Light-induced multistability and Freedericksz transition in nematic liquid crystals

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We study light transmission through a homeotropically oriented nematic liquid crystal cell and solve self-consistently a nonlinear equation for the nematic director coupled to Maxwell’s equations. We demonstrate that above a certain threshold of the input light intensity, the liquid-crystal cell changes abruptly its optical properties due to the light-induced Freedericksz transition, demonstrating multistable hysteresis-like dependencies in the transmission. We suggest that these properties can be employed for tunable all-optical switching photonic devices based on liquid crystals.

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I. INTRODUCTION

Liquid crystals (LCs) play an important role in the modern technologies being used for numerous applications in electronic imaging, display manufacturing, and optoelectronics [1, 2]. A large variety of electro-optical effects that may occur in LCs can be employed for a design of photonic devices. For example, the property of LCs to change its orientational structure and the refractive index in the presence of a static electric field suggests one of the most attractive and practical schemes for tuning the photonic bandgap devices [3, 4]. Nonlinear optical properties of LCs and multistability of 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 the light intensity exceeds some threshold value [6]. This effect is widely known as the light-induced Freedericksz transition (LIFT), and its theory was developed more than two decades ago in a number of the pioneering papers [7, 8, 9]. In particular, Zel’dovich et al. [7] demonstrated that the light-induced Freedericksz transition can generally be treated as the second-order orientational transition, but in some types of LCs hysteresis-like dependencies and two thresholds can be observed, for the increasing and decreasing intensity of the input light. The results obtained later by Ong [10] confirmed that for the MBBA nematics the Freedericksz transition is of the second order and there is no hysteresis behavior, whereas for the PAA nematics the Freedericksz transition is of the first order and the hysteresis-like behavior with two distinct thresholds should be observed. Although these conclusions have been confirmed to some extent in later experiments [11], the theory developed earlier was based on the geometrical optics and by its nature is approximate. The similar approximation was used later [11] for taking into account a backward wave in a LC film placed in a Fabry-Perot resonator, where it was shown that the threshold of the Freedericksz transition depends periodically on the LC cell thickness.

Nonlinear optical properties of a nematic LC film in a Fabry-Perot interferometer was studied by Khoo et al. [12], who considered the propagation of light polarized under an acute angle to the LC director and observed experimentally bistability in the output light intensity caused by giant nonlinearity of the LC film. Cheung et al. [13] observed experimentally the effects of multistability in a similar system, including oscillations of the output light intensity.

However, in spite of numerous theoretical studies and experimental observations, a self-consistent theory of the light-induced Freedericksz transition based on a systematic analysis of the coupled equations for the nematic director and electromagnetic field is still missing. Therefore, the purpose of this paper is twofold. First, we consider a general problem of the light transmission through a homeotropically-oriented nematic LC and analyze the specific conditions for the multistability and light-induced Freedericksz transition, for possible applications in all-optical switching photonic devices. Second, for the first time, we consider this problem self-consistently and solve numerically a coupled system of the stationary equations for the director and Maxwell’s equations. We present our results for two kinds of nematic liquid crystal, para-azoxyanisole (PAA) and Np-methoxybenzylidene-np-butylaniline (MBBA), which show quite dissimilar behavior of the nematic director at the Freedericksz transition in the previous theoretical studies [10], and also discuss light transmission and bistability thresholds as functions of the cell thickness.

The paper is organized as follows. Sections II and III present our basic equations and outline our numerical approach. Section IV summarizes our results for two kinds of nematic liquid crystal and discusses in detail both bistability and hysteresis-type behavior of the light transmission. Section V concludes the paper.

II. BASIC EQUATIONS

We consider a nematic LC cell confined between two planes \((z = 0 \text{ and } z = L)\) with the director initially oriented along the \(z\) axis (see Fig. I). The LC cell interacts with a normally incident monochromatic electromagnetic
wave described by the electric field \( \mathbf{E}(r, t) \),

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

(1)

FIG. 1: (colour online) Schematic representation of the problem. A LC cell is placed between two walls \((z = 0, z = L)\), the vector \( \mathbf{n} \) describes the molecules orientation in the cell.

To derive the basic equations, we write the free energy of the LC cell in the presence of the electromagnetic wave as follows \[7\]

\[
F = \int (f_{el} + f_E) dV ,
\]  

(2)

where

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

\[
(\sin^2 \phi + K_{33} \cos^2 \phi) \frac{d^2 \phi}{dz^2} - (K_{33} - K_{11}) \sin \phi \cos \phi \frac{d \phi}{dz} + \frac{\varepsilon_{||} \varepsilon_{\perp}}{16\pi} \sin 2\phi |E_x|^2 = 0 ,
\]  

(4)

where we take into account that, as follows from Maxwell’s equations, the electric vector of the light field inside the LC cell has the longitudinal component \( E_z(z) = -(\varepsilon_{||}/\varepsilon_{\perp})E_x(z) \).

From Maxwell’s equations, we obtain the scalar equation for the z-component of the electric field,

\[
\frac{d^2 E_z}{dz^2} + k^2 \frac{\varepsilon_{||} \varepsilon_{\perp}}{\varepsilon_{\perp} + \varepsilon_a \cos^2 \phi} E_z = 0 ,
\]  

(5)

where \( k = 2\pi \lambda/c \), and \( \lambda \) 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 cell \[6, 10\], and it can be used for characterizing different regimes of the nonlinear transmission.

\[
f_E = -\frac{1}{8\pi} \varepsilon_{ik} E_i E_k^* , \quad \varepsilon_{ik} = \varepsilon_{\perp} \delta_{ik} + \varepsilon_a n_i n_k .
\]

Here \( f_{el} \) is the LC elastic energy density, \( f_E \) is a contribution to the free energy density from the light field, \( \mathbf{n} \) is the nematic director, \( K_{ii} \) are the elastic constants, \( \varepsilon_{ik} \) is the LC dielectric permittivity tensor, \( \varepsilon_a = \varepsilon_{||} - \varepsilon_{\perp} \) describes anisotropy of the LC dielectric susceptibility, where \( \varepsilon_{||} \) and \( \varepsilon_{\perp} \) are the main components of the tensor \( \varepsilon_{ik} \) parallel and perpendicular to the director, respectively.

We assume that outside the LC cell the electric field is directed along the \( x \) axis (see Fig. 1), which can cause the director reorientation in the \( xz \) plane inside the LC cell. When the incident beam is broad, we can describe it as a plane wave, so that all functions inside the LC cell will depend only on the \( z \)-coordinate. Therefore, we can seek the spatial distribution of the nematic director in the form

\[
\mathbf{n}(r) = e_x \sin \phi(z) + e_z \cos \phi(z) ,
\]  

(3)

where \( \phi \) is the angle between the director and the \( z \) axis (see Fig. 1). \( e_x \) and \( e_z \) are the unit vectors in the Cartesian coordinate frame.

After minimizing the free energy \[2\] with respect to the director angle \( \phi \), we obtain the stationary equation for the LC director orientation in the presence of the light field

\[
\mathbf{n}(r) = e_x \sin \phi(z) + e_z \cos \phi(z) ,
\]

(3)

where \( \phi \) is the angle between the director and the \( z \) axis (see Fig. 1). \( e_x \) and \( e_z \) are the unit vectors in the Cartesian coordinate frame.

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We solve the system of coupled nonlinear equations \[41\] and \[55\] in a self-consistent manner together with the proper boundary conditions. For the director, we assume the strong anchoring at the cell boundaries, i.e.

\[
\phi(0) = \phi(L) = 0 ,
\]  

(6)

whereas for the electric field we consider the standard scattering conditions

\[
E_x(0) = E_{in} + E_{ref} , \quad E_x(L) = E_{out} .
\]  

(7)

Here \( E_{in} \), \( E_{ref} \), and \( E_{out} \) are the amplitudes of the incident, reflected, and outgoing waves, respectively. In all
The boundary conditions imply that we consider two counter-propagating waves on the left side of the LC cell, incoming and reflecting, whereas only an outgoing wave appears on the right side. Therefore, in order to solve this nonlinear problem, first we fix the amplitude of the outgoing wave \( E_{\text{out}} \). It allows us to find the unique values of the incident \( E_{\text{in}} \) and reflected \( E_{\text{ref}} \) waves.

Equation for the director is similar to a general-type equation for a nonlinear pendulum with the fixed boundary conditions. This means that we should look for its periodic solutions with the period 2\( L \). In fact, there exist many periodic solutions of Eq. 4. First of all, a trivial solution \( \phi(z) = 0 \) corresponds to the undisturbed orientation distribution of the director and the absolute minimum of the free energy. The Freedericksz transition occurs when this trivial solution becomes unstable for larger values of the input light intensity, and a finite energy barrier can appear between the minima of the free energy. The Freedericksz transition is a second order transition.

IV. RESULTS AND DISCUSSIONS

We solve the nonlinear transmission problem for two kinds of nematic liquid crystals, para-azoxyanisole (PAA) and Np-methoxybenzylidene-np-butylaniline (MBBA), which possess different signs of the parameter \( B = (1 - 9e_{\parallel}^2)/(4e_L) - (K_{33} - K_{11})/K_{33} \). We also take into account that a finite energy barrier can appear between the minima of the free energy; thus, the transition to the state \( \phi(z) = 0 \) takes place only when this energy barrier disappears. This leads to a hysteresis-like dependence of the director and the different threshold values for the "up" and "down" transitions in the director orientation.

FIG. 2: (colour online) (a,b) Spatial distributions of the field amplitude \( |E_x| \) in the cell of MBBA, before (dashed) and after (solid) the light-induced Freedericksz transition, \( L = \lambda/n_0 \), \( \lambda = 6328 \text{ nm}, n_0 = 1.544 \). (b) Spatial distributions of the director deviation angle \( \phi(z) \) in the cell of MBBA after the light-induced Freedericksz transition, for \( L = \lambda/n_0 \) (solid), \( L = 100 \mu\text{m} \) (dashed), are shown together with the function \( \phi_0 \sin(\pi z/L) \) at \( \phi_0 = 1.483 \) (dash-dotted).

According to their approach, the sign of this parameter \( B \) determines the order of the Freedericksz transition. For PAA \( B < 0 \) and the Ffreedericksz transition should be of the first order, while for MBBA \( B > 0 \) and there should be the second order transition.

We take the following physical parameters: (a) for PAA, \( K_{11} = 9.26 \times 10^{-7} \text{ dyn/cm}^2, K_{33} = 18 \times 10^{-7} \text{ dyn/cm}^2, n_0 = 1.595, n_e = 1.995 \), at \( \lambda = 4800 \text{ nm}, n_0 = 1.544, n_e = 1.758 \), at \( \lambda = 6328 \text{ nm} \); and consider two values for the cell thickness, \( L = \lambda/n_0 \) and \( L = 100 \mu\text{m} \). Spatial distributions of the electric field amplitude \( |E_x(z)| \) in the LC cell before and after the light-induced Freedericksz transition occurs is presented in Fig. 2(a) for the parameters of MBBA and the cell thickness \( L = \lambda/n_0 \). For the other value of the LC cell thickness the geometrical optics approximation is valid.
(\(L = 100\mu m\)), the spatial distribution of the electric field is similar, but the number of the oscillations of the electric field \(|E_x|\) inside the LC cell increases due to a larger value of \(L/\lambda\). For PAA, a very similar distribution of the electric field is found. Thus, we reveal an essentially inhomogeneous spatial distribution of the electric field inside the LC cell, and the functions \(|E_x(z)|\) are different before and after the Freedericksz transition.

Spatial distribution of the director orientation angle \(\phi(z)\) inside the LC cell after the Freedericksz transition is shown in Fig. 3b) for the parameters of MBBA, for \(L = \lambda/n_0\) and \(L = 100\mu m\), respectively. On the same plot, we show the function \(\phi_0 \sin(\pi z/L)\) at \(\phi_0 = 1.483\) for comparison. We notice that the position of the maximum of the director deviation angle can shift from the point \(z = L/2\), as a consequence of an asymmetric distribution of the field \(|E_x(z)|\) inside the LC cell. Spatial distribution of the director angle \(\phi(z)\) in the PAA cell has the same character as that shown in Fig. 3b) for MBBA.

In Fig. 3 we present our numerical results for a change of the maximum deformation angle \(\phi_{\text{max}}\) of the director as a function of the power density inside LC \(S_z\) for increasing and decreasing light intensity, for both PAA and MBBA and two values of the cell thickness, \(L = \lambda/n_0\) and \(L = 100\mu m\). For both kinds of LC, we observe a hysteresis-like dependence of the angle \(\phi_{\text{max}}\) and two different thresholds of the light-induced director reorientation: \(S'_z\) for the increasing intensity, and \(S''_z\), for the decreasing intensity. In both cases, these two thresholds correspond to the first-order transition. The results are similar for two values of the LC cell thickness, see Figs. 3(a-d). Thus, our results suggest that at the light-induced Freedericksz transition the cells of both kinds of LCs, PAA and MBBA, reveal hysteresis-like behavior with the respect to \(S_z\).

Dependencies of the amplitude of the outgoing wave \(|E_{\text{out}}|\) on the amplitude of the incident wave \(|E_{\text{in}}|\) are shown in Figs. 4(a-d) for the parameters of both PAA and MBBA. Depending on the LC cell thickness \(L\), the cell transmission is characterized by either hysteresis or multistability with respect to the incident wave amplitude. In the case of small thickness of the LC cell
FIG. 4: (colour online) Multistable transmission of the LC cell, shown as the outgoing wave $|E_{\text{out}}|$ vs. the incident wave $|E_{\text{in}}|$ for PAA: (a) $L = \lambda/n_0$ and (b) $L = 100\mu m$, and for MBBA: (c) $L = \lambda/n_0$ and (d) $L = 100\mu m$, respectively.

$(L = \lambda/n_0)$ only the hysteresis-like transmission is observed; it is caused by the hysteresis behavior of the director reorientation between "up" and "down" thresholds, as presented in Figs. 4(a,c). However, for larger thickness $(L = 100\mu m)$ we observe the transmission multistability, above the "up" threshold for increasing light intensity, and above the "down" threshold for decreasing light intensity [see Figs. 4(b,d)]. Multistability in our system is similar to that of a nonlinear resonator, and is determined by the resonator properties of a finite thickness of the LC cell.

The thresholds of the director reorientation for increasing and decreasing light intensities are shown in Figs. 4(a,b), for PAA and MBBA, respectively, as functions of the normalized thickness of the LC cell. Similar to the results of the geometrical optics approximation [8, 10], the threshold values are proportional to $(1/L)^2$, but they increase approximately in two times due to an essentially inhomogeneous spatial distribution of the electric field inside the LC cell. A similar increase of the threshold value for an inhomogeneous distribution of the electric field in the LC cell was also mentioned by Lednei et al. [13]. In addition, for the "up" threshold we observe an additional periodic dependence of the threshold value on the cell thickness $L$, which is typical for resonators and is caused by an interference of two counter-propagating waves in the LC cell. This result agrees with the results obtained for LC in a Fabry-Perot resonator [11]. The "up" threshold is determined by a competition between the electric field forces and elastic forces of the liquid crystal, and thus the interference distribution of the electric field in the LC cell is important. However, the "down" threshold is defined by the condition of the disappearance of a barrier between the local and absolute minima of the LC free energy [7]. We suppose that difference of these mechanisms leads to the different type of $L$-dependencies for the "up" and "down" thresholds.

We should mention that our results differ qualitatively from the results of earlier studies [2, 10], where for MBBA both hysteresis and bistability were not predicted. In the simplest case of one traveling wave [7, 11], the conserva-
1.2 1.4 1.6 1.8 2
L, (λ/\(n_0\)) units

0 50 100 150 200 250 300
\(S_z\) (MW/cm\(^2\))

"Up" threshold
"Down" threshold

FIG. 5: (colour online) Thresholds of the director reorientation for increasing (solid) and decreasing (dotted) light intensities vs. the cell thickness \(L\): (a) PAA, (b) MBBA.

tion of the value of \(S_z\) during the Freedericksz transition leads to the conservation of the electromagnetic field amplitudes at the boundaries of the LC cell. However, in the general case there always exists a reflected wave, so that we have \(S_z = (c/8\pi)E_x(0)H_y(0) = (c/8\pi)(E_{in} + E_{ref})(E_{in} - E_{ref}) = (c/8\pi)(E_{in}^2 - E_{ref}^2) = S_{in} - S_{ref}\), where \(S_{in}, S_{ref}\) are the power densities of the incident and reflected waves, respectively. In such a case, the conserva-

V. CONCLUSIONS

We have analyzed the light transmission through homeotropically-oriented cell of a nematic liquid crystal, and studied multistability and light-induced Freedericksz transition. We have solved numerically the coupled stationary equations for the nematic director and electric field of the propagating electromagnetic wave, for two kinds of liquid crystals (PAA and MBBA). We have found that the liquid crystals of both kinds possess multistability and hysteresis behavior in the transmission characterized by two thresholds of the director reorientation, so that for the increasing and decreasing light intensities the Freedericksz transition is of the first order.

We have demonstrated that the resonator effects of the liquid-crystal cell associated with the light reflection from two boundaries are significant, and they are responsible, in particular, for the observed periodic dependence of the threshold values and multistability of the transmitted light as a function of the cell thickness. We expect that these features will become important for the study of periodic photonic structures with holes filled in liquid crystals [17] where multiple reflection effects and nonlinear light-induced Freedericksz transition should be taken into account for developing tunable all-optical switching devices based on the structure with liquid crystals.

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