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Light-induced Frederiks transition in a nonlinear liquid crystal waveguide

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The analysis of a nonlinear optical effect in two waveguides separated by a nematic liquid crystal is presented. The nonlinear effect is caused by a light-induced reorientation occurring in a homeotropic or planar oriented nematic film and can be enhanced by a d.c. external bias (electric) field. It is shown that the nonlinear optical effect has a threshold nature of the light-induced Frederiks transition which is discussed in three classical geometries corresponding to *splay*, *twist*, and *bend* deformations of the nematic liquid crystal.

1. Introduction

Although the first observations of liquid crystalline behaviour were made in 1888 by Reinitzer and Lehmann, the nonlinear optics of liquid crystals (LCs) has been actively studied for not more than the last twenty years. Early studies have concentrated on the isotropic phase in which LCs reveal a very interesting pretransitional (from isotropic to nematic phase) behaviour manifesting itself in great molecular reorientations and macroscopic collective phenomena. In the nematic phase, the correlation among molecules is very strong because of the high anisotropy as well as the collective behaviour of the molecules. This is responsible for the fact that LC molecules can be easily reoriented even with a very low externally applied field.

Additionally, due to

– extraordinary large optical nonlinearities of various forms,

– a large birefringence and good transparency (from near UV to far IR spectrum),

– both liquid and crystalline properties suitable to optoelectronic elements, and

– unlimited possibilities of new syntheses in view of specific nonlinear properties,

liquid crystals are unique nonhomogeneous anisotropic objects for nonlinear optics [1].

The use of liquid crystals leads to the numerous nonlinear optical phenomena arising from molecular reorientation or/and thermal effects [2]–[7] such as intrinsic bistability, temporal instabilities and stochastic processes for light-induced reorientation, nonlinear phenomena on a surface and on boundaries, fluctuations and nonlinear light scattering at phase transitions. All these optical nonlinear phenomena seem to be very promising in applications to optoelectronic waveguided functional elements.

In this paper, we discuss the light-induced Frederiks transition (LIFT) occurring in a nematic liquid crystal (NLC) waveguide structure. The optically-induced reorientation is imposed by the evanescent field of the light guided in the waveguide. Three geometries of the LIFT which correspond to three basic deformations of an NLC *splay*, *twist*, and *bend* are discussed and a comparison with the optical Frederiks transition observed in a homeotropically aligned nematic film with a cw laser beam [8] is presented.

2. Nonlinear optical properties of liquid crystals

Liquid crystals are composed of highly anisotropic and strongly correlated molecules that can be easily reoriented by external fields. Typically, a d.c. field of $E \approx 100\ \mathrm{V\,cm^{-1}}$ or $H \approx 0.1$ T is sufficient to induce a significant molecular reorientation leading to a refractive index change Δn as large as 0.01 to 0.1 [8]. Since an optical field can be treated equivalently to a d.c. field (so long as there is no strong permanent dipole on the molecules), the same refractive index change can be obtained by a laser intensity of about $100\ \mathrm{W\,cm^{-2}}$. Depending on the mechanism involved (electronic or non-electronic), optical properties of LCs can be described by two different models. The electronic nonlinearities of LCs, *i.e.*, $\chi^{(2)}$, $\chi^{(3)}$ for second and third harmonic generations, respectively, require a quantum mechanical approach. On the other hand, the non-resonant nonlinearities arising from one of the three mechanisms:

– optically-induced molecular reorientation, $\Delta n = \dfrac{dn}{d\theta}\Delta\theta$ (θ – reorientation angle),

– thermal effect, $\Delta n = \dfrac{dn}{dS}\dfrac{dS}{dT}\Delta T + \dfrac{dn}{d\rho}\dfrac{d\rho}{dT}\Delta T$ (S – nematic order parameter, T – temperature), or

– density effect, $\Delta n = \dfrac{dn}{d\rho}\Delta\rho$ (ρ – density).

are generally much greater in LCs than electronic nonlinearities and result in a light intensity dependent refractive index change Δn [1].

3. Light-induced Frederiks transition (LIFT)

It is well known that if an external field is applied in a direction perpendicular to the mean molecular alignment defined by a unit vector $\boldsymbol{n}$, the molecular reorientation can only happen when the field is above a certain threshold value. This phenomenon initially observed by Frederiks in early '30 [9] for static electric and magnetic fields is known as the Frederiks transition and exhibits all the characteristic behaviour of a phase transition. The static field Frederiks transitions have been studied in three basic configurations of liquid crystal cells corresponding to three possible deformations: *splay*, *twist*, and *bend* described, respectively, by free energy densities

i) $F_1 = \dfrac{1}{2}K_1(\mathrm{div}\,\boldsymbol{n})^2$,

ii) $F_2 = \frac{1}{2} K_2 (\boldsymbol{n} \cdot \operatorname{rot} \boldsymbol{n})^2$,

iii) $F_3 = \frac{1}{2} K_3 (\boldsymbol{n} \times \operatorname{rot} \boldsymbol{n})^2$

where K_1, K_2 and K_3 are the corresponding elastic (Frank) constants.

In 1981, DURBIN, ARAKELIAN and SHEN [8] demonstrated the evidence of the optical (light-induced) Frederiks transition. They presented both the experimental data and the theoretical calculations of the induced phase shift $\Delta\Phi = \int_{-d/2}^{+d/2} (\omega/c)\Delta n dz$ as a function of the laser intensity I, in a thin film of homeotropically aligned nematic PCB of thickness $d = 250$ μm (Fig. 1), where Δn as a funtion of z arises from the induced molecular reorientation. Because of the strong anchoring of the molecules at the boundary surfaces Δn is not uniform across the film ($\Delta n(z \approx \pm d/2) \approx 0$ and $\Delta n(z = 0) := \max$). In the case of a normally incident laser beam, the homeotropically oriented molecules cannot reorient unless the field intensity is above a threshold value and the transition is of the second order. As it was also demonstrated in [8], the threshold of the LIFT can be greatly reduced with the help of a bias field (d.c. or optical).

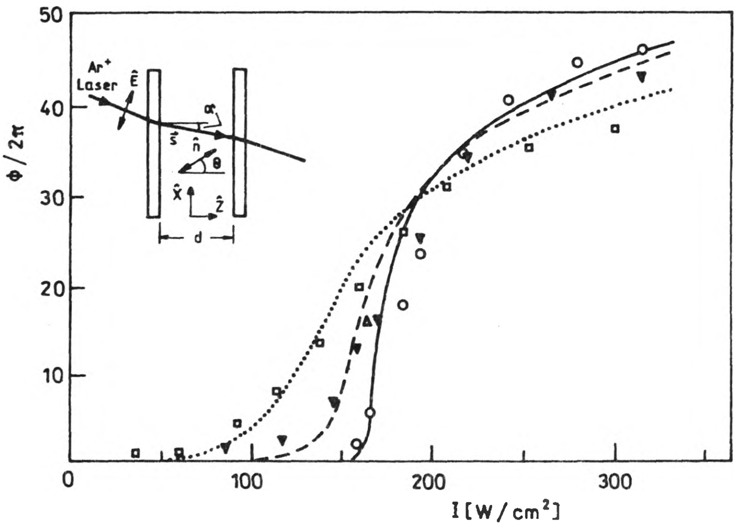

Fig. 1. Experimentally observed laser-induced phase shift in an NLC (PCB) film experienced by a probe beam as a result of the refractive index change induced by a pump laser along with theoretical curves. Circles and solid curve correspond to the normal incidence ($\alpha = 0°$) of the pump beam, hence to the light-induced Frederiks transition with the threshold of about 155 W/cm^{-2} for the cell thickness $d = 250$ μm (from [8])

The next section presents an analysis of the LIFT in a waveguide configuration. The main difference between cell (mentioned above) and waveguide configuration is limited to the origin of the optical field. In the waveguide configuration, molecular reorientation is induced through the evanescent field of the light guided in the waveguide structure while, in the cell configuration there is a normally incident laser beam. In both cases, the optical field is in a conflict with orientations imposed by surfaces. In addition to the optically-induced reorientation we will also discuss an influence of a d.c. external bias (electric) field which enhances the action of the optical field. Both fields cause changes in the effective refractive index of the NLC layer due to the optical nonlinearity and due to the direct action of the bias field.

4. LIFT in a waveguide configuration

We consider a step-index single-mode lossless planar optical waveguide. These assumptions cause that the guided electromagnetic field is homogeneous in one direction perpendicular to the propagation direction and confined in the second direction. The cover of the waveguide consists of an NLC layer with a positive optical anisotropy (*i.e.*, the extraordinary refractive index of a liquid crystal $n_\parallel$ is greater than the ordinary refractive index $n_\perp$: $n_\parallel > n_\perp$). A transversal component of the electric field of the optical wave guided in the waveguide structure (see Fig. 2) is given by [10]

$$E_{opt}(x,z) = \Psi(x) - \exp(-ik_0 n_{eff} z) \tag{1}$$

where $k_0 = 2\pi/\lambda$, n_{eff} is an effective refractive index of the guided mode, and $\Psi(x)$ is a waveguide mode field profile. In the nematic cover the electric field has a form

$$\Psi(d > x > 0) = A_0 \exp(-\gamma x) \tag{2}$$

where A_0 is an amplitude and $\gamma = k_0\sqrt{n_{eff}^2 - n_\perp^2}$ is a transversal component of the wavevector. The intensity of light guided by the analyzed structure is defined as follows:

$$I = \sqrt{\frac{\varepsilon_0}{\mu_0}} \int\limits_{-\infty}^{+\infty} \Psi^2 dx.$$

Additionally, in the parallel direction to the electric field of the electromagnetic wave, a homogeneously distributed static electric field characterized by its average value E_{st} is applied to the NLC layer. Both fields: the strong electric field of the light wave E_{opt} and the static electric field E_{st} rotate nematic molecules to fulfil the condition of the free energy density minimization. In this work, we analyze three possible configurations of the nonlinear waveguide with the NLC cover (see Fig. 2) corresponding to three basic deformations of an NLC:

1) planar NLC texture and a TM-polarized electromagnetic wave (*splay* deformation),

2) planar NLC texture and a TE-polarized electromagentic wave (*twist* deformation),

3) homeotropic NLC texture and a TE-polarized electromagnetic wave (*bend* deformation).

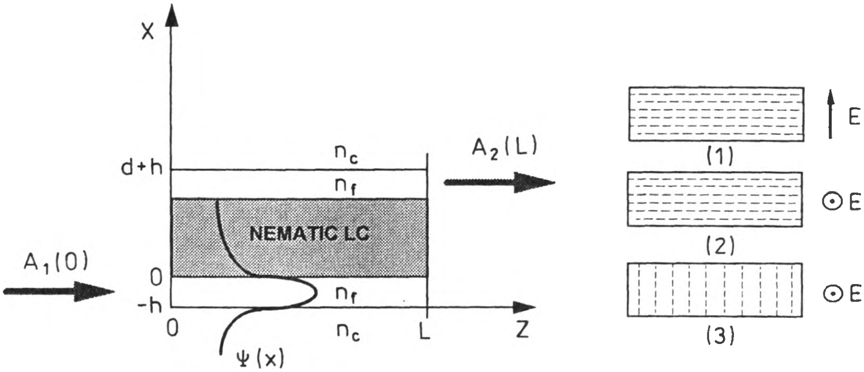

Fig. 2. Schematic drawing of the analyzed three configurations of the nonlinear waveguide structure with the NLC layer: 1 — planar NLC texture and a TM-polarized electromagnetic wave (*splay* deformation), 2 — planar NLC texture and a TE-polarized electromagnetic wave (*twist* deformation), 3 — homeotropic NLC texture and a TE-polarized electromagnetic wave (*bend* deformation)

The reorientation angle θ of the liquid crystal molecules is calculated from the Euler–Lagrange equations which describe minimization of the free energy. For the analyzed configurations the equations take the form

$$\frac{d^2\theta}{dx^2}[(k_j-1)\sin^2\theta+1]+\frac{1}{2}\left(\frac{d\theta}{dx}\right)^2(k_j-1)\sin 2\theta+\eta_j[A_0^2\exp(-2\gamma x)+2E_{st}^2]\sin 2\theta=0 \tag{3}$$

where $j=1$ for *splay*, $j=2$ for *twist*, and $j=3$ for *bend* geometries, k_j is the elastic constants ratio: $k_1=K_{33}/K_{11}$, $k_2=1$, $k_3=K_{11}/K_{33}$, and η_j is a normalizing coefficient defined by the formula

$$\eta_j=\frac{(n_\parallel^2-n_\perp^2)\varepsilon_0}{4K_{jj}}. \tag{4}$$

In the one-elastic-constant approximation ($K_{11}=K_{22}=K_{33}$, *i.e.*, $k_j=1$ for all configurations) and for small angles (*i.e.*, $\sin\theta\approx\theta$ and $|(d\theta/dx)^2(k_j-1)| \ll \eta_j[A_0^2e^{-2\gamma x}+2E_{st}^2)]$ Eq. (3) yields

$$\frac{d^2\theta}{dx^2}+\eta_j[A_0^2\exp(-2\gamma x)+2E_{st}^2]\theta=0, \tag{5}$$

which can be solved analytically in terms of the Bessel functions J_v and Y_v:

$$\theta(x)=b_1J_v(\sqrt{\eta_j}A_0\gamma^{-1}e^{-\gamma x})+b_2Y_v(\sqrt{\eta_j}A_0\gamma^{-1}e^{-\gamma x}) \tag{6}$$

where b_j ($j=1$, 2) are constants and $v^2=-2\eta_jE_{st}^2/\gamma^2$.

The solution (6) allows us to determine the threshold value for the light-induced orientation effect (LIFT). For light intensities above the threshold the larger reorientation angles appear and the numerical integration of Eq. (3) is necessary.

Two types of the boundary conditions corresponding to two different regimes of interactions between nematic molecules and limiting glass surfaces located at $x = 0$ and $x = d$ can be taken into consideration.

1. In the regime of strong interactions the molecules are rigidly anchored at surfaces, hence the boundary conditions $\theta(0) = \theta(d) = 0$ (strong anchoring conditions) are fulfilled. For a small thickness d ($\gamma d \ll 1$), the threshold intensity $\eta_j I_{th} \sim \eta_j A_0^2 \approx (\pi/d)^2 - 2\eta_j E_{st}^2$. However, for larger thicknesses d ($\gamma d \gg 1$) the threshold intensity becomes independent of the thickness and for $E_{st} = 0$ is approximated by the first root of J_0 Bessel function: $\eta_j A_0^2 \approx (2.405\gamma)^2$.

2. In the regime of weak anchoring at $x = 0$ interface, the boundary conditions $d\theta/dx(0) = \theta(d) = 0$ are applied and then the threshold intensity is approximated for a small thickness d by the condition: $\eta_j A_0^2 \approx (\pi/2d)^2 - 2\eta_j E_{st}^2$.

The molecular reorientation calculated from Eq. (3) with proper boundary conditions leads to changes in the local electric permittivity

$$\delta\varepsilon = \frac{n_\perp^2}{\cos^2\theta + (n_\perp/n_\parallel)^2 \sin^2\theta} - n_\perp^2 \tag{7}$$

and this causes a change in the guided mode effective refractive index which finally is dependent on the intensity of external electric field (electrooptic effect) and the light intensity (optical nonlinear effect).

5. Results and conclusions

Results presented in Figures 3–5 have been obtained for the planar waveguide structure with $n_f = 1.74$, $n_c = 1.52$, $n_s = 1.72$, $h = 2$ μm (see Fig. 2) and for the wavelength $\lambda = 1.06$ μm. The nematic liquid crystal chosen for the calculations was PCB (nematic range from 21 °C to 35 °C) characterized by refractive indices $n_\perp = 1.52$, $n_\parallel = 1.72$, and by elastic constants of the order of 10^{-12} N (at 21 °C: $K_{11} = 12.9 \times 10^{-12}$ N, $K_{22} = 6.7 \times 10^{-12}$ N, $K_{33} = 17.8 \times 10^{-12}$ N) which causes that the coefficient η is of the order of 1.5 V^{-2}. As it is schematically sketched in Fig. 2, the evanescent field E_{opt} in the nematic cover, enhanced by the external electric field E_{st}, is always perpendicular to the initial mean molecular orientation (molecular director) and this direction is described by the refractive index $n_\perp \equiv n_c$. The strong anchoring conditions are taken into account.

Figures 3**a**,**b** present variations of the reorientation angle θ along the NLC layer, for various intensities ηI above the threshold value of the LIFT in the absence of external electric field $E_{st} = 0$ (Fig. 3**a**) and for various values of the static electric field E_{st} for the light intensity $\eta I = 10\ \Omega^{-1}\mu\mathrm{m}^{-1}$ (Fig. 3**b**). The data were obtained by numerical integration of Eq. (3) for $k_j = 1$ (in the one-elastic-constant approximation or twist deformation) and for the thickness of the NLC layer $d = 2$ μm. The largest reorientation effect (close to $\pi/2$) exhibits near the border $x = 0$.

Figure 4**a** presents the guided light intensity threshold of the LITF *vs.* the thickness of the NLC layer plotted for selected values of the external electric field, while in Fig. 4**b**, the threshold was plotted *vs.* the electric field, for selected thick-

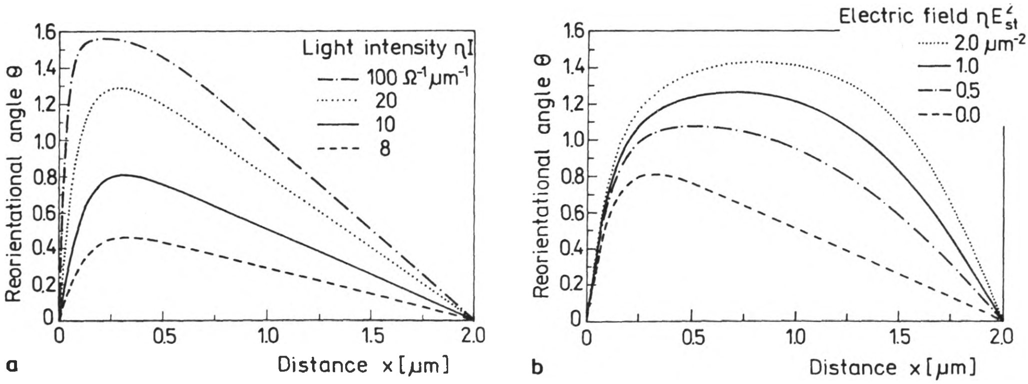

Fig. 3. Reorientation angle θ along the NLC layer of the thickness $d = 2$ μm: **a** — for various intensities ηI above the threshold value of the light-induced Frederiks transition and the electric static field $E_{st} = 0$, **b** — for various electrical static fields E_{st} and for the light intensity $\eta I = 10\ \Omega^{-1}\mu\mathrm{m}^{-1}$

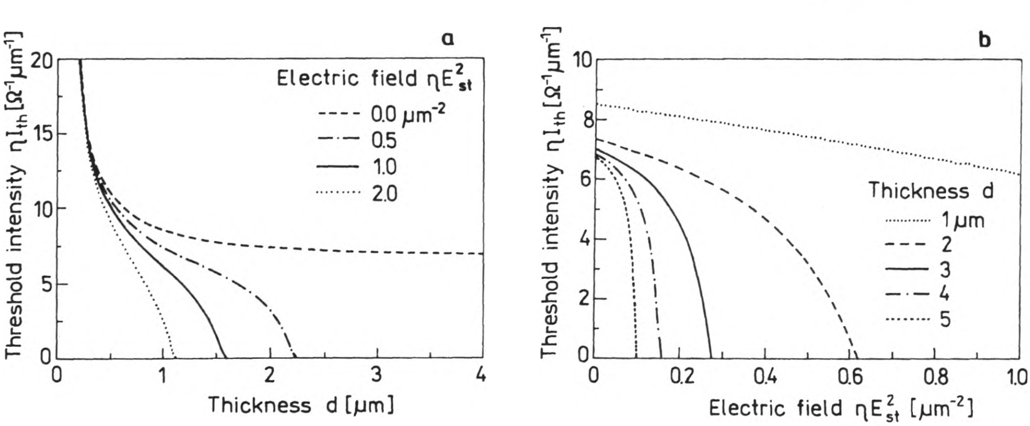

Fig. 4. Threshold intensity of the LIFT : **a** — *vs.* the thickness of the NLC layer for selected values of the external electric field E_{st}, **b** — *vs.* the external electric field E_{st} for various NLC layer thicknesses d. The elastic constants ratio $k_j = 1$

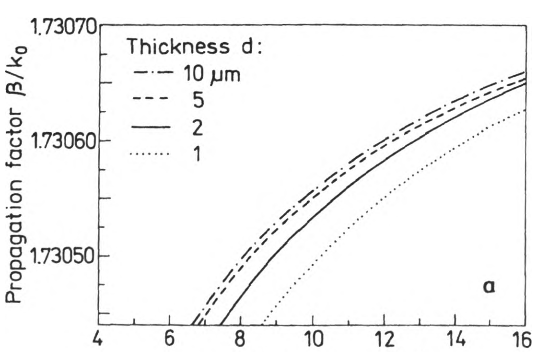

Fig. 5a

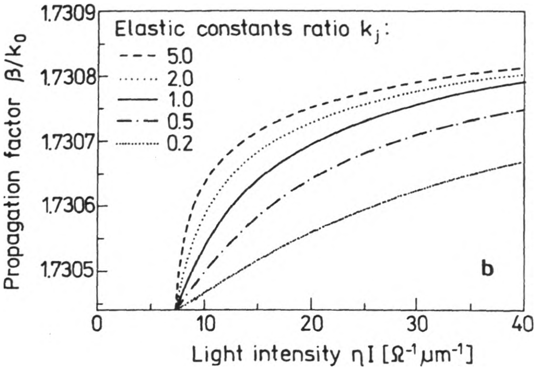

Fig. 5b

Fig. 5. Propagation factor β changes *vs.* the input light intensity ηI: **a** — for various thicknesses d and the elastic constants ratio $k_j = 1$, **b** — for various elastic constants conditions at three geometries: *splay* ($k_j = 2, 5$), *twist* or one-elastic-constant approximation ($k_j = 1$), *bend* ($k_j = 0.5, 0.2$), and for the thickness $d = 2$ μm. Threshold behaviour of the LIFT is clearly visible

nesses of the nematic layer. The dependence of the threshold intensity on the thickness d as well as on the electric field E_{st} for small thicknesses ($d < 1$ μm) and for large thicknesses ($d > 2$ μm) is in agreement with theoretical predictions for strong anchoring conditions.

Figure 5 presents propagation factor β defined (for a small perturbation) in form [10]

$$\beta_l = k_0\left[n_{\mathrm{eff}}^{(l)} + \frac{1}{2n_{\mathrm{eff}}^{(l)}}\,\frac{\int\limits_0^d \delta\varepsilon \Psi^2(x)dx}{\int\limits_{-\infty}^{+\infty} \Psi^2(x)dx}\right] \tag{8}$$

($l = 1, 2$) plotted against the input light intensity for various thicknesses d (Fig. 5**a**) and for various elastic constants ratio (Fig. 5**b**). For thicker NLC layers the reorientation of molecules in the centre of the layer is less dependent on the forces at liquid crystal/dielectric waveguide boundary and then changes of the propagation factor appear for lower intensities (see Fig. 5**a**). In Figure 5**b**, the elastic constants ratio $k_j > 1$ corresponds to splay deformations and $k_j < 1$ corresponds to bend deformations since typically the following relation occurs: $K_{11} < K_{33}$. Note that the light intensity $\eta_j I$ is normalized to the elastic constant corresponding to the type of deformation. Therefore, in the splay geometry an increase of the ratio $k_1 = K_{33}/K_{11}$ is followed by an increase of the bend elastic constant K_{33} and, consequently, in the bend geometry an increase of the ratio $k_3 = K_{11}/K_{33}$ is followed by an increase of the splay deformation constant K_{11}. The case $k_j = 1$ corresponds to the twist deformation or to the one-elastic-constant approximation.

Finally, we have obtained the light-induced Frederiks transition with the nematic liquid crystal waveguide structure for various geometries and selected polarizations of incoming light. It has been shown that the external electric field enhances the nonlinear optical effect and lowers the threshold value of the LIFT. The same effect could be obtained by using the magnetic field. The results obtained have been applied to analyze the performance of a nonlinear liquid crystalline directional coupler [11].

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