Light-induced Freedericksz transition and optical multistability in nematic liquid crystals

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Abstract

We revisit the problem of light transmission through a slab of homeotropically oriented nematic liquid crystal and solve *self-consistently* a system of coupled nonlinear equations describing orientation of the nematic director in the slab and the Maxwell equation for the electric field. We demonstrate that optical transmission of the slab as a function of input power shows a multistable hysteresis-like behaviour. We suggest that this multistability can be useful for creating tunable all-optical switching devices based on the liquid-crystal infiltration of photonic crystals.

Keywords: optical Freedericksz transition, liquid crystal, optical bistability

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1. Introduction

Liquid crystals (LCs) play an important role in the modern technology and are used for numerous applications in electronic imaging, display manufacturing and optoelectronics [1,2]. A large variety of electro-optical effects that occur in LCs could be employed when designing photonic devices. For example, a property of LC to change its orientational structure and the refractive index in the presence of static electric fields suggests one of the most attractive and practical schemes for tuning photonic bandgap devices [3,4]. Nonlinear optical properties of LCs and multistability of their light transmission are of a great interest for the future applications of LCs in photonics [5].

Light polarized perpendicular to the LC director changes its orientation provided that the light intensity exceeds some threshold value [6]. This effect is widely known as *a light-induced Freedericksz transition* (LIFT). Its theory has been developed more than two decades ago in a number of pioneering papers [7–9]. In particular, Zeldovich *et al.* [7] have demonstrated that LIFT could generally be treated as a second-order orientational transition, though hysteresis-like dependences and two thresholds can be observed in some types of LCs, which concern increasing and decreasing intensities of the input light. The results obtained later by Ong [10] have confirmed that LIFT for MBBA nematics is of the second order and no hysteresis behaviour is observed, whereas LIFT for PAA nematics is of the first order and a hysteresis-like behaviour, with two distinct thresholds, should be observed for a single beam at normal incidence. Although these conclusions have been confirmed to some extent in later experiments [10], the theories developed earlier have been based on the approximations of geometric optics and they are approximate by their nature. Similar approximations have been used later [11] for taking into account a backward wave in the LC film placed in Fabry-Perot resonator. It has been shown that the threshold of LIFT depends periodically on the thickness of LC slab. Nonlinear optical properties of nematic LC films placed into Fabry-Perot interferometer have been studied by Khoo *et al*. [12]. They have considered propagation of light polarized under acute angle with respect to the LC director and experimentally observed a bistability in the output light intensity caused by a giant nonlinearity of LC films. Cheung *et al.* [13] have detected experimentally the effects of multistability in the similar systems, including the oscillations of the output light intensity.

In this paper, we revisit this classical problem and solve numerically, for the first time to our knowledge, a complete set of coupled equations for the nematics and the propagating electric field. We demonstrate a possibility for optical bistability occurring after LIFT takes place. This provides a straightforward verification of the earlier results and also builds a solid background for the studies of more complex photonic structures that employ orientational nonlinearity of LCs.

First, we consider a general problem of the light transmission through homeotropically oriented nematic LC and analyze specific conditions for the multistability and LIFT. Second, we consider this problem self-consistently, by solving numerically the coupled system of stationary equations for the director orientation and the Maxwell equations for the electric field. We present our results for two different nematic LCs, para-azoxyanisole (PAA) and a mixture E7, which have been shown in the previous theoretical studies [10] to demonstrate quite dissimilar behaviours at the LIFT.

The paper is organized as follows. Sections 2 and 3 present our basic equations and outline our numerical approach. Section 4 summarizes the numerical results and compares them for the two nematic LCs. In addition, both the bistability and the hysteresis-type behaviours of the light transmission are in detail discussed in this section. Finally, Section 5 concludes the paper.

2. Basic equations

Consider a nematic LC confined between two vertical planes ($z=0$ and $z=L$), with the director initially oriented along the z axis (see Fig. 1). The LC slab interacts with a normally incident monochromatic electromagnetic wave characterized by the electric field $\mathbf{E}(\mathbf{r},t)$,

$$\mathbf{E}(\mathbf{r},t)=\frac{1}{2}\left[\mathbf{E}(\mathbf{r})e^{-i\omega t}+\mathbf{E}^*(\mathbf{r})e^{i\omega t}\right]. \quad (1)$$

In order to derive basic equations for the nematic director and the electric field, we write the free energy of LC in the presence of electromagnetic wave in the form [7]

$$F=\int(f_{\mathrm{el}}+f_{\mathrm{E}})dV, \quad (2)$$

where

$$f_{\mathrm{el}}=\frac{K_{11}}{2}(\nabla\cdot\mathbf{n})^2+\frac{K_{22}}{2}(\mathbf{n}\cdot\nabla\times\mathbf{n})^2+\frac{K_{33}}{2}(\mathbf{n}\times\nabla\times\mathbf{n})^2,$$

$$f_{\mathrm{E}}=-\frac{1}{8\pi}\varepsilon_{ik}E_iE_k^*, \qquad \varepsilon_{ik}=\varepsilon_\perp\delta_{ik}+\varepsilon_a n_i n_k.$$

Here f_{el} is the elastic energy density for the LC, f_{E} the contribution to its free energy density from the light field, $\mathbf{n}$ the nematic director, K_{ii} the elastic constants, ε_{ik} the dielectric permittivity tensor, while $\varepsilon_a=\varepsilon_\parallel-\varepsilon_\perp>0$ describes the LC response anisotropy, with $\varepsilon_\parallel$ and $\varepsilon_\perp$ being respectively the principal components of the ε_{ik} tensor parallel and perpendicular to the director.

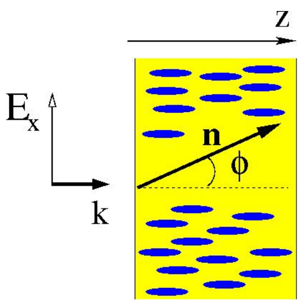

Fig. 1. Schematic representation of the problem: a slab of LC is placed between two walls ($z=0$ and $z=L$). The $\mathbf{n}$ vector describes orientation of molecules in the slab.

We assume that the electric field outside the LC slab is directed along the x axis (see Fig. 1) and it causes director reorientation in the xz plane inside the LC slab. Therefore, all the functions inside the LC slab would depend only on the z coordinate, so that we may look for spatial distribution of the nematic director in the following form: $\mathbf{n}(\mathbf{r})=\mathbf{e}_x\sin\varphi(z)+\mathbf{e}_z\cos\varphi(z)$, where φ is the angle between the LC director and the z axis (see Fig. 1) and $\mathbf{e}_x$ and $\mathbf{e}_z$ the unit vectors of the Cartesian frame.

After minimizing the free energy given by Eq. (2) with respect to the director angle φ, we obtain the stationary equation for the LC director orientation in the presence of light field:

$$(K_{11}\sin^2\varphi+K_{33}\cos^2\varphi)\frac{d^2\varphi}{dz^2}=(K_{33}-K_{11})\sin\varphi\cos\varphi\left(\frac{d\varphi}{dz}\right)^2 -\frac{\varepsilon_a\varepsilon_\parallel\varepsilon_\perp}{16\pi}\frac{\sin 2\varphi}{(\varepsilon_\perp+\varepsilon_a\cos^2\varphi)^2}|E_x|^2, \quad (3)$$

where we have taken into consideration that the electric field inside the LC slab has the longitudinal component only, $E_z(z) = -(\varepsilon_{xz}/\varepsilon_{zz})E_x(z)$.

From the Maxwell equations, one can obtain scalar equation for the x component of the electric field,

$$\frac{d^2E_x}{dz^2} + k^2 \frac{\varepsilon_\perp \varepsilon_\parallel}{\varepsilon_\perp + \varepsilon_a \cos^2 \varphi} E_x = 0, \quad (4)$$

where $k = 2\pi\lambda/c$ and λ is the wavelength of the incident light. The time-averaged z component of the Poynting vector, $S_z = c/(8\pi)E_x H_y^*$, remains unchanged inside the LC slab [7,10] and so it could be employed for characterizing different regimes of the nonlinear transmission of light.

3. Numerical approach

We solve the system of coupled nonlinear equations (3) and (4) *in a self-consistent way*, taking the appropriate boundary conditions into account. For the director, we assume a strong LC anchoring at the boundaries, i.e.

$$\varphi(0) = \varphi(z) = 0, \quad (5)$$

whereas for the electric field we consider the reflecting conditions

$$E_x(0) = E_{\text{in}} + E_{\text{ref}}, \qquad E_x(L) = E_{\text{out}}. \quad (6)$$

Here E_{in}, E_{ref} and E_{out} denote the amplitudes of the incident, reflected and transmitted waves, respectively. In all of the above equations we put $\mu = 1$ for the magnetic susceptibility and consider that $H_y = (1/ik)(dE_x dz)$.

The boundary conditions expressed by Eqs. (6) imply that two counter-propagating waves are considered on the left side of the LC slab, the incoming and reflected ones, whereas only one outgoing wave appears on the right side. Therefore, we first fix the amplitude of the outgoing wave E_{out}, in order to solve this nonlinear eigenvalue problem. This allows us to determine unique values of the incident (E_{in}) and reflected (E_{ref}) waves.

Eq. (3) for the director is similar to that describing a nonlinear pendulum, with the fixed boundary conditions given by Eq. (5). This means that we should search for periodic solutions, with the period $2L$. In fact, there exist many periodic solutions of Eq. (3). First of all, this is a trivial solution $\varphi(z) = 0$ that corresponds to unperturbed orientation of the director and absolute minimum of the free energy given by Eq. (2). The LIFT occurs when this trivial solution gets unstable for larger values of the input light intensity and the value of the director angle $\varphi(z)$ becomes finite. We find this solution numerically by using the well-known *shooting method* [15]. By fixing the amplitude of the outgoing wave E_{out} and taking $\varphi(L) = 0$ at the right boundary, we find the values of derivative $d\varphi / dz_{z=L}$ such that a vanishing value of the director angle at the left boundary, $\varphi(0) = 0$, is obtained after integration. After analyzing the nonlinear equation (3) in the two-dimensional phase space, one can show that the corresponding solution lies just below the separatrix curve and it has no node between the points $z = 0$ and $z = L$. This observation allows us to reduce significantly the parameter region for the required derivative $d\varphi / dz_{z=L}$ values. Then we apply stability analysis for the zero solution, which gives us the threshold value.

4. Results and discussions

We have solved the nonlinear transmission problem for the parameters of two nematic LCs, para-azoxyanisole (PAA) and a mixture E7. They are characterized by different signs of the parameter $B = (1 - 9\varepsilon_a / (4\varepsilon_\perp) - (K_{33} - K_{11}) / K_{33}) / 4$ appearing in the geometric optics approximation [7,10], where the sign of B determines the order of the LIFT. For the PAA, $B < 0$ is found and the LIFT should be of the first order, while we have $B > 0$ for the LC E7 and the corresponding transition should be of the second order. Below we will verify these results by applying our self-consistent approach. We take the following physical parameters: (a) $K_{11} = 9.26 \cdot 10^{-7}\,\text{dyn}$, $K_{33} = 18 \cdot 10^{-7}\,\text{dyn}$, $n_0 = 1.595$ and $n_e = 1.995$ for PAA and (b) $K_{11} = 11.09 \cdot 10^{-7}\,\text{dyn}$, $K_{33} = 15.97 \cdot 10^{-7}\,\text{dyn}$, $n_0 = 1.522$ and $n_e = 1.746$ for E7. A single wavelength ($\lambda = 532$ nm) is considered for both LCs.

In Fig. 2 we present our numerical results for the changes in the maximum orientation angle $\varphi_{\max}$ of the director as a function of normalized input power I / I_F calculated for the PAA and E7. The threshold power I_F (for instance,

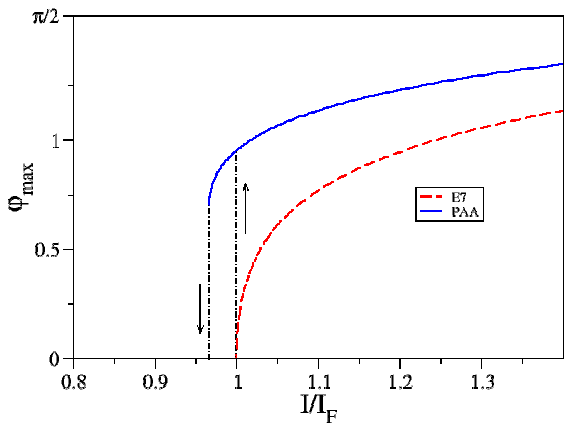

Fig. 2. Dependence of the maximum deformation angle $\varphi_{\max}$ vs. the normalized input intensity for the two types of LCs, E7 and PAA.

I_F = 0.152 MW/cm^2 for the E7) corresponds to the critical value when the zero solution becomes unstable. A hysteresis-like dependence of the angle $\varphi_{\max}$ is observed for the PAA. In the case of LC E7, this dependence exhibits a smooth second-order transition, without any hysteresis. The dependences represented in Fig. 2 are rather universal. They are independent of the LC cell thickness L or the refractive index n_s of the surrounding medium. Yet, in order to be more realistic, one should correct these dependences by the effect of light absorption in the slab, which is not accounted for in this study.

The light transmission versus the input power for the case of LC E7 is shown in Fig. 3. It is evident from Fig. 3 that variations in the refractive index of the surrounding medium n_s cause transmission oscillations with respect to the incident power, in accordance with the Fabry-Perot phenomenon. Depending on the LC cell thickness, these oscillations may lead to the appearance of *optical multistability*. This is similar to that peculiar to a nonlinear resonator and is determined by the resonator properties of LC slab of a finite thickness [14]. Here the reflected wave plays an essential role, as it has been shown in Ref. [13] by taking a second counter-propagating wave into account. In frame of our approach, both waves have been considered from the very beginning, when simply solving the full set of the Maxwell equations with the scattering boundary conditions. As expected, we observe variations of the threshold value I_F with respect to the geometrical optics approximation [10].

Thus, the self-consistent approach gives the same results as obtained in framework of the geometric optics for LIFT in nematic LCs. Nevertheless, our approach does not provide a complete solution for the problem. Indeed, we consider the time-averaged problem though, as shown earlier [16,17], taking into account the LC dynamics would be also important, because both the transverse and azimuthal instabilities may take place. Moreover, our discussion is based on the assumption that the incident laser light is

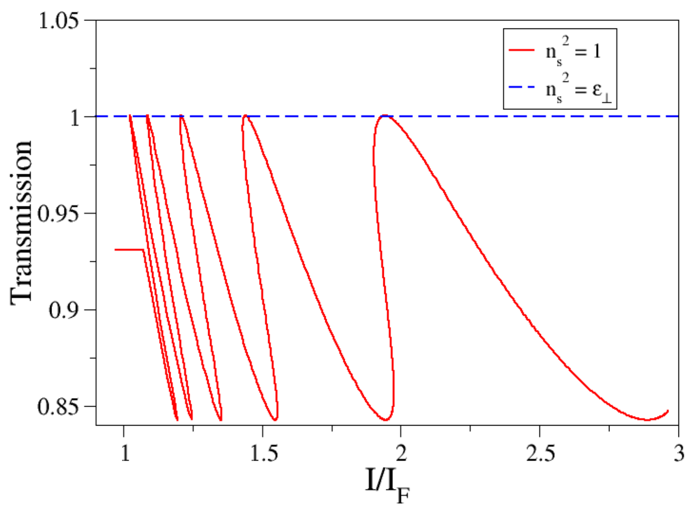

Fig. 3. Light transmission through a slab of LC E7 (the thickness $L = 10 \mu m$) for the two refractive indices of the surrounding media, $n_s = 1$ and $n_s = n_e = \sqrt{\varepsilon_\perp}$. Optical mulitstability is observed in the former case and the latter case corresponds to the condition of perfectly matched refractive indices, when the reflection is absent.

described by a plane wave. In reality, lasers yield focused beams with the sizes comparable to the slab thickness, and this might lead to the appearance of transverse effects. As a consequence, our approach may be considered as a first step towards a clear understanding of the problem. It can be further generalized as to accounting for all of these phenomena, which will shed light on the entire process of LIFTs.

5. Conclusions

We have analyzed the transmission of light through a slab of homeotropically oriented nematic LC and have studied in detail both the optical multistability and LIFT. We have solved numerically the stationary version of the coupled equations for the nematic director and the propagating electric field, taking specific parameters of two different LCs (PAA and E7) as examples. It is shown that this self-consistent approach confirms the main results obtained earlier in the framework of geometric optics approximation.

We have also demonstrated that the resonator effect of the LC slab associated with light reflection from the two boundaries gives a significant effect and, in particular, it is responsible for the observed periodic dependence of the threshold values and the multistability of transmitted light on the slab thickness. We expect that these features may be useful while studying periodic photonic structures with the holes infiltrated by LCs [18]. Multiple reflections and nonlinear LIFT should be properly taken into account in the latter case, in order to develop tunable all-optical switching devices based on the specific nonlinear and tunable properties of LCs.

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