ (m even), however, the eigenvalue α_0 is positive, and the fluctuations will grow exponentially. The initial growth of the instability is governed by a time constant proportional to $1/\alpha_0$. For $\tilde{I}_m\le\tilde{I}\le\tilde{I}_m^*$ (m odd), the state $\phi=0$ is also unstable, but the initial growth of the fluctuations has a double-exponential character with two time constants proportional to $1/\alpha_0$ and $1/\alpha_1$, respectively. At the points $\tilde{I}=\tilde{I}_m^*$ (m odd) the eigenvalues α_0 and α_1 coalesce. In any case, in the unstable regions, deviation from the undistorted state will grow in time, until the nonlinear terms, neglected in Eq. (5), become significant and drive the system to a steady-state saturation regime. This behavior has no analog in the corresponding dc Fréedericksz transition.

The threshold intensities $\tilde{I}_m$, at which the solution $\hat{\mathbf{s}}=(-1,0,0)$, $\phi=0$ switches from stable to unstable can be obtained as a function of $\tilde{L}$ by setting $\alpha=0$ in Eq. (8). We find $x_1=0$ and $x_2=\tilde{L}\sqrt{1+2\tilde{I}}$, so that Eq. (7) reduces to

$$2\tilde{I}_{\rm th}\left[\frac{\sin(\tilde{L}\sqrt{1+2\tilde{I}_{\rm th}})}{\tilde{L}\sqrt{1+2\tilde{I}_{\rm th}}}\right]=-1 , \tag{9}$$

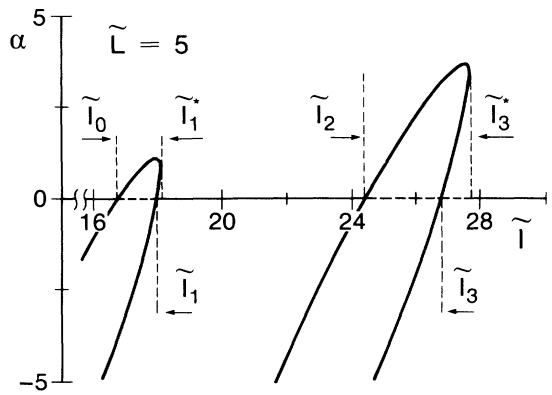

FIG. 1. Plot of the eigenvalues α vs $\tilde{I}$ for $\tilde{L}=5$ from Eq. (7). The dotted part on the abscissa denotes regions where the trivial solution $\hat{\mathbf{s}}=(-1,0,0)$, $\phi=0$ becomes unstable.

from which $\tilde{I}_{\rm th}(\tilde{L})$ can be found. This is plotted in Fig. 2. For any fixed $\tilde{L}$, we have a discrete set of values $\tilde{I}_{\rm th}=\tilde{I}_m$ ($m=0,1,2,\ldots$), corresponding to the zeros of α [for example, the zeroes of α in Fig. 1 correspond to the intersecting points of the $\tilde{L}=5$ line with the curves $\tilde{I}_{\rm th}(\tilde{L})$ in Fig. 2]. The solid curves in Fig. 2 describe $\tilde{I}_m(\tilde{L})$ with even m and the dashed curves $\tilde{I}_m(\tilde{L})$ with odd m. Only the former denotes the threshold intensities at which the system switches from stable to unstable. The lowest threshold for a given $\tilde{L}$ corresponds to $\tilde{I}_{\rm th}=\tilde{I}_0$. The result can be better understood if we know the molecular orientation (eigenfunction) associated with the present eigenvalue problem.

For a given L, we find the twisted molecular reorientation corresponding to the eigenvalue α_n to be

$$\phi_n(u)=A\left[\frac{\sin[x_1(\alpha_n)u]}{\sin[x_1(\alpha_n)]}-\frac{\sin[x_2(\alpha_n)u]}{\sin[x_2(\alpha_n)]}\right], \quad 0\le u\ (\equiv z/L)\le 1 \qquad (10)$$

where A is a constant. This eigenfunction $\phi_n(u)$ vanishes at $u=0$ and L, and exhibits a number of sinusoidal-like oscillations in between. A plot of the eigenfunction $\phi_0(u)$ is shown in Fig. 3 for $\tilde{I}$ just above the minimum threshold $\tilde{I}_0$ and $\tilde{L}=2$, 5, and 10. We see that the molecular distortion from the unperturbed state is an oscillating function having a number of zeros, which is of the order of $\tilde{L}$. This is also generally true for ϕ_n. Since $\tilde{L}$ is close to (L/λ), the number of oscillations is usually very high, unless very thin samples are used. For this reason, the free energy associated with $\phi_0(u)$ is relatively large and the corresponding threshold intensity I_0 is expected to be high, as compared with the optical Freédericksz transition in a homeotropic sample having the same thickness. Also, $\tilde{I}_0$ is larger for larger $\tilde{L}$, as seen in Fig. 2. The smallest $\tilde{I}_0$ occurs at $\tilde{L}\cong 2$, where the molecular distortion ϕ_0 has only half an oscillation between $z=0$ and L (Fig. 3). For $\tilde{L}<2$, $\tilde{I}_0$ is proportional to $1/L^2$ for sufficiently small $\tilde{L}$. This latter behavior is similar to that

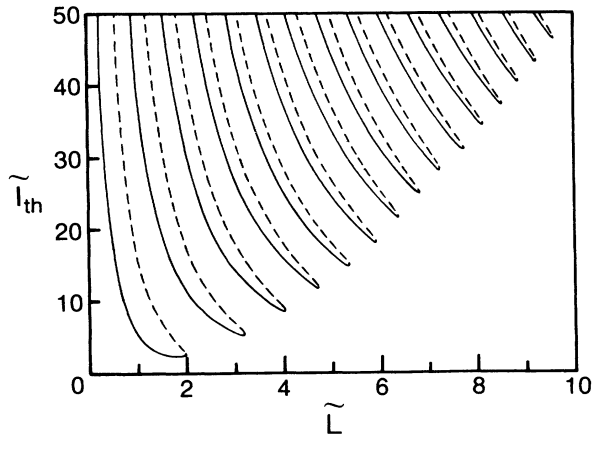

FIG. 2. Threshold intensity $\tilde{I}_{\rm th}$ vs sample thickness $\tilde{L}$. The intersecting points of an $\tilde{L}=$constant line with the solid curves yield the threshold intensities where the solution switches from stable to unstable, for that given $\tilde{L}$. Dashed lines correspond to unstable solutions.

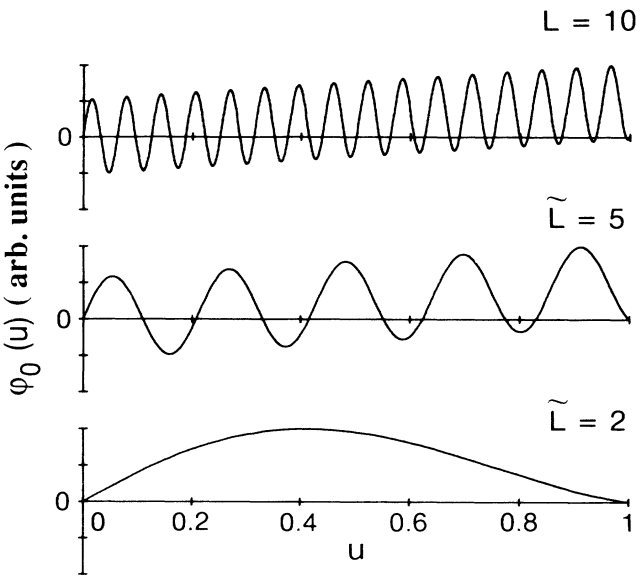

FIG. 3. Director reorientation $\phi_0(u)$ $(0\le u\le 1)$ as a function of position u in the sample at the minimum threshold intensity $\tilde{I}=\tilde{I}_0$ for three different sample thicknesses $\tilde{L}=2$, 5, and 10.

of the corresponding dc case and also to that of the optical Fréedericksz transition in a homeotropic sample, as one would expect from the distortion profile.

For a typical nematic sample ($n_0=1.5$; $n_e=1.7$; $k_{22}/k_{33}\simeq 0.6$) with a thickness of 100 μm and $\lambda=0.5$ μm, we find $\tilde{L}=250$ and $I_{\rm th}$ (homeotropic)$/I_0$ (planar)$\cong 1.5\times 10^{-4}$. Only for very thin samples with L being of the order of unity, the two thresholds are of comparable magnitude, but then they are both very high ($\simeq 1$ MW/cm^2). We note, however, that in the present case, if Δn is smaller, then for a given sample thickness, both $\tilde{L}$ and the threshold intensity I_0 are smaller. Considering the case with $\Delta n=10^{-3}$ and $k_{22}=2.5\times 10^{-7}$ dyne, we find, for $L=160$ μm (corresponding to $\tilde{L}=2$) and $\lambda=0.5$ μm, a threshold intensity $I_0=30$ kW/cm^2, which could be obtained in reality by a focused CW laser beam.

The above analysis gives no information about the character of the transition, i.e., whether it is first order or second order. To answer this question, we have solved numerically the set of nonlinear equations (1) and (2) in the stationary case ($\partial\phi/\partial t=0$ and $\partial\hat{\mathbf{s}}/\partial t=0$). In Fig. 4 the ellipticity S_3 of the output beam is plotted as a function of the input intensity $\tilde{I}$. The stable branches of stationary solutions are drawn in solid lines, while the unstable branches are dashed. An alternate series of stable and unstable bifurcations is found, according to Fig. 1. The curve in Fig. 4 clearly shows that the transition is second order.

When the intensity $\tilde{I}$ gradually increases from zero, the system remains undistorted with $\hat{\mathbf{s}}=(-1,0,0)$ and $\phi=0$ until the first branching point $\tilde{I}=\tilde{I}_0$ is reached. At this point, a second-order optical twist Fréedericksz transition occurs and the output ellipticity begins to deviate from zero, following the solid line in Fig. 4. To reach the second steady-state branch in Fig. 4, the intensity $\tilde{I}$ should be suddenly switched to the stable region and then

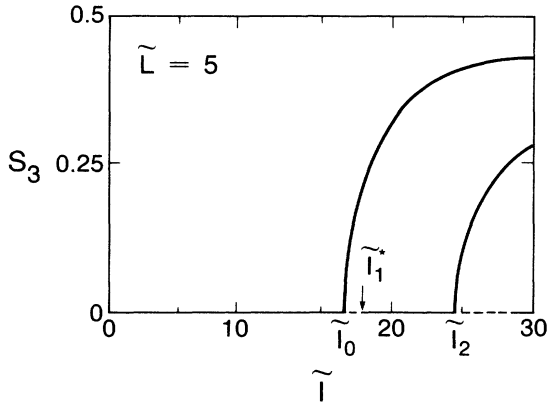

FIG. 4. Ellipticity s_3 of the transmitted light beam through the sample as a function of $\tilde{I}$ for $\tilde{L}=5$. The dashed part on the horizontal abscissa corresponds to the unstable solutions shown in Fig. 1.

gradually increased. At the second branching point $\tilde{I}=\tilde{I}_2$, the system again undergoes a second-order Fréedericksz transition and moves onto the second steady-state branch. If $\tilde{I}$ is slowly decreased instead, then as it reaches the end of the stable interval $\tilde{I}=\tilde{I}_1^*$, the system should switch to the first steady-state branch via a first-order transition. Note that in the stable regions (e.g., $\tilde{I}_1^* < \tilde{I} < \tilde{I}_2$), the system is intrinsically bistable, with the two stable solutions given by the unperturbed state $\hat{\mathbf{s}}=(-1,0,0)$, $\phi=0$ and the nonlinearly perturbed state on the steady-state branch. If $\tilde{I}$ is suddenly switched to an unstable region, the system would vary in time until the corresponding nonlinearly perturbed state on the steady-state branch is reached.

The present theory applies also to smectic-c liquid crystals in Rapini's N-configuration.[7] In this case, the following formal substitutions should be used:

$$\Delta n = n_0 \left[\frac{n_e}{(n_e^2\cos^2\theta + n_0^2\sin^2\theta)^{1/2}} - 1 \right] ,$$

$$\tilde{I} = \frac{(I/ck_{22}\Delta n)(\lambda/2\pi)^2}{(1-k\cos^2\theta)\sin^2\theta} ,$$

$$k = (k_{33}-k_{22})/k_{22} ,$$

where k_{33} is the bending elastic constant and θ is the tilt angle characteristic of the smectic material. Samples with small θ are expected to have lower threshold $\tilde{I}_{\rm th}$, which is roughly scaled by the factor $\sin^2\theta$.

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