Partially polarized light-induced Fréedericksz transition in nematic liquid crystals

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The combination of two noncoherent copropagating beams enables us to study the roles of the electromagnetic field symmetry and angular momentum in optically induced Fréedericksz transition. The particular choice of the interaction geometry allows one to achieve all-optical control of the spatial and dynamic behavior of orientational modes of this transition. The collective molecular precession rate is continuously controlled via the light angular momentum transfer. This control is coupled with the change of twist deformation of molecular orientation. Corresponding theoretical model is proposed and analytical solutions are obtained, providing insight into the multitype deformation behavior of the orientational transition and the possibility of its optical control. Excellent quantitative agreement between experimental and theoretical results is demonstrated. [S1063-651X(98)10110-1]

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I. INTRODUCTION

The dielectric torque (DT) $\mathbf{\Gamma}_E = \mathbf{P} \times \mathbf{E}$ (where $\mathbf{P}$ is the polarization of the medium and $\mathbf{E}$ is the electric field of the light) exerted by electromagnetic fields (EMF) on transparent anisotropic medium is one of the fascinating phenomena in physics [1]. Light-matter interactions via the DT have been studied in different material systems, ranging from individual molecules [2] to macroscopic solids (e.g., a half-wave plate [1]). It was established that the long scale orientational correlation of microscopic molecular axes in liquid-crystal materials may provide very efficient DT exerted by high (optical) frequency EMF [3,4]. These interactions, however, are relatively complicated for simulation because of strong intrinsic feedback and light-matter coupling. Namely, the DT depends upon the polarization state of light, which may be strongly modified due to the collective and spatially nonlocal reorientation of liquid-crystal molecules via the DT. Rich nature of the optical DT in nematic liquid crystal (NLC) has been demonstrated both experimentally and theoretically (see, e.g., [4–6]). The light-induced Fréedericksz transition (LIFT) and the transfer of angular momentum (AM) from the EMF to the director $\mathbf{n}$ (the average direction of microscopic molecular axes) of the NLC are important examples of that [6,7]. Coherent EMF has been applied for these experimental and theoretical studies, where the AM and azimuthal symmetry (AS) of the light were coupled. The rotating plane of polarization of the light has been used also to study the LIFT [8]. Recently, we have used two noncoherent copropagating cross polarized beams (Fig. 1) to study separately the roles of the AM and AS of the EMF in the LIFT [9]. Optical excitation and control of periodic molecular precession have been experimentally demonstrated [9]. However, corresponding theory has not been provided until now.

We report in the present work the detailed experimental and theoretical study of the LIFT in the field of two noncoherent copropagating oppositely polarized beams, as further development of the study started in Ref. [9]. In particular, a theoretical model is reported, which provides very important information concerning the molecular orientation behavior in both space and time. The obtained analytical solution is in excellent quantitative agreement with experimental data.

The organization of this paper is as follows. In the beginning of Sec. II we describe very shortly the interaction geometry, which allows one to implement the light driven molecular motor [9]. A general theoretical background is then provided. The concrete theoretical model is then established

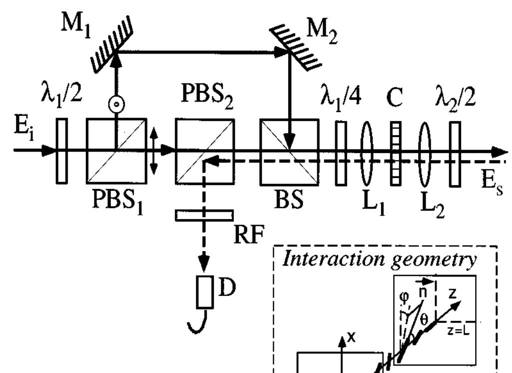

FIG. 1. Experimental setup and interaction geometry. E_i: initial linearly polarized beam of argon-ion laser (operating at 514 nm); $\lambda_1/2$: half-wave plate (for 514 nm), $PBS_{1,2}$: polarization beam splitters, $M_{1,2}$: mirrors, BS: 50/50% beam splitter, $\lambda_1/4$: quarter-wave plate (for 514 nm), C: homeotropic liquid crystal cell (thickness is 90 μm), L_1: lens (with focal length $f_1 = 13$ cm), E_s: linearly polarized weak probe He-Ne laser beam, L_2: lens (with focal length $f_2 = 10$ cm), $\lambda_2/2$: half-wave plate (for 632.8 nm), RF: red filter, D: detector. INSET: $\vec{n}$: director of NLC (initially parallel with $\vec{z}$), θ: polar angle, φ: azimuthal angle, $E_{x;y}$: electric field components of incident beams, K: wave vector of incident beams, L: thickness of the cell, squares are glass substrates (placed at $z=0$ and $z=L$). x, y, z: coordinate system.

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and analytically solved for two principal geometries, when the roles of the light AM and AS are separately analyzed in LIFT. A short description of principal experimental conditions and comparison of experimental and theoretical results is presented in the Sec. II. A summary of the work is then provided.

II. THEORY

A. Recall

Let us recall that the use of two copropagating noncoherent beams with orthogonal polarizations (Fig. 1) allowed us to optically induce, study, and control the director reorientation in a homeotropic (director is perpendicular to cell substrates) sample of NLC [9]. Two principal geometries of interaction have been studied, when two excitation beams have (a) opposite circular and (b) orthogonal linear polarizations. The variation of the intensity ratio of these two beams (for fixed total intensity) allowed the continuous modulation of the AM of combined EMF for a given AS [case (a)] and modulation of its AS for a given AM [case (b)]. Only a qualitative discussion of obtained experimental results has been provided, and, for example, a plane reorientation of the director was assumed in the case of the precession rate control by the input AM of the light. We will show below that the situation is more complicated, which is, however, possible to model correctly and to control experimentally.

B. General theoretical background

In the corresponding theoretical model, we shall consider an infinite layer of homeotropically aligned NLC cell of thickness L (inset to Fig. 1). Two noncoherent plane waves with frequency ω and wave number $k_0=\omega/c$, traveling in the positive direction of the z axis, are normally incident on the NLC at the plane $z=0$ (see below). The director $\mathbf{n}$ may be described by polar angles θ and φ, where θ is the tilt angle between n and the z axis, and φ is the azimuthal angle between the local (n,z) and (x,z) planes. We shall thus describe the perturbation of the director as

$$\mathbf{n}=\mathbf{e}_x\sin\,\theta\,\cos\,\varphi+\mathbf{e}_y\sin\,\theta\,\sin\,\varphi+\mathbf{e}_z\cos\,\theta, \tag{1}$$

where $\mathbf{e}_x$, $\mathbf{e}_y$, and $\mathbf{e}_z$ are unit vectors along the x, y, and z axes.

All functions depend upon the coordinate z only, since the interaction geometry is invariant to translation in the (x,y) plane. In addition, we assume the following dependence of the azimuthal angle φ on time ''t'' and space ''z'':

$$\varphi(z,t)=\Omega t+\alpha(z), \tag{2}$$

where Ω is the frequency of rotation of the complex director configuration around the z axis and $\alpha(z)$ represents the twist of the director deformation.

In order to find the photoinduced configuration of the director, it is necessary to take into account the perturbations θ and φ for solution of Maxwell's equations in the NLC. We shall consider the weak and slow perturbation (see below) regime of the director, which will allow us to seek these solutions in the form $\mathbf{E}=\mathbf{E}_a e^{ik_0\psi(z)-i\omega t}$ in the geometrical optics approximation [4]:

$$[(\partial\psi/\partial z)^2(\delta_{zi}\delta_{zj}-\delta_{ij})+\varepsilon_{ij}]E_{aj}=0, \tag{3a}$$

$$P_z=(c/8\pi)|\partial\psi/\partial z|\{|E_x|^2+|E_y|^2\}=\text{const}, \tag{3b}$$

where E_a and ψ are the slowly varying amplitude and phase, respectively. P_z is the value of the z component of the Poynting vector in the medium, $\varepsilon_{ij}=\varepsilon_\perp\delta_{ij}+\varepsilon_a n_i n_j$ is the local dielectric susceptibility tensor at light frequency (with $\varepsilon_a=\varepsilon_\parallel-\varepsilon_\perp$), and E_{aj} are x,y,z components of the field E_a. The P_z is constant, since the ε_{ij} depends upon z only [4]. We will keep in mind the harmonic character of our waves, but in our further discussion we will omit the term $e^{-i\omega t}$ for the sake of shortness.

In general there are two solutions of Eqs. (3): the ordinary wave $\mathbf{E}_{\text{ord}}$ with the phase $k_0\psi_0$ and amplitude E_0 (expressed by the corresponding constant Pointing vector P_0):

$$\mathbf{E}_{\text{ord}}=E_0(-\sin\,\varphi,\cos\,\varphi,0)e^{ik_0\psi_o(z)},$$
$$d\psi_o/dz=\pm\sqrt{\varepsilon_\perp}, \tag{4a}$$
$$|E_0|^2=8\pi P_0/(c\sqrt{\varepsilon_\perp}),$$

and the extraordinary wave $\mathbf{E}_{\text{ext}}$ with the phase $k_0\psi_e$ and amplitude E_e (expressed by the corresponding constant Poynting vector P_e):

$$\mathbf{E}_{\text{ext}}=E_e(\varepsilon_{zz}/\varepsilon_\parallel)^{1/4}(\cos\,\varphi,\sin\,\varphi,$$
$$-\varepsilon_a\sin 2\theta/(2\varepsilon_{zz}))e^{ik_0\psi_e(z)},$$
$$d\psi_e/dz=\pm\sqrt{\varepsilon_\perp\varepsilon_\parallel/\varepsilon_{zz}}, \tag{4b}$$
$$|E_e|^2=8\pi P_e/(c\sqrt{\varepsilon_\perp}).$$

One can see that the incident light is split into ''e'' and ''o'' waves, in such a manner that the electric field of the ''e'' wave is in the (n,z) plane, while the electric field of the ''o'' wave is perpendicular to that plane. Note that the initial energy splitting follows the local (n,z) plane, as in the linear Mauguin regime [3]. However, several important differences are present in our case, e.g., the director configuration is influenced by the light field, the phase velocity, and the amplitude of the extraordinary wave are changed along the propagation, etc. Consequently, in the case of multiple incident waves, each wave will be split into the mentioned ''o'' and ''e'' waves, and the total fields will be presented as

$$\mathbf{E}_{\text{ord}}=S_0(-\sin\,\varphi,\cos\,\varphi,0),\quad S_0=\sum_m E_{0m}e^{ik_0\psi_{om}(z)}, \tag{5a}$$

$$\mathbf{E}_{\text{ext}}=S_e[\cos\,\varphi,\sin\,\varphi,-\varepsilon_a\,\sin 2\theta/(2\varepsilon_{zz})],$$

$$S_e=(\varepsilon_{zz}/\varepsilon_\parallel)^{1/4}\sum_m E_{em}e^{ik_0\psi_{em}(z)}. \tag{5b}$$

The behavior of φ and θ is then found by seeking the balance condition among elastic, electromagnetic, and viscous torques acting on the director [3–5]:

$$\mathbf{T}_{\text{elast}}+\mathbf{T}_{\text{em}}+\mathbf{T}_{\text{visc}}=0, \tag{6}$$

where T_{elast} and T_{em} are found using the derivatives of corresponding free energy densities F [4,5,7]:

$$F_{\text{elast}}=0.5K_1(\text{div }\mathbf{n})^2+0.5K_2(\mathbf{n}\cdot\text{curl }\mathbf{n})^2+0.5K_3[\mathbf{n}\times\text{curl }\mathbf{n}]^2, \tag{7a}$$

$$F_{\text{em}}=-(1/16\pi)\varepsilon_{ij}E_iE_j. \tag{7b}$$

The viscous torque is defined by the dissipation function:

$$R=(\gamma/2)(\partial\mathbf{n}/\partial t)^2, \tag{7c}$$

where K_i are Frank's elastic constants, γ is the orientational viscosity of the NLC, $\mathbf{E}=\mathbf{E}_{\text{ord}}+\mathbf{E}_{\text{ext}}$. The obtained φ and θ components of torques are

$$\begin{aligned}T_{\text{elast},\theta}&=\frac{\partial}{\partial z}\frac{\partial F_{\text{elast}}}{\partial(\partial\theta/\partial z)}-\frac{\partial F_{\text{elast}}}{\partial\theta}\\&=(K_3\cos^2\theta+K_1\sin^2\theta)\frac{\partial^2\theta}{\partial z^2}\\&\quad-0.5(K_3-K_1)\left(\frac{\partial\theta}{\partial z}\right)^2\sin 2\theta\\&\quad-\left(\frac{\partial\varphi}{\partial z}\right)^2(0.25K_3\sin 4\theta+K_2\sin^2\theta\sin 2\theta),\end{aligned}$$

$$T_{\text{em},\theta}=\frac{\partial}{\partial z}\frac{\partial F_{\text{em}}}{\partial(\partial\theta/\partial z)}-\frac{\partial F_{\text{em}}}{\partial\theta}=\frac{\varepsilon_a\varepsilon_\perp}{16\pi\varepsilon_\parallel}(\varepsilon_\parallel/\varepsilon_{zz})^2\sin 2\theta|S_e|^2, \tag{8a}$$

$$T_{\text{visc},\theta}=-\frac{\partial R}{\partial(\partial\theta/\partial t)}=0,$$

and

$$\begin{aligned}T_{\text{elast},\varphi}&=\frac{\partial}{\partial z}\frac{\partial F_{\text{elast}}}{\partial(\partial\varphi/\partial z)}-\frac{\partial F_{\text{elast}}}{\partial\varphi}\\&=\sin^2\theta(K_3\cos^2\theta+K_2\sin^2\theta)\frac{\partial^2\varphi}{\partial z^2}+\sin 2\theta(K_3\cos 2\theta\\&\quad+2K_2\sin^2\theta)\left(\frac{\partial\theta}{\partial z}\right)\left(\frac{\partial\varphi}{\partial z}\right),\end{aligned}$$

$$\begin{aligned}T_{\text{em},\varphi}&=\frac{\partial}{\partial z}\frac{\partial F_{\text{em}}}{\partial(\partial\varphi/\partial z)}-\frac{\partial F_{\text{em}}}{\partial\varphi}\\&=\frac{\varepsilon_a\varepsilon_\perp}{16\pi\varepsilon_\parallel}(\varepsilon_\parallel/\varepsilon_{zz})\sin^2\theta(S_eS_0^*+\text{c.c.}),\end{aligned} \tag{8b}$$

$$T_{\text{visc},\varphi}=-\frac{\partial R}{\partial(\partial\varphi/\partial t)}=-\gamma\Omega\sin^2\theta.$$

Note [from Eq. (8a)] that the polar deformation θ is defined by the extraordinary component of the total field $|S_e|^2$, while the twist deformation (the z dependence of φ) is defined [from Eq. (8b)] by the crossed term ($S_eS_o^*+\text{c.c.}$). The φ component of Eq. (6), $T_{\text{elast},\varphi}+T_{\text{em},\varphi}+T_{\text{visc},\varphi}=0$, is solved for following boundary conditions [5]:

$$\begin{aligned}&\theta(z=0)=\theta(z=L)=0,\\&d\alpha/dz(z=0)=d\alpha/dz(z=L)=0,\end{aligned} \tag{9}$$

to give the z dependence of the director:

$$\frac{d\alpha}{dz}=\frac{\int_0^z\sin^2\theta(z')[\gamma\Omega-(\varepsilon_a\varepsilon_\perp/16\pi\varepsilon_\parallel)(\varepsilon_\parallel/\varepsilon_{zz})(S_eS_0^*+\text{c.c.})]dz'}{K_3\sin^2\theta(z)[1-\sin^2\theta(z)(K_3-K_2)/K_3]}. \tag{10}$$

The conditions (9), used in Eq. (10), lead to the following condition for the definition of Ω [to keep the value of $d\alpha/dz(z=L)$ bounded]:

$$\Omega=\frac{\varepsilon_a\varepsilon_\perp}{16\pi\varepsilon_\parallel\gamma}\frac{\int_0^L\sin^2\theta(z')(\varepsilon_\parallel/\varepsilon_{zz})(S_eS_0^*+\text{c.c.})dz'}{\int_0^L\sin^2\theta(z')dz'}. \tag{11}$$

Equations (10) and (11) are general solutions of material equations.

We shall further consider two concrete configurations of incident electromagnetic fields (used in our experiment for the NLC perturbation) to find the corresponding director perturbations, defined by the set of Ω,α,θ.

C. Circular polarizations

Let us consider the case when two circularly polarized noncoherent waves ($\mathbf{E}_1$ and $\mathbf{E}_2$) with opposite circularity are incident on the NLC:

$$\begin{aligned}\mathbf{E}_1&=E_1\frac{i\mathbf{l}_\perp(0)+\mathbf{l}_\parallel(0)}{\sqrt{2}}e^{ik_0z},\\\mathbf{E}_2&=E_2\frac{-i\mathbf{l}_\perp(0)+\mathbf{l}_\parallel(0)}{\sqrt{2}}e^{ik_0z+i\delta(t)},\end{aligned} \tag{12}$$

where $\mathbf{l}_\perp(z)=[-\sin\varphi(z),\cos\varphi(z)]$, $\mathbf{l}_\parallel(z)=[\cos\varphi(z),\sin\varphi(z)]$. The $\mathbf{l}_\parallel$ and $\mathbf{l}_\perp$ are unit vectors in the (x,y) plane. The $\mathbf{l}_\parallel$ is parallel to the transverse vector component $\mathbf{n}_\perp$ of the director $\mathbf{n}$ (where $\mathbf{n}_\perp=\mathbf{n}-\mathbf{n}_z$, $\mathbf{n}_z$ is the "$\mathbf{z}$" vector component of $\mathbf{n}$). The $\mathbf{l}_\perp$ is perpendicular to $\mathbf{l}_\parallel$. The $E_{1,2}$ are wave amplitudes and $\delta(t)$ is the chaotic phase shift between two waves (thus we have $\langle E_1 E_2 e^{i\delta(t)}\rangle = 0$, where the $\langle\ \rangle$ assumes time averaging).

Using solutions (4), we can represent the fields (12) in the medium. Thus we have for the two polarization components of E_1:

$$\mathbf{E}_{1,o} = iA_1(-\sin\varphi,\cos\varphi,0)\exp(ik_0\sqrt{\varepsilon_\perp}z),$$

$$\mathbf{E}_{1,e} = (\varepsilon_{zz}/\varepsilon_\parallel)^{1/4}A_1[\cos\varphi,\sin\varphi,-\varepsilon_a\sin 2\theta/(2\varepsilon_{zz})] \times\exp\left(ik_0\int_0^z\sqrt{\varepsilon_\perp\varepsilon_\parallel/\varepsilon_{zz}}dz'\right), \tag{13a}$$

and for the two polarization components of E_2:

$$\mathbf{E}_{2,o} = -iA_2(-\sin\varphi,\cos\varphi,0)\exp[ik_0\sqrt{\varepsilon_\perp}z + i\delta(t)],$$

$$\mathbf{E}_{2,e} = (\varepsilon_{zz}/\varepsilon_\parallel)^{1/4}A_2[\cos\varphi,\sin\varphi,-\varepsilon_a\sin 2\theta/(2\varepsilon_{zz})] \times\exp\left(ik_0\int_0^z\sqrt{\varepsilon_\perp\varepsilon_\parallel/\varepsilon_{zz}}dz' + i\delta(t)\right). \tag{13b}$$

The obtained total ordinary and extraordinary fields will be expressed by Eq. (5), with

$$S_e = (\varepsilon_{zz}/\varepsilon_\parallel)^{1/4}(A_1 + A_2 e^{i\delta(t)})\exp\left(ik_0\int_0^z\sqrt{\varepsilon_\perp\varepsilon_\parallel/\varepsilon_{zz}}dz'\right),$$

$$S_o = i(A_1 - A_2 e^{i\delta(t)})\exp(ik_0\sqrt{\varepsilon_\perp}z), \tag{14}$$

where

$$A_{1,2} = \tau E_{1,2}/\sqrt{2},\quad \tau = 2/(1+\sqrt{\varepsilon_\perp}),$$

$$P_{i,o} = P_{i,e} = c\sqrt{\varepsilon_\perp}|A_i|^2/(8\pi) = P_i/2,\quad i = 1,2,$$

and, e.g., $P_{i,o}$ represents the ''o'' component of the Pointing vector of the ''i'' incident field in the medium, etc.

We will make some further assumptions to simplify the solutions (10) and (11). Thus, we will seek the polar perturbation in the following form [4,5]:

$$\theta(z) = \theta_0 \sin qz,\quad q = \pi/L, \tag{15}$$

and we will assume a weak perturbation regime:

$$\Delta = k_0\sqrt{\varepsilon_\perp}\int_0^L\sqrt{\varepsilon_\parallel/\varepsilon_{zz}}dz' - k_0\sqrt{\varepsilon_\perp}z = k_0\sqrt{\varepsilon_\perp}L\theta_0^2\varepsilon_a/(4\varepsilon_\parallel) \ll 1, \tag{16}$$

which supposes a small nonlinear phase shift between extraordinary and ordinary waves. Final forms obtained from Eqs. (10) and (11) for the φ perturbation are

$$\Omega = \frac{\Delta}{2}\frac{1-R}{1+R}\frac{K_3 q^2}{\gamma},$$

$$\alpha(z) = \frac{\Delta}{4}\frac{1-R}{1+R}f\left(\frac{z}{L}\right), \tag{17}$$

where $f(\xi) = 1 - (1/2)[\xi + \sin(2\pi\xi)/(2\pi)] - \pi\xi(1-\xi)\cot(\pi\xi)$, $R \equiv |A_2|^2/|A_1|^2$.

Note that the parameter R represents the total AM of the light incident on the NLC. Namely, we have a maximal AM for $R=0$ (when A_1 is maximal) and a minimal AM for $R=1$.

The polar perturbation amplitude θ_0 is found under the same assumptions [Eqs. (15) and (16)], using the θ component of Eq. (6): $T_{\mathrm{elast},\theta} + T_{\mathrm{em},\theta} + T_{\mathrm{visc},\theta} = 0$. To solve this equation, we will consider terms up to θ^3 only and we will use the Galerkin method, which consists in multiplying the equation by $2\sin(qz)$ and integrating through the z [4,5]. We obtain finally

$$-K_3 q^2\left(\theta_0 - \frac{K_3 - K_1}{2K_3}\theta_0^3\right) + \frac{\varepsilon_a\varepsilon_\perp}{8\pi\varepsilon_\parallel}(|A_1|^2 + |A_2|^2)\left(\theta_0 - \frac{4\varepsilon_\perp - 5\varepsilon_a}{8\varepsilon_\parallel}\theta_0^3\right) = 0, \tag{18}$$

where the first and second terms represent the space-averaged values of the elastic and electromagnetic torques, respectively. The nontrivial solution of this equation is

$$\theta_0^2 = \frac{(|A_1|^2 + |A_2|^2)/I_{\mathrm{lin}} - 1}{(4\varepsilon_\perp - 5\varepsilon_a)/(8\varepsilon_\parallel) - (K_3 - K_1)/(2K_3)}, \tag{19}$$

where $I_{\mathrm{lin}} = 8\pi\varepsilon_\parallel K_3 q^2/(\varepsilon_a\varepsilon_\perp)$ (see below). The obtained solutions (17) and (19) demonstrate several important characteristics of the LIFT in the case of two noncoherent circularly polarized copropagating waves.

First, the polar perturbation is achieved only when the electromagnetic torque overcomes the elastic torque: $|A_1|^2 + |A_2|^2 \geqslant I_{\mathrm{lin}}$. That means, by the way, that the value of the total intensity corresponding to the LIFT threshold will be $I_{\mathrm{tot}} = 2(|A_1|^2 + |A_2|^2) \geqslant 2I_{\mathrm{lin}} = I_{\mathrm{th}}$. Thus, this threshold intensity corresponds to the total Poynting vector value:

$$P_{\mathrm{tot}} = \sum_{i=1}^{2}(P_{i,o} + P_{i,e}) = I_{\mathrm{tot}}c\sqrt{\varepsilon_\perp}/(8\pi) = 2P_{\mathrm{lin}}, \tag{20}$$

where P_{lin} represents the Pointing vector value for the LIFT threshold in the case of single linearly polarized excitation wave (I_{lin} is the corresponding intensity). This threshold is independent of the ratio R. Thus, the obtained threshold value is always equal to the LIFT threshold corresponding to the case of single circularly polarized ($R=0$) or nonpolarized wave [4].

Second, the spatial and temporal behavior of the director $\mathbf{n}$ depends on two optical parameters and, thus, may be optically controlled. Indeed, the polar reorientation angle θ depends upon the total intensity I_{tot} only. At the same time, the twist component of the director deformation and the precession rate may be controlled also (in addition to I_{tot}, since Δ depends upon θ) by the intensity ratio R [see Eq. (17)]. For instance, a given polar excitation (θ) would become stronger for an increase of total intensity δI_{tot}, while the behavior of the α and Ω would depend on how the δI_{tot} would change the R. Both of these parameters (α and Ω) are larger for $R \to 0$ and suppressed for $R \to 1$. Thus the director would be reoriented in a plane ($\alpha=0$) and has stationary ($\Omega=0$) configuration for $R=1$, while we would obtain maximal Ω and α when $R=0$. It is important to note that the continuous optical control of Ω via R is coupled with the corresponding twist deformation (α). Another interesting possibility is the control of the director rotation direction (the sign of the Ω) by a proper choice of the intensity ratio of used beams.

D. Linear polarizations

Now let us consider the case when two linearly polarized noncoherent waves with orthogonal polarizations are incident on the NLC:

$$\mathbf{E}_1=E_1\mathbf{e}_y e^{ik_0z},\qquad \mathbf{E}_2=E_2\mathbf{e}_x e^{ik_0z+i\delta(t)}. \tag{21}$$

Recall that only the AS of the input EMF may be modulated in this case, while its AM is always vanishing. As before, the perturbation of the director $\mathbf{n}$ (the θ and φ) will be expressed by Eq. (1) and Eq. (2), and we will assume that all functions depend only on z.

Using solutions (4), we can represent the fields (21) in the medium for the polarization components of E_1 as

$$\mathbf{E}_{1,o}=A_1\cos\varphi_0(-\sin\varphi,\cos\varphi,0)\exp(ik_0\sqrt{\varepsilon_\perp}z),$$
$$\mathbf{E}_{1,e}=(\varepsilon_{zz}/\varepsilon_\parallel)^{1/4}A_1\sin\varphi_0[\cos\varphi,\sin\varphi,-\varepsilon_a\sin 2\theta/(2\varepsilon_{zz})]\times\exp\left(ik_0\int_0^z\sqrt{\varepsilon_\perp\varepsilon_\parallel/\varepsilon_{zz}}dz'\right), \tag{22}$$

and for the E_2 as

$$\mathbf{E}_{2,o}=A_2(-\sin\varphi_0)(-\sin\varphi,\cos\varphi,0)\exp[ik_0\sqrt{\varepsilon_\perp}z+i\delta(t)],$$
$$\mathbf{E}_{2,e}=(\varepsilon_{zz}/\varepsilon_\parallel)^{1/4}A_2\cos\varphi_0\times[\cos\varphi,\sin\varphi,-\varepsilon_a\sin 2\theta/(2\varepsilon_{zz})]\times\exp\left(ik_0\int_0^z\sqrt{\varepsilon_\perp\varepsilon_\parallel/\varepsilon_{zz}}dz'+i\delta(t)\right).$$

The total ordinary and extraordinary fields will be expressed by Eq. (5), where

$$S_e=(\varepsilon_{zz}/\varepsilon_\parallel)^{1/4}(A_1\sin\varphi_0+A_2\cos\varphi_0e^{i\delta(t)})\times\exp\left(ik_0\int_0^z\sqrt{\varepsilon_\perp\varepsilon_\parallel/\varepsilon_{zz}}dz'\right),$$
$$S_o=(A_1\cos\varphi_0-A_2\sin\varphi_0e^{i\delta(t)})\exp(ik_0\sqrt{\varepsilon_\perp}z), \tag{23}$$

and

$$A_{1,2}=\tau E_{1,2},\quad \varphi_0=\varphi(z=0,t),$$
$$P_{1,o}=c\sqrt{\varepsilon_\perp}|A_1|^2\cos^2\varphi_0/(8\pi)=\text{const},$$
$$P_{1,e}=c\sqrt{\varepsilon_\perp}|A_1|^2\sin^2\varphi_0/(8\pi)=\text{const},$$
$$P_{2,o}=c\sqrt{\varepsilon_\perp}|A_2|^2\sin^2\varphi_0/(8\pi)=\text{const},$$
$$P_{2,e}=c\sqrt{\varepsilon_\perp}|A_2|^2\cos^2\varphi_0/(8\pi)=\text{const}.$$

Note that we have constant Poynting vectors for ordinary and extraordinary waves, which suggests that the φ_0 is also constant. Thus we obtain from Eq. (2) that the dynamic precession of the director is absent:

$$\varphi_0=\text{const}\Rightarrow\Omega=0. \tag{24}$$

Here also, we will assume the same conditions, expressed by Eq. (15) and (16), to simplify the solutions (10) and (11). We obtain thus

$$\Omega=\frac{\varepsilon_a\varepsilon_\perp}{16\pi\varepsilon_\parallel\gamma}\sin 2\varphi_0(|A_1|^2-|A_2|^2),\qquad d\alpha/dz=0. \tag{25}$$

Taking into account Eq. (24) we find from Eq. (25) that in the general case $\sin 2\varphi_0=0$ and $\varphi(z,t)=\text{const}$, which finally gives

$$\varphi=\pi m/2,\quad m=0,\pm1,\pm2,\ldots\ . \tag{26}$$

We see that the vector component $\mathbf{n}_\perp$ of the director in the (x,y) plane is parallel to the polarization of one of the incident beams (along $\mathbf{e}_y$ for $m=\pm1,\pm3,\ldots$ and along $\mathbf{e}_x$ for $m=0,\pm2,\ldots$). Indeed, we have obtained this degeneracy of orientation using only the condition of the vanishing torque:

$$T_{\text{em},\varphi}+T_{\text{elast},\varphi}=-\frac{\partial(F_{\text{em}}+F_{\text{elast}})}{\partial\varphi}=\frac{\varepsilon_a\varepsilon_\perp}{16\pi\varepsilon_\parallel}\sin^2\theta\sin 2\varphi(|A_1|^2-|A_2|^2)=0.$$

The following condition of the minimal free energy density will break this degeneracy and will define the final orientation of φ:

$$\frac{\partial^2(F_{\text{em}}+F_{\text{elast}})}{\partial\varphi^2}=-(\varepsilon_a\varepsilon_\perp/8\pi\varepsilon_\parallel)\sin^2\theta\times\cos 2\varphi(|A_1|^2-|A_2|^2)>0. \tag{27}$$

Using Eqs. (26) and (27) we obtain the condition $(-1)^m(|A_2|^2-|A_1|^2)>0$, which leads to

$$\varphi=\begin{cases}\pi/2+\pi m & \text{for } |A_1|^2>|A_2|^2,\\ \pi m & \text{for } |A_1|^2<|A_2|^2.\end{cases} \tag{28}$$

We see, therefore, that the linearly polarized wave with higher intensity will always determine the direction of $\mathbf{n}_\perp$.

We will find the final polar perturbation amplitude θ_0 following the same way as for the circular wave excitation [described after the Eq. (17)]:

$$\theta_0^2=\frac{(|A_1|^2\sin^2\varphi+|A_2|^2\cos^2\varphi)/I_{\text{lin}}-1}{(4\varepsilon_\perp-5\varepsilon_a)/(8\varepsilon_\parallel)-(K_3-K_1)/(2K_3)}. \tag{29}$$

Note that the only difference in this case would be the contribution of each incident wave in the electromagnetic torque, which is expressed here by corresponding weights

"$\sin^2\varphi$" and "$\cos^2\varphi$." Obviously, we have to take into account Eq. (28) to define the wave with principal contribution in Eq. (29). Thus, the stronger wave becomes the "e" wave and the value of the LIFT threshold is defined by its intensity, since the threshold condition becomes $\max(|A_1|^2,|A_2|^2) \geqslant I_{\mathrm{lin}}$. The corresponding total Poynting vector value will be

$$P_{\mathrm{tot}}=P_1+P_2=(|A_1|^2+|A_2|^2)c\sqrt{\varepsilon_\perp}/(8\pi)$$
$$=I_{\mathrm{lin}}c\sqrt{\varepsilon_\perp}/(8\pi)(1+R)=P_{\mathrm{lin}}(1+R), \qquad (30)$$

where $R=\min(|A_1|^2,|A_2|^2)/\max(|A_1|^2,|A_2|^2)$. These results are in agreement with those reported in Ref. [4]. Namely, the threshold and the initial growth of the director perturbation in the case of the LIFT induction by an arbitrarily polarized single beam is described there as $\mathbf{n}=(n_{0,x},n_{0,y})e^{\Gamma t}\sin qz$, where

$$\Gamma=K_3q^2/\gamma\left(\frac{|A_1|^2+|A_2|^2}{I_{\mathrm{lin}}}\,\frac{1+\sqrt{\xi_1^2+\xi_3^2}}{2}-1\right),$$
$$\frac{n_{0,y}}{n_{0,x}}=\frac{\xi_1}{\xi_3+\sqrt{\xi_1^2+\xi_3^2}}, \qquad (31)$$
$$(|A_1|^2+|A_2|^2)_{\mathrm{th}}=I_{\mathrm{lin}}\,\frac{2}{1+\sqrt{\xi_1^2+\xi_3^2}},$$

and ξ_j are Stokes parameters:

$$\xi_3=(|A_2|^2-|A_1|^2)/(|A_1|^2+|A_2|^2),$$
$$\xi_1=2|A_1||A_2|\langle\cos\,\delta(t)\rangle/(|A_1|^2+|A_2|^2).$$

In our case $\xi_1=0$ (for noncoherent cross-polarized copropagating beams) we should obtain

$$(n_{0,y}/n_{0,x})\xrightarrow[\xi_1\to 0]{}\begin{cases}\xi_1/(2\xi_3)=0 & (n_{0,y}=0), \quad \text{when } \xi_3>0,\\ 2|\xi_3|/\xi_1=\infty & (n_{0,x}=0), \quad \text{when } \xi_3<0,\end{cases} \qquad (32)$$

and

$$(|A_1|^2+|A_2|^2)_{\mathrm{th}}\xrightarrow[\xi_1\to 0]{} I_{\mathrm{lin}}(1+R).$$

The system of Eq. (32) shows the same results as our derivation [Eqs. (28) and (30)].

Note that the parameter R represents here the AS of the incident light, in contrast to the previous case. Obviously, the case of two *coherent* copropagating beams would be different, depending, for instance, upon the phase shift between these beams. This case was intensively studied by number of research groups [4–6].

The solutions obtained above [Eqs. (28)–(30)] demonstrate important differences of the LIFT with respect to the previous geometry. First, for any ratio R of intensities of impinging waves, they suggest planar ($\alpha=$const) and stationary ($\Omega=0$) director reorientation [compare Eq. (17) and Eq. (25)]. Second, the total input intensity corresponding to the LIFT threshold will depend linearly upon the intensity ratio R [see Eq. (30)], since the transition threshold is overcome by one (stronger) beam only, while the second beam does not contribute in this process. This is the reason of the dependence upon R. Indeed, its value changes from $P_{\mathrm{th}}=P_{\mathrm{lin}}$ (when we direct all the input power into single linearly polarized wave, i.e., $R=0$) to $P_{\mathrm{th}}=2P_{\mathrm{lin}}$ (when the input power is initially distributed between two noncoherent cross-polarized impinging waves of equal intensity, i.e., $R=1$). Similar doubling of the LIFT threshold was predicted in Ref. [4] for a single excitation beam when its polarization is changed from linear to a nonpolarized or circular. However, as we have mentioned above, the latter case may be accompanied by complex AM transfer phenomenon, which is absent in our case.

III. COMPARISON WITH EXPERIMENTAL RESULTS

A. Experimental setup and interaction geometry

We describe first the experimental setup [9] and results (Fig. 1) to compare with our theoretical model. The linearly polarized Gaussian beam E_i of an argon ion laser (operating at 514.5 nm) is divided in two separate arms after traversing the $\lambda_1/2$ half-wave plate (for 514.5 nm) and the polarizational beam splitter PBS_1. The deviated beam is reflected from two mirrors M_1 and M_2, and then returned to the initial optical path and combined with the directly transmitted beam by the simple beam splitter BS. The optical path difference of two beams exceeds the coherence length of the laser used (as checked by interferometry). The further control of the polarizations of the two beams is performed by the $\lambda_1/4$ quarter-wave plate (for 514.5 nm). In order to obtain an optical field composed of two beams with orthogonal linear or circular polarizations, the optical axis of the $\lambda_1/4$ plate is oriented parallel or at 45° with respect to the plane of initial polarizations of two beams, correspondingly. The intensity ratio R of these two beams is easily controlled by rotating the $\lambda_1/2$ plate. The combined excitation beam is focused by the lens L_1 and is normally incident on the NLC sample C. In our experiment, the sample studied was a 90-μm-thick homeotropic NLC film. The NLC used is E7 from Merck Ltd. The combined beam spot diameter at the sample is 80 μm. The weak linearly polarized probe beam E_s of the He-Ne laser (operating at 632.8 nm) is counterpropagating with respect to the excitation beams, and is focused in the director perturbation area by the lens L_2. The transmitted probe beam is then collimated by the lens L_1 and is reflected from the second polarization beam splitter PBS_2 out of the principal axis. The polarization plane of the probe beam is rotated by means of the second half-wave plate $\lambda_2/2$ (for the 632.8

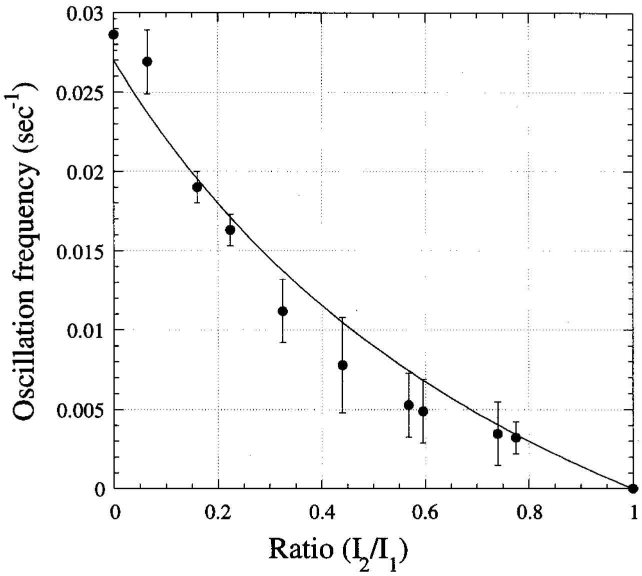

FIG. 2. Molecular precession rate Ω vs the intensity ratio $R = I_1/I_2$ of two circularly polarized noncoherent copropagating beams for a total input intensity of $I = 1.65\ \mathrm{kW/cm^2}$. The line is a theoretical fit by formula (17).

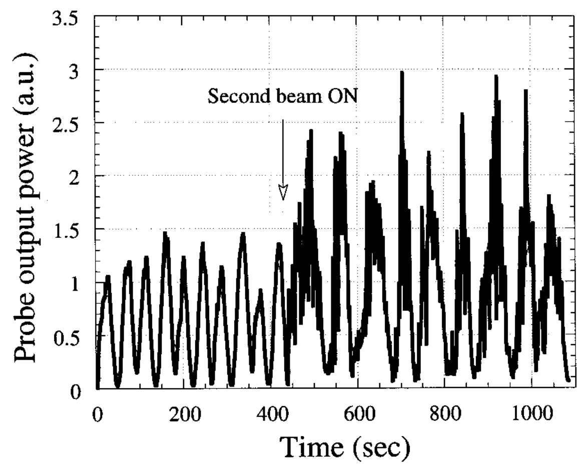

FIG. 3. Demonstration of control of molecular precession rate (Ω) and polar reorientation (θ) by simultaneous modulation of the total intensity and angular momentum of input EMF. The initial perturbation is induced by single circularly polarized beam and a weak beam of opposed circularity is then added.

nm). The polarization state of the output probe beam is analyzed by means of the detector D. The noise or reflections of the excitation beams are cut off by the red filter RF. The period of the output probe beam intensity modulation (resulted from its polarization dynamical change) is detected for different fixed excitation conditions and orientations of $\lambda_2/2$ plate.

Note that I_{tot} and R are varied independently in this experimental setup. That is, the rotation of the half-wave plate $\lambda_1/2$ leads to variation of the R at constant I_{tot}, and fixing its orientation allows separate variation of the I_{tot}.

B. Circular polarization case

First, we use two beams with opposite circular polarizations, normally incident on the sample (optical axis of the $\lambda_1/4$ plate is oriented at 45° with respect to the initial linear polarization plane).

Optically induced Fréedericksz transition (or LIFT) is observed above a certain threshold value of the total intensity I_{th}. The value of I_{th} in this case appears to be independent from the value of AM (defined by R) carried by the combined beam [9]. Its value (the I_{th}) is twice as high as the one corresponding to the single linearly polarized beam. These results are in agreement with the predictions of our theory [see Eq. (20)].

Once the threshold is achieved, the behavior of the NLC system is essentially affected by the value of the input AM (or R). Namely, when two circularly polarized beams are of equal intensities ($R=1$) the reorientation is quite stable, thus confirming our theoretical predictions ($\theta \neq 0$, $\Omega = 0$). Periodic rotation of the NLC director around the z axis occurs for all other cases (i.e., when the $R \neq 1$, and hence the incident light carries a nonvanishing AM). Strong dependence of the precession period on R is observed (at fixed I_{tot}). Inverted oscillation period versus R is plotted in Fig. 2 for total input intensity $I = 1.65\ \mathrm{kW/cm^2}$. The inverted period tends to zero while approaching $R=1$. The corresponding theoretical curve is drawn in the same figure using Eq. (17) (for the Ω) of our theory. We have used the following material parameters for this curve: $\varepsilon_{\parallel} = 2.92$, $\varepsilon_{\perp} = 2.28$, $\gamma = 0.5$ P, $(K_3 - K_1)/K_3 = 0.379$, $K_3 = 7.5 \times 10^{-7}$ dyne (see, e.g., in Ref. [3]). Excellent quantitative agreement is achieved.

An example of the action of simultaneous variation of I_{tot} and R on the director reorientation and precession is demonstrated in Fig. 3. This corresponds to the situation when the reorientation and precession are initiated by a single beam ($R=0$), and a small quantity (few percent) of oppositely polarized copropagating photons is added to the first beam. One can see that the modulation depth (defined by the θ) is enhanced, while the precession rate Ω is decreased, as predicted by our theory [see Eq. (17)].

We have no direct experimental tools to check the prediction of our theory concerning the twist component of the director reorientation [see Eq. (17)]. However, these predictions may be easily interpreted qualitatively. Namely, let us suppose that we have a small and initially ''plane'' director

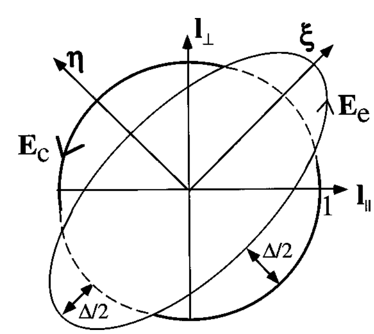

FIG. 4. Schematic demonstration of origins of the twist. The ''plane'' of the initial director reorientation is supposed to be in the plane ($l_\parallel, z$); E_c: circularly polarized input wave; $\mathbf{E}_e$: elliptically polarized output wave (with principal axis ξ); Δ: relative phase shift between ordinary and extraordinary waves, η: principal axis of the output ellipse of a copropagating opposed circularly polarized wave.

reorientation [say, in the plane $(l_\parallel, z)$, Fig. 4]. Consider an input wave $\mathbf{E}_c$ which is *initially* circularly polarized. The corresponding ordinary and extraordinary components of $\mathbf{E}_c$ will propagate in perturbed NLC with different phase velocity. Thus, along the propagation, the circular polarization (at $z=0$ interface) will be transformed into elliptical polarization $\mathbf{E}_e$ (near the output $z=L$ interface), with a principal axis $\boldsymbol{\xi}$, which is tilted at 45 degrees, say in the $+l_\parallel$, $+l_\perp$ quarter. Note that the ellipticity will be defined by the relative phase shift Δ between ordinary and extraordinary waves. Recall also that the dielectric torque of the light (exerted on the director) is very sensitive to the polarization state of light. This would give rise a ''preferred direction'' of reorientation (parallel with ξ) near the output $z=L$ interface. At the same time, there is an azimuthal symmetry (around the z axis) near to the input plane $z=0$, since we have a circularly polarized input beam. The above-mentioned ellipticity axis (near the $z=L$) will try to reorient the director from the plane $(l_\parallel, z)$ towards a new plane, which would contain the direction $\boldsymbol{\xi}$. Any small azimuthal reorientation of the initial plane of the director will immediately create a new preferred direction, which will make 45° with respect to the new plane of the director reorientation, etc. Two important consequences follow due to this effect. First, persistent director precessions may be achieved in the dynamically stabilized regime. Second, the driving torque is always applied asymmetrically (from the side of the output $z=L$ plane), which will give rise the twisting effect.

Let us suppose now that we have another copropagating (with the first wave) circularly polarized wave. Then, the *same* director deformation would lead to the formation of another preferred direction $\boldsymbol{\eta}$ (near to the $z=L$ interface, Fig. 4), which will be crossed with $\boldsymbol{\xi}$, if two copropagating waves had opposed circularity at the input. Thus, director plane deformations would be favored, since the two preferred directions ($\boldsymbol{\xi}$ and $\boldsymbol{\eta}$) will compensate each other. These phenomena are predicted and mathematically described by our model.

C. Linear polarization case

Now, the principal optical axis of the quarter-wave plate $\lambda_1/4$ is chosen so that it does not change the polarization states of two linearly cross-polarized noncoherent copropagating beams.

Here also a LIFT is observed above a certain threshold value of the total intensity, since the initial director orientation in this geometry is normal to both incident linear polarizations (as in Sec. III B). The observed threshold value depends on the parameter R (Fig. 5, circles), as was predicted by our model. We emphasize that the total intensity of the combined beam depends on R, while the transition is initiated and controlled only by the stronger excitation field. The corresponding theoretical curve is presented in the same figure. We use the same material parameters as for the previous fit and we obtain excellent quantitative agreement of experimental data with the Eq. (30) without any adjustment. As one can see, the threshold value at $R=1$ (circular symmetry) is twice as high as the one corresponding to $R=0$ (single linearly polarized beam), confirming our theoretical predictions. Finally, the above-threshold director configuration is found to be quite stable, also confirming our theoretical predictions [see Eq. (24)].

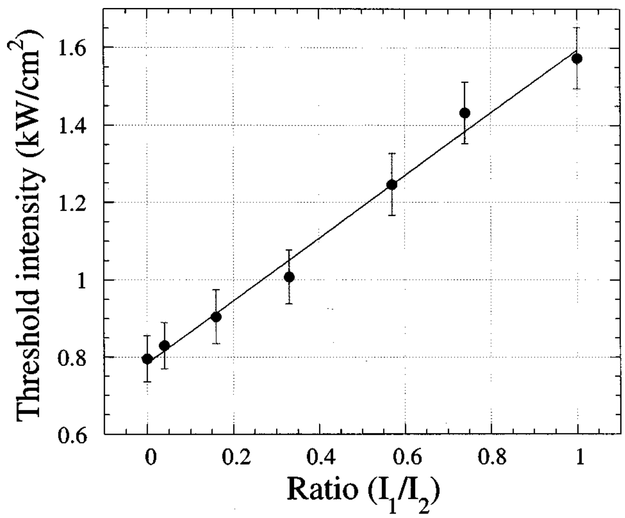

FIG. 5. Dependence of the threshold power for the light-induced Fréedericks transition on the intensity ratio $R=I_1/I_2$ of two cross polarized noncoherent copropagating beams. The line is a theoretical fit by the formula (30).

IV. SUMMARY

The combination of two noncoherent beams enabled us to separately study the roles of the EMF symmetry and angular momentum in optically induced Fréedericksz transition. We have established (both theoretically and experimentally) several important characteristics of the collective molecular reorientation behavior. Thus, the director precession rate (Ω) may be controlled by the AM of input EMF. However, this control is coupled with configurational changes, in particular, with orientational twist deformation. The angular momentum of the light does not change the LIFT threshold, while the azimuthal symmetry of EMF may change it twice, etc.

A rich variety of multistability phenomena have been observed in our experiment in both static and dynamic excitation regimes. However, we have presented here the experimental results corresponding to relatively small reorientations, which allowed us to obtain analytical solutions of our model. The model provides for multistability phenomena as well, but consistent comparison with a corresponding experiment is possible only in terms of numerical simulation. These results are to be presented shortly elsewhere.

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