

Director grating and two-beam energy exchange in a hybrid photorefractive cholesteric cell with a helicoidal polymer network

V. Yu. Reshetnyak¹, I. P. Pinkevych^{1,1}, M. E. McConney², and D. R. Evans²

¹*Physics Faculty, Taras Shevchenko National University of Kyiv, Volodymyrs'ka Street 64, Kyiv 01601, Ukraine*

²*Air Force Research Laboratory, Materials and Manufacturing Directorate, Wright-Patterson Air Force Base, Ohio 45433, USA*

We develop a theory describing two-beam energy exchange in a cholesteric liquid crystal (CLC) stabilized in the planar state by a helicoidal polymer network. The CLC layer is placed between photorefractive and non-photorefractive substrates and illuminated by two intersecting coherent light beams. An interference pattern created by the incident beams induces a spatially periodic space-charge electric field in the photorefractive substrate. The field penetrates into the adjacent CLC layer and interacts with the charges trapped on the polymer fibrils forcing the fibrils to move along the helicoidal axis. At new positions, the fibrils reorient the CLC director and therefore induce a director grating. The light beams propagating across the cell couple within the grating. We calculate the energy exchange between the coupled beams and the gain of the weak light beam. We analyze the dependence of the gain coefficient on the parameters of the polymer network and the CLC, and show that it can reach values greater than those obtained in typical solid photorefractive crystals.

1. Introduction

Due to a high refractive index modulation of order 0.1-0.2 caused by the liquid crystal (LC) director reorientation, a very strong energy transfer between coupled light beams is observed in LC based systems.¹⁻³ The gain coefficient in such systems is more than two orders of magnitude larger than that in typical solid photorefractive crystals.^{4,5} A whole set of analogous photorefractive liquid crystalline systems is treated in the literature.⁶⁻¹¹ In recent years the hybrid organic-inorganic photorefractives, in which an LC sample is placed adjacent to a photorefractive or photoconducting inorganic layers have been extensively studied.⁹⁻¹¹ The photo-generated charges contained in the inorganic layers create a space-charge field, which penetrates into the LC layer and modulates the LC director creating a director grating and in turn a refractive index grating. As shown in Refs. 12-14, the Bragg regime can be realized in these systems, where the coupled light beams generate only first order diffracted beams.

It has been proposed that the formation of a director grating in hybrid organic-inorganic photorefractives is governed by the interaction of the space-charge field with the LC flexoelectric polarization¹⁵, rather than by static dielectric anisotropy coupling.^{16,17} It allowed for a description of the experimental results obtained for both nematic¹⁵ and cholesteric LC cells.^{18,19}

The gain of the two-beam energy exchange in hybrid organic-inorganic photorefractive cells depends strongly on the director boundary conditions at the LC cell substrates, i.e., director pre-tilt angle and anchoring energy. In the cells with a flexoelectric mechanism of the director reorientation, the gain becomes negligibly small when there is no director pre-tilt at the LC cell

¹ ipinkevych@gmail.com

substrates. This takes place in the planar state of the cholesteric liquid crystal (CLC) with the CLC helical axis perpendicular to the cell substrates. However, in recent times, the stable CLC planar states are obtained using polymer networks. Such polymer stabilization provides additional functionalities to the CLC.²⁰⁻²⁵ In particular, as was shown in Ref. 26, the polymer network can exhibit translational motion in response to an external electric field, and due to the coupling of the network with the LC this polymer network motion changes the CLC helical pitch.

In the present paper, we speculate that the formation of the space-charge field in the photorefractive substrate of the CLC cell may control translational motion of the polymer network, and therefore affect the director grating formation. We study two-beam coupling enabled by the director grating formed in the planar state of the CLC due to the polymer network.

This paper is organized as follows. In Sec. 2 we introduce a model of the hybrid cell with the CLC layer stabilized in the planar state by the polymer network, and derive and solve equations for the CLC director profile under a space-charge electric field. In Sec. 3 we consider propagation and coupling of two light beams in the CLC cell with a director grating, derive expressions for the exponential gain coefficient, and analyze the influence of parameters characterizing the polymer network and CLC on the gain coefficient. In Sec. 4 we present brief conclusions.

2. Director grating induced by the polymer network

Consider a CLC layer placed between photorefractive (inorganic crystal) and non-photorefractive substrates, where the CLC is stabilized in the planar state by a helicoidal polymer network with the helix axis perpendicular to the CLC layer substrates. The z -axis is directed along the helix axis and the CLC layer is bound by the plane $z=-L/2$ from the side of photorefractive substrate and by the plane at $z=L/2$ from the side of the non-photorefractive substrate (Fig. 1). Due to the anchoring, the CLC director and the polymer fibrils are initially aligned in the same direction.

Two intersecting coherent light beams $\mathbf{E}_1 = A_1 \mathbf{e}_1 \exp(i\mathbf{k}_1 \mathbf{r} - i\omega t)$ and $\mathbf{E}_2 = A_2 \mathbf{e}_2 \exp(i\mathbf{k}_2 \mathbf{r} - i\omega t)$ illuminate the hybrid cell. The bisector of the beams is directed along the z -axis, and the wave vectors $\mathbf{k}_1, \mathbf{k}_2$ lay in the xz -plane. The polarization vectors of the beams, $\mathbf{e}_1, \mathbf{e}_2$, lay in the xz -plane at the entrance plane of the CLC cell, $z=-L/2$, but they can rotate as the beams propagate across the cholesteric cell. The beams produce a light intensity interference pattern in the photorefractive substrate,

$$I(z) = (I_1 + I_2) \left[1 + \frac{1}{2} (m \exp(iqx) + c.c.) \right], \quad (1)$$

where the modulation parameter $m = 2 \cos(2\delta) A_1 A_2^* / (I_1 + I_2)$, 2δ is the angle between incident beams in the photorefractive substrate, $I_1 = A_1 A_1^*$, $I_2 = A_2 A_2^*$ are the intensities of the incident beams, and $q = k_{1x} - k_{2x}$ is the wave number of the intensity pattern.

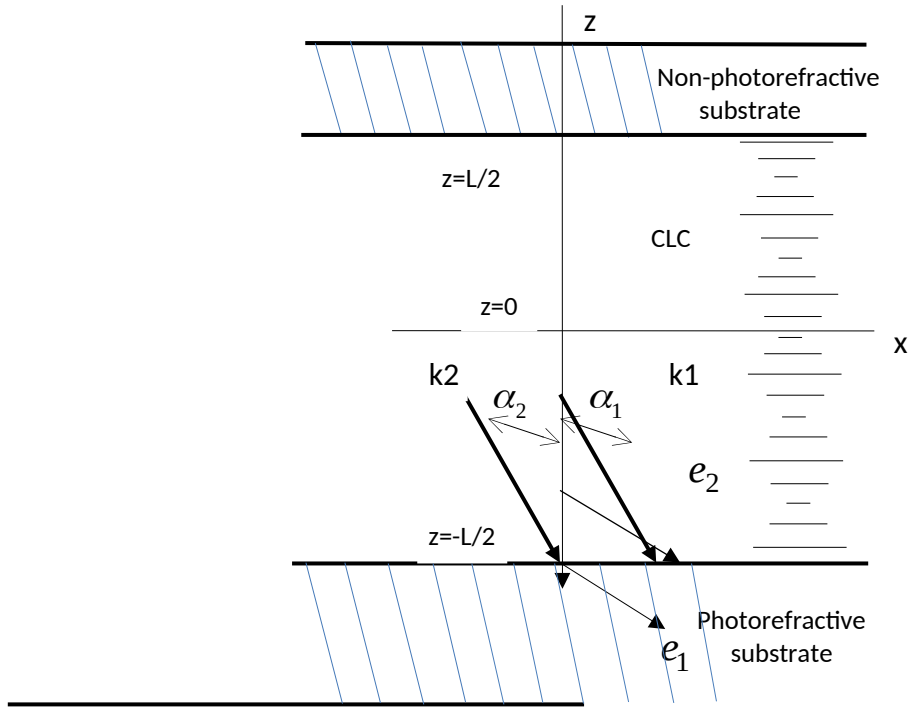


FIG.1. Schematic of the CLC cell, showing light beams incident from a photorefractive medium, together with associated wave- and polarization vectors. The quantities $\mathbf{k}^{(1,2)}$, $\alpha_{1,2}$, $\mathbf{e}_{1,2}$ are defined in the text. The horizontal lines to spanning the cell depict the CLC helical twist.

The light intensity pattern given by eq. (1) induces a space charge inside the photorefractive substrate. The space-charge density is modulated along the x -axis with a period equal to $\Lambda = 2\pi/q$ and gives rise to the space-charge field with a magnitude $E_{sc}(q)$, which penetrates into the adjacent CLC layer. In particular, in an infinite photorefractive medium and for a diffusion-dominated space-charge field $E_{sc}(q)$ is as follows:²⁷

$$E_{sc}(q) = \frac{iE_d}{1 + \frac{E_d}{E_q}}, \quad E_d = q \frac{k_b T}{e}, \quad E_q = \left(1 - \frac{N_a}{N_d}\right) \frac{eN_a}{\epsilon_0 \epsilon_{ph} q}, \quad (2)$$

where E_d is the diffusion field, E_q is the saturation field, N_a and N_d are respectively the acceptor and donor impurity densities, ϵ_{ph} is the dielectric permittivity of the photorefractive material, e is the electron charge, and k_b is the Boltzmann constant.

However, the photorefractive medium is rather semi-infinite and, in general, the solution for the electric fields in the photorefractive substrate and the LC is actually a complex coupled problem. Nevertheless, if we suppose that expression (2) remains valid at the CLC-photorefractive medium boundary, we can use eq. (2) as the boundary condition for the electric

field in the CLC cell. Then, supposing that at the non-photorefractive substrate the electric potential can be chosen to be a zero and solving the Poisson equation, we obtain the following expressions for the electric field in the CLC bulk:¹⁹

$$\mathbf{E} = iE_x + jE_z, \quad E_x = E_{0x} \exp(iqx) + c.c., \quad E_z = E_{0z} \exp(iqx) + c.c., \quad (3)$$

where

$$E_{0x} = -\frac{E_{sc}(q)m(-L/2)}{2\sinh(\tilde{q}L)} \sinh \tilde{q}(z - L/2), \quad E_{0z} = \frac{i\tilde{q}E_{sc}(q)m(-L/2)}{2q\sinh(\tilde{q}L)} \cosh \tilde{q}(z - L/2). \quad (4)$$

Here $\tilde{q} = q\sqrt{(\tilde{\epsilon}_\perp + \tilde{\epsilon}_\parallel)/2\tilde{\epsilon}_\perp}$, $m(-L/2)$ is the modulation parameter at the entrance plane $z = -L/2$, $\tilde{\epsilon}_\parallel$ and $\tilde{\epsilon}_\perp$ are the CLC static dielectric constants parallel and perpendicular to the director.

We confine ourselves to just studying the influence of the polymer network under the space charge field on the CLC director spatial profile. The polymer network is comprised of uniformly distributed fibrils aligned during the polymerization in the same direction as the CLC director. As in Refs. 26 and 28, we suppose that the ions (positive or negative) are trapped on the polymer fibrils, which therefore can exhibit translational motion in response to the electric field. For the CLC in the planar state, we can introduce the director in the form $\mathbf{n} = (\cos\phi, \sin\phi, 0)$. In the absence of the space-charge field, the CLC director matches the orientation of the polymer fibrils, which is described by the angle $\varphi_0(z) = (2\pi/p_0) \times (z + L/2)$ where p_0 is the initial cholesteric pitch (i.e. before the fibril translation influences the CLC pitch). Under the component $E_x(x, z)$ of the photorefractive electric field (3), the charged fibrils move along the z -axis to new positions; the translation of the polymer fibrils along the z -axis under this field is denoted by $u(x, z)$. The polymer fibril initially located at $z - u(x, z)$ with the twist angle $\varphi_0[z - u(x, z)]$ is moved to the position z where the twist angle of the CLC director is $\phi(x, z)$. Because of this translation, there appears to be a difference in the twist angles of the CLC director and the polymer fibril, such that $\phi(x, z) - \varphi_0[z - u(x, z)] = \phi(x, z) - (2\pi/p_0) \times [z + L/2 - u(x, z)]$. Due to the anchoring of the director with the polymer fibrils, this difference influences the director spatial distribution. Thus, following Ref. 26 we can write the contribution of the polymer network into a free energy functional of the CLC cell in the form $F_{PN} = \int f_{PN} dV$, where

$$f_{PN} = \frac{1}{2} B \left[\frac{du(x, z)}{dz} \right]^2 + \frac{1}{2} A \left[\phi(x, z) - \frac{2\pi}{p_0} [(z + L/2) - u(x, z)] \right]^2 - \frac{1}{2} CE_z u(x, z). \quad (5)$$

In eq. (5) the first term is the volume density of the polymer network elastic energy, the second term is the surface energy of the CLC on the fibrils surface per unit volume, and the third term

describes the interaction of the photorefractive electric field with the fibrils due to the ions trapped on the fibrils.

In the initial state at zero space-charge field the homogeneous CLC twist does not produce any flexopolarization; therefore, the flexoelectric effect contribution to the CLC free energy, which may arise due to the CLC helix deformation will be of a second order in the space-charge field. Neglecting also the direct action of the photorefractive and light fields on the CLC director, we must minimize the total free energy functional $F = \int (f_{LC} + f_{PN}) dV$ where f_{LC} is the volume density of the CLC elastic energy, which in the one-constant approximation is as follows,

$$f_{el} = \frac{K}{2} \left[\left(\frac{\partial \phi}{\partial z} - \frac{2\pi}{p_0} \right)^2 + \left(\frac{\partial \phi}{\partial x} \right)^2 \right]. \quad (6)$$

Then, minimizing the functional F we obtain the Euler-Lagrange equations for the fibril translation $u(x, z)$ and the director angle $\phi(x, z)$:

$$B \frac{d^2 u(x, z)}{dz^2} - A \frac{2\pi}{p_0} \left[\phi(x, z) - \frac{2\pi}{p_0} [(z + L/2) - u(x, z)] \right] + \frac{1}{2} C E_z = 0, \quad (7)$$

$$K \left[\frac{\partial^2 \phi(x, z)}{\partial z^2} + \frac{\partial^2 \phi(x, z)}{\partial x^2} \right] - A \left[\phi(x, z) - \frac{2\pi}{p_0} [(z + L/2) - u(x, z)] \right] = 0. \quad (8)$$

Expressions for the polymer fibrils translation $u(x, z)$ and the director angle $\phi(x, z)$ can be decomposed as follows,

$$u(x, z) = u(z) \exp(iqx) + c.c., \quad (9)$$

$$\phi(x, z) = \varphi_0(z) + [\varphi(z) \exp(iqx) + c.c.] \quad (10)$$

Substituting eqs. (9) and (10) in eqs. (7) and (8) we obtain equations for the magnitudes $u(z)$ and $\varphi(z)$:

$$\frac{d^2 u(z)}{dz^2} - \frac{A}{B} \left(\frac{2\pi}{p_0} \right)^2 u(z) - \frac{A}{B} \frac{2\pi}{p_0} \varphi(z) = - \frac{1}{2} \frac{C}{B} E_{0z}, \quad (11)$$

$$\frac{\partial^2 \varphi(z)}{\partial z^2} - \left(q^2 + \frac{A}{K} \right) \varphi(z) - \frac{A}{K} \frac{2\pi}{p_0} u(z) = 0 \quad (12)$$

In eq. (11) we can neglect contribution to the fibril translation magnitude $u(z)$ from the small angle φ in comparison with contribution from the electric field and arrive at

$$\frac{d^2 u(z)}{dz^2} - \frac{A \left(\frac{2\tau}{p_0} \right)^2}{B} u(z) = - \frac{1}{2} \frac{C}{B} E_{0z} \quad (13)$$

We accept “soft” boundary conditions at the photorefractive substrate $z = -L/2$ assuming the fibril anchoring and the director azimuth anchoring with the substrate to be weak and setting the fibril translation magnitude at the substrate to be $u(z = -L/2) = u_0$. The fibril translation causes the director rotation at the photorefractive substrate from the initial angle $\varphi_0(z = -L/2) = 0$, and for the director angle φ we accept the free boundary condition: $\partial \varphi(z) / \partial z|_{z=-L/2} = 0$. As the photorefractive field is localized near the photorefractive substrate, such boundary conditions allow the photorefractive field to maximize its effect on the charged polymer fibrils, the CLC director, and therefore on the CLC pitch. The experimental realization of these conditions may be based on a modification of a technique suggested in Ref. 29, where a polymer network is fabricated in the CLC mixture. Due to the photo-initiator embedded in one of the two alignment layers coating the cell substrates, a polymer network is fabricated in the way that it is localized close to one surface, but not touching this surface. We also refer to Refs. 30 and 31, where it is shown that the LC director can easily slide over the substrate. At the non-photorefractive substrate $z = L/2$ we suppose “strong” fibril and director anchoring: $u(z = L/2) = 0$, $\varphi(z = L/2) = 0$.

It is plausible to assume the fibril translation magnitude at the photorefractive substrate u_0 to be proportional to the magnitude of the photorefractive field at this substrate, $E_{0z}(z = -L/2)$:

$$u_0 = D E_{0z}(-L/2), \quad (14)$$

where D is a constant, which, in fact, depends on the charge localized on the polymer fibril and elasticity of the polymer network in CLC.

Now taking into account boundary conditions for the fibril translation magnitude u and eq. (4) for $E_{0z}(z = -L/2)$, the solution to eq. (13) takes the form

$$u(z) = \frac{\tilde{q} |E_{sc}(q)| m(-L/2)}{2q \sinh(\tilde{q}L) \sinh(sL)} \left[c_1 \sinh s(z + L/2) + c_2 \sinh s(z - L/2) + c_3 \cosh \tilde{q}(z - L/2) \right], \quad (15)$$

where

$$c_1 = - \frac{C}{2B} \frac{1}{(\tilde{q}^2 - s^2)}, \quad c_2 = \frac{C \cosh(\tilde{q}L)}{2B (\tilde{q}^2 - s^2)} + D \cosh(\tilde{q}L), \quad c_3 = \frac{C \sinh(sL)}{2B (\tilde{q}^2 - s^2)}, \quad s = \frac{2\tau}{p_0} \sqrt{A/B}. \quad (16)$$

As it is seen from eqs. (14) - (16) and (5), contribution to the fibrils translation magnitude arises from the interaction of the photorefractive field with the fibrils in the cell bulk (terms with coefficient C/B) and at the boundary of the photorefractive substrate (terms with coefficient D).

Substituting eq. (15) in eq. (12) and using boundary conditions for the director angle $\varphi(z)$, we obtain a solution to this angle as follows,

$$\varphi(z) = \frac{|E_{sc}(q)|m(-L/2)}{2q} f(z), \quad (17)$$

where

$$f(z) = \frac{(2\pi/p_0)\tilde{q}}{\sinh(\tilde{q}L)\sinh(sL)} \frac{A}{K} [d_1 \cosh h(z+L/2) + d_2 \sinh h(z-L/2)] \\ + \frac{c_1}{s^2-h^2} \sinh s(z+L/2) + \frac{c_2}{s^2-h^2} \sinh s(z-L/2) + \frac{c_3}{\tilde{q}^2-h^2} \cosh \tilde{q}(z-L/2), \quad (18)$$

and

$$d_1 = -\frac{1}{\cosh(hL)} \left[\frac{c_1 \sinh(sL)}{s^2-h^2} + \frac{c_3}{\tilde{q}^2-h^2} \right], \quad h = \sqrt{q^2 + A/K}, \\ d_2 = \frac{1}{\cosh(hL)} \left[+\frac{\tilde{q} c_3 \sinh(\tilde{q}L)}{h \tilde{q}^2-h^2} - \frac{s c_1 + c_2 \cosh(sL)}{h s^2-h^2} \right]. \quad (19)$$

Eqs. (10) and (17) - (19) describe the director grating induced in the polymer stabilized CLC by the charged polymer fibrils translation under the photorefractive field.

For numerical calculations of the fibril translation and the director grating magnitudes we use the polymer network parameters $A = 1.6 \times 10^2 \text{ Jm}^{-3}$, $B = 10^3 \text{ Jm}^{-3}$, $C = 45 \text{ Cm}^{-3}$ and the initial CLC pitch $p_0 = 0.371 \text{ }\mu\text{m}$ obtained in Ref. 26 for the cell with thickness $L = 10 \text{ }\mu\text{m}$. As for the CLC parameters we use the following typical numbers $\tilde{\epsilon}_{\parallel} = 9.1$ and $\tilde{\epsilon}_{\perp} = 4.1$ ¹² and set the elastic constant to be $K = 8 \times 10^{-12} \text{ N}$. In order to evaluate a magnitude of the space-charge field $E_{sc}(q)$, we follow formula (2) and Ref. 12, where the photorefractive substrate SBN:Ce was used; the dielectric permittivity of the photorefractive layer is given by $\epsilon_{ph} = 200$ at temperature $T = 300 \text{ K}$. The value of parameter $m(-L/2)$ is taken to be equal to 0.2 corresponding to the incident wave amplitudes ratio $A_1(-L/2)/A_2(-L/2) = 0.1$, which is typical for the experiments on the two-beam energy exchange.

Below in Figs. 2 – 4 we present results of calculations of the fibrils translation magnitude and the director grating magnitude for different values of the polymer network parameters. It allows us to demonstrate an impact on these magnitudes of the fibrils anchoring strength with the CLC director (A), the polymer network elasticity (B), the photorefractive field interaction with charged fibrils in the cell bulk (C) and at the photorefractive substrate (D), as well as the CLC elastic constant K .

In Fig. 2a, we show a ratio of the fibrils translation magnitude at the photorefractive substrate to the initial cholesteric pitch, u_0/p_0 , as a function of the grating spacing Λ for different values of the parameter D . According to eq. (14), this ratio increases with an increase of the parameter D and the Λ -dependence of this ratio reproduces the corresponding behavior of the z -component of the photorefractive field E_{0z} . The value of the parameter D is unknown, therefore using Fig. 2a we can choose its value to be such that the maximum of the ratio u_0/p_0 does not exceed 0.1. Therefore, for our further calculations, we use values D not exceeding $6 \times 10^{-12} \text{V}^{-1} \text{m}^2$.

In Fig. 2b, we present the ratio $u(z)/p_0$ as a function of position in the cell for different values of parameter D for a director grating spacing $\Lambda = 2 \mu\text{m}$. It is seen that the polymer fibrils translation in the cell bulk, $u(z)$, increases with an increase of parameter D , but the most significant effect is only near the photorefractive substrate. The plots of the ratio $u(z)/p_0$ as a function of the position in the cell for different values of parameters A and B are shown in Figs. 3a and 3b, respectively.

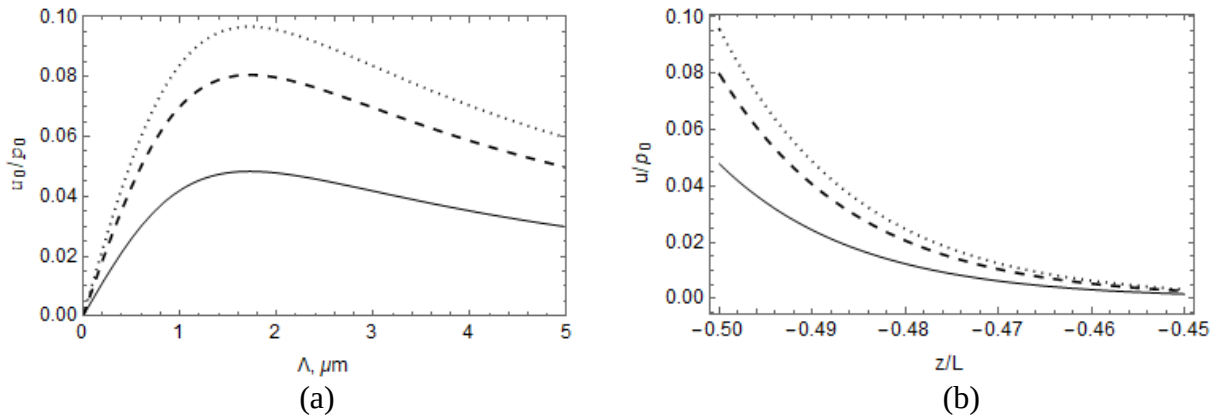


FIG. 2. (a) Ratio of the fibrils translation magnitude at the photorefractive substrate to the initial cholesteric pitch versus the director grating spacing. (b) Ratio of the fibrils translation magnitude to the initial cholesteric pitch as a function of position in the cell, $\Lambda = 2 \mu\text{m}$. D (in $10^{-12} \text{V}^{-1} \text{m}^2$) = 3 - solid line, 5 - dashed, 6 - dotted.

The magnitude of the fibrils translation decreases with an increase of parameter A , which is caused by retarding of the fibrils motion due to their anchoring with the LC director. At the same time, magnitude $u(z)$ increases with an increase of parameter B connected with the term describing the polymer network elasticity. For the fixed fibril translation at the photorefractive substrate, u_0 , an increase of the network elasticity leads to a more slowly decreasing displacement of the fibrils in the cell bulk. However, a contribution to the magnitude $u(z)$ from terms describing direct interactions of the photorefractive field with charged fibrils in the cell bulk [terms in eq. (16) with coefficient C] is negligibly small due to the rapid decay of the photorefractive field with distance from the substrate. Therefore, impact of the network

parameters A and B on $u(z)$ is provided mainly in the form of ratio A/B that is seen at the comparison of Figs. 3a and 3b.

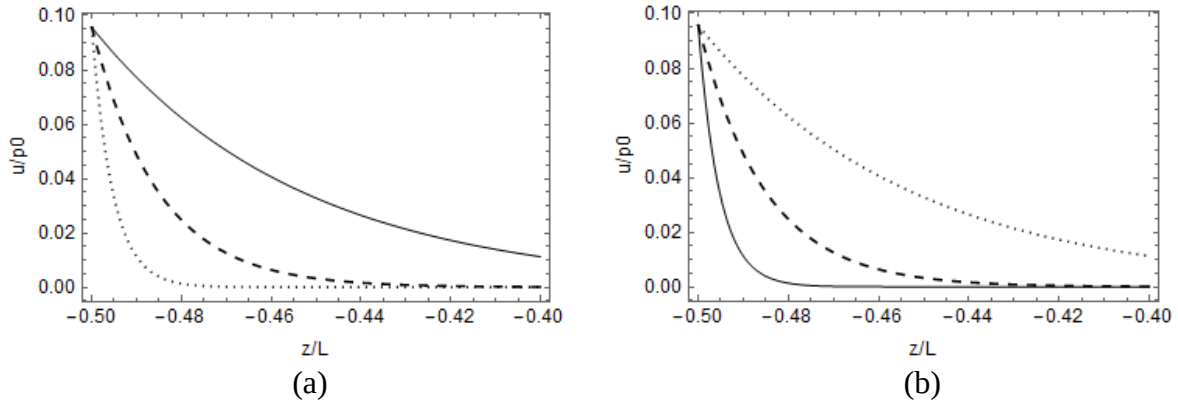
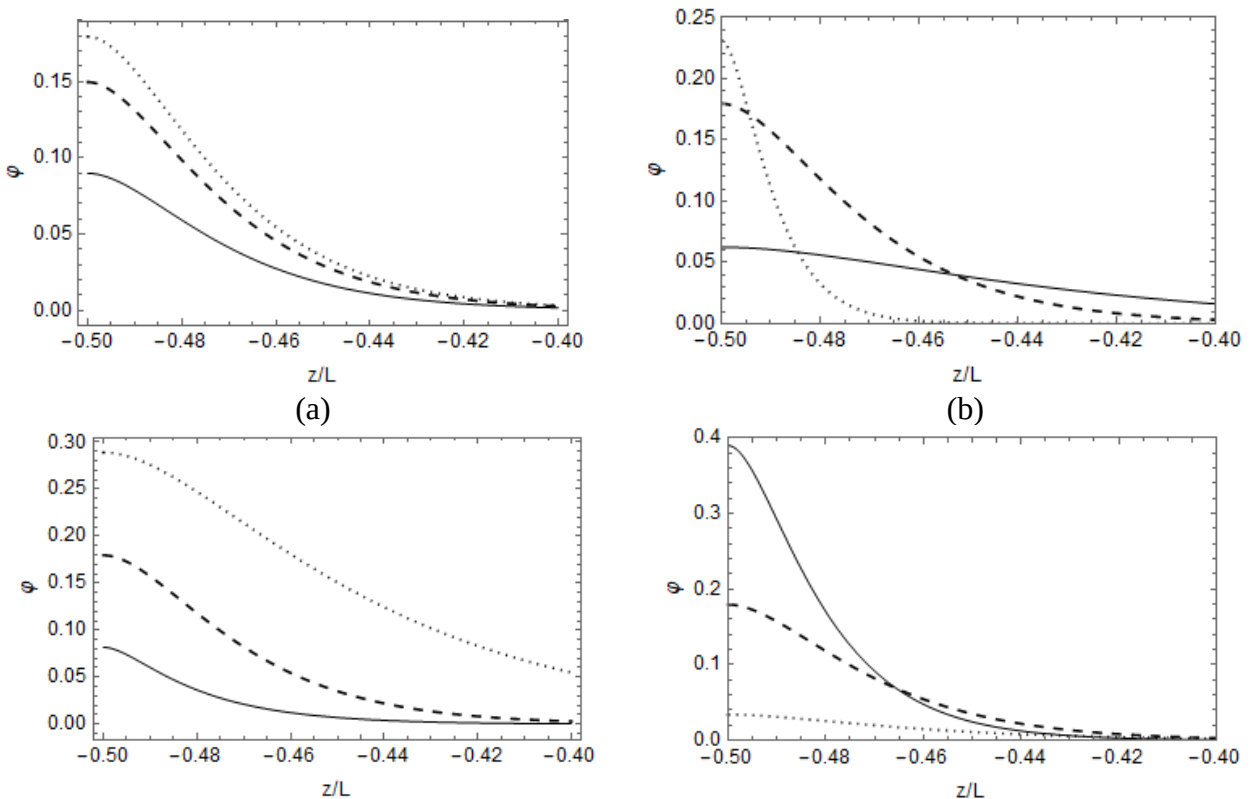


FIG. 3. Ratio of the fibrils translation magnitude to the initial cholesteric pitch as a function of position in the cell for different values of parameters A and B . (a) A (in Jm^{-3}) = 1.6×10^1 - solid line, 1.6×10^2 - dashed, 1.6×10^3 - dotted, $B = 10^3 Jm^{-3}$; (b) B (in Jm^{-3}) = 10^2 - solid line, 10^3 - dashed, 10^4 - dotted, $A = 1.6 \times 10^2 Jm^{-3}$; $D = 6 \times 10^{-12} V^{-1} m^2$.

The plots of the director grating magnitude described by the angle $\varphi(z)$ as a function of position in the CLC cell for different values of the parameters D , A , B , and the CLC elastic constant K are shown in Fig. 4a, 4b, 4c, and 4d, respectively.



(c) (d)

FIG. 4. Director grating magnitude as a function of position in the CLC cell for different values of the parameters D , A , B and elastic constant K . (a) D (in $10^{-12}V^{-1}m^2$) = 3 - solid line, 5 - dashed, 6 - dotted; (b) A (in Jm^{-3}) = 1.6×10^1 - solid line, $1.6 \cdot 10^2$ - dashed, 1.6×10^3 - dotted, $B = 10^3 Jm^{-3}$, $D = 6 \times 10^{-12} V^{-1} m^2$; (c) B (in Jm^{-3}) = 10^2 - solid line, 10^3 - dashed, 10^4 - dotted, $A = 1.6 \times 10^2 Jm^{-3}$, $D = 6 \times 10^{-12} V^{-1} m^2$; (d) K (in N) = 8×10^{-13} - solid line, 8×10^{-12} - dashed, 8×10^{-11} - dotted.

One can see from Figs. 4a and 4c that as in the case of the fibrils translation magnitude $u(z)$ the director grating magnitude $\varphi(z)$ increases with an increase of the parameters D and B . Dependence $\varphi(z)$ on the parameter A , as we can see from eqs. (17)-(19) and Fig. 4b, is more complicated: the magnitude $\varphi(z)$ increases with an increase of A in points near the photorefractive substrate, but rapidly decreases as the distance increases from the substrate. We also can see from Fig. 4d that smaller elasticity, K , of the CLC promotes greater values of $\varphi(z)$.

As in the case of the fibrils translation magnitude the terms with coefficient C give negligibly small contributions to the magnitude $\varphi(z)$; therefore, at further study of the light beams coupling on the director grating we do not take into account these terms in eqs. (17)-(19).

3. Beams coupling and gain of the signal beam

The incident light beams propagate in the CLC cell, where the director grating is induced by the polymer fibrils translation. The electric field of the light beams in the CLC satisfies the wave equation

$$\nabla \times \nabla \times \mathbf{E}_{hv} - \frac{\omega^2}{c^2} \hat{\varepsilon} \mathbf{E}_{hv} = 0, \quad (20)$$

where $\hat{\varepsilon}$ is the CLC dielectric tensor in the optical frequency regime. In the planar CLC it has the form

$$\hat{\varepsilon} = \begin{vmatrix} \varepsilon_{\perp} + \varepsilon_a \cos^2 \phi & (\varepsilon_a / 2) \sin 2\phi & 0 \\ (\varepsilon_a / 2) \sin 2\phi & \varepsilon_{\perp} + \varepsilon_a \sin^2 \phi & 0 \\ 0 & 0 & \varepsilon_{\perp} \end{vmatrix}. \quad (21)$$

Here $\varepsilon_a = \varepsilon_{\parallel} - \varepsilon_{\perp}$, and ε_{\parallel} , ε_{\perp} are the principal values of the dielectric tensor at optical frequency along and perpendicular to the LC director.

In our case, the director angle ϕ is described by eq. (10). Substituting eq. (10) into eq. (21) and neglecting second order and higher terms in the small angle $\varphi(z)$, one can rewrite the dielectric tensor $\hat{\varepsilon}$ in the following way:

$$\hat{\varepsilon} = \hat{\varepsilon}_1(z) + [\hat{\varepsilon}_2(z) \exp(iqx) + c.c.] , \quad (22)$$

$$\hat{\varepsilon}_1 = \begin{vmatrix} \varepsilon_{\perp} + \varepsilon_a \cos^2 \varphi_0 & (\varepsilon_a / 2) \sin 2\varphi_0 & 0 \\ (\varepsilon_a / 2) \sin 2\varphi_0 & \varepsilon_{\perp} + \varepsilon_a \sin^2 \varphi_0 & 0 \\ 0 & 0 & \varepsilon_{\perp} \end{vmatrix}, \quad \hat{\varepsilon}_2 = \varepsilon_a \varphi(z) \begin{vmatrix} -\sin 2\varphi_0 & \cos 2\varphi_0 & 0 \\ \cos 2\varphi_0 & \sin 2\varphi_0 & 0 \\ 0 & 0 & 0 \end{vmatrix}, \quad (23)$$

where $\hat{\varepsilon}_1$ is a dielectric tensor of the CLC without the director grating.

The electric field vector \mathbf{E}_{in} in eq. (20) is a superposition of the electric vectors of the two light beams, $\mathbf{E}_{in} = \mathbf{E}_1 + \mathbf{E}_2$. We study a case when the wavelength of incident light beams λ is outside of the cholesteric gap and the waveguide regime takes place. In this regime the condition $\lambda > p(n_c - n_0)$ holds, where n_0 , n_c are respectively the ordinary and extraordinary wave refraction indices, and the eigenmodes of electromagnetic waves are nearly circular.³² Then in the CLC with dielectric tensor $\hat{\varepsilon}_1$, the Cartesian components of each light beam can be written in the form of the superposition of two waves with opposite circular polarization,¹⁹

$$\begin{aligned} E_{1x} &= (A_1 / 2) (e^{i(\omega/c)n_1^{(1)}(z+L/2)} + e^{i(\omega/c)n_1^{(2)}(z+L/2)}) e^{ik_1 x}, \\ E_{1y} &= i(A_1 / 2) (e^{i(\omega/c)n_1^{(1)}(z+L/2)} - e^{i(\omega/c)n_1^{(2)}(z+L/2)}) e^{ik_1 x}, \\ E_{1z} &= i\alpha_1 \frac{n_1^{(1)} + n_1^{(2)}}{2} \frac{c}{\varepsilon_{\perp} \omega} \frac{\partial E_{1x}}{\partial z}, \end{aligned} \quad (24)$$

and

$$\begin{aligned} E_{2x} &= (A_2 / 2) (e^{i(\omega/c)n_2^{(1)}(z+L/2)} + e^{i(\omega/c)n_2^{(2)}(z+L/2)}) e^{ik_2 x}, \\ E_{2y} &= i(A_2 / 2) (e^{i(\omega/c)n_2^{(1)}(z+L/2)} - e^{i(\omega/c)n_2^{(2)}(z+L/2)}) e^{ik_2 x}, \\ E_{2z} &= -i\alpha_2 \frac{n_2^{(1)} + n_2^{(2)}}{2} \frac{c}{\varepsilon_{\perp} \omega} \frac{\partial E_{2x}}{\partial z}, \end{aligned} \quad (25)$$

where $n_1^{(1)}, n_1^{(2)}$ are the refractive indices for waves with the left and right circular polarizations, respectively; $i = 1, 2$ denotes the beams, α_1, α_2 are the beams' incidence angles.

We set the wave vectors of the light beams to be symmetric with regard to the cell normal, so that $\alpha_1 = \alpha_2 = \alpha$. Since the angle α is small, the refractive indices for waves with the same circular polarization may be regarded as equal, $n_1^{(1)} = n_2^{(1)} = n^{(1)}$, $n_1^{(2)} = n_2^{(2)} = n^{(2)}$.

The term $\hat{\varepsilon}_2(z) \exp(iqx) + c.c.$ in eq. (22) is responsible for the coupling between the light beams in the CLC. We follow a procedure outlined in Ref. 33 and used in our previous papers.^{18,19}

It involves setting electric field magnitudes in eqs. (24), (25) $A_1 = A_1(z)$, $A_2 = A_2(z)$, and

assuming these magnitudes vary slowly across the cell. We define beam 1 to be the signal and beam 2 to be the pump, with the consequence that the pump magnitude $|A_2| \gg |A_1|$. Substituting the electric fields given by eqs. (24) and (25) into the wave equation (20) we adopt the undepleted pump approximation²⁷, for which the signal has a negligible effect on the pump magnitude. In this case, the pump magnitude $A_2(z)$ may be regarded as constant and therefore the set of coupled equations for the electric field magnitudes $A_1(z)$ and $A_2(z)$ reduces to a single equation for the signal beam magnitude $A_1(z)$,

$$\frac{\partial}{\partial z} A_1(z) = -iS(z) A_2, \quad (26)$$

where

$$S(z) = \frac{\omega}{c} \frac{\epsilon_a}{n^{(1)} + n^{(2)}} \varphi(z) \sin \left\{ \left[\frac{\omega}{c} (n^{(1)} - n^{(2)}) + \frac{4\pi}{p_0} \right] (z + L/2) \right\}. \quad (27)$$

The solution to eq. (26) has the form:

$$A_1(z) = A_1(-L/2) - iA_2 \int_{-L/2}^z S(z') dz' \quad (28)$$

The signal energy gain caused by the CLC layer is defined as

$$G = \left| \frac{A_1(L/2)}{A_1(-L/2)} \right|^2, \quad (29)$$

where

$$A_1(L/2) = A_1(-L/2) - iA_2 \int_{-L/2}^{L/2} S(z') dz' \quad (30)$$

Substituting eq. (30) into eq. (29) and using eqs. (27) and (17) for $\varphi(z)$, we arrive at the following expression for the signal gain:

$$G = \left| 1 + \frac{\omega}{c} \frac{\epsilon_a}{n^{(1)} + n^{(2)}} \frac{|E_{sc}(q)| \cos(2\delta)}{q} \int_{-L/2}^{L/2} dz f(z) \sin \left[\left[\frac{\omega}{c} (n^{(1)} - n^{(2)}) + \frac{4\pi}{p_0} \right] (z + \frac{L}{2}) \right] \right|^2. \quad (31)$$

Evaluating the integral in eq. (31), we can express the result in terms of the exponential gain

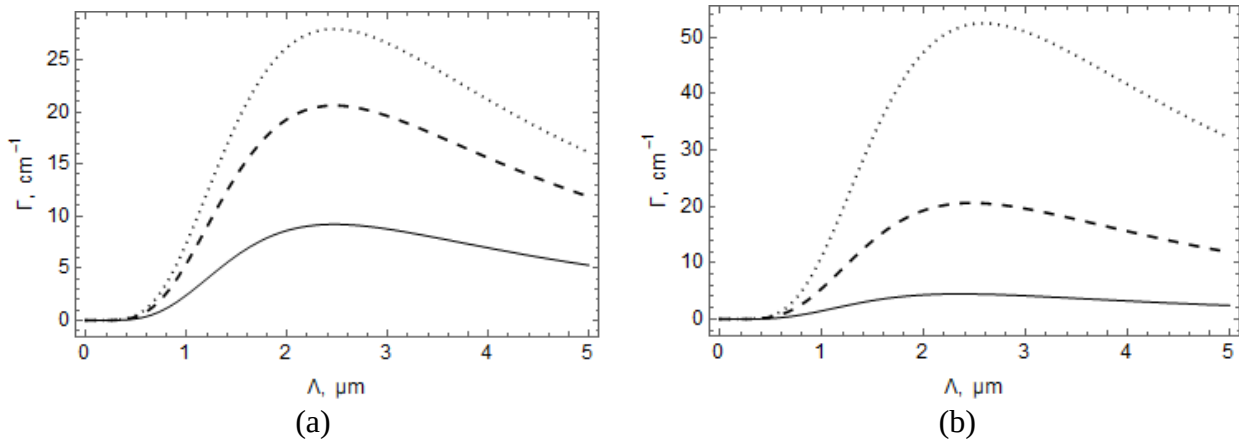
coefficient $\Gamma = \frac{1}{L} \ln |G|$:

$$\Gamma = \frac{1}{L} \ln \left[1 + \left(\frac{\omega}{c} \frac{\varepsilon_a}{n^{(1)} + n^{(2)}} \frac{|E_{sc}(q)| \cos(2\delta)}{q} \frac{A}{K} \frac{D}{s^2 - h^2} \frac{(2\pi/p_0) \tilde{q} \cosh(\tilde{q}L)}{\sinh(sL) \sinh(\tilde{q}L)} \right)^2 \right. \\ \left. \times \left(\frac{t \sinh(sL) - s \sin(tL)}{s^2 + t^2} - \frac{s}{h} \frac{t \sinh(hL) - h \sin(tL)}{h^2 + t^2} \frac{\cosh(sL)}{\cosh(hL)} \right)^2 \right], \quad (32)$$

where $t = (\omega/c)(n^{(1)} - n^{(2)}) + 4\pi/p_0$.

For numerical calculations of the gain coefficient, we use the above mentioned parameters of the polymer network and the CLC TL205/CB15, a mixture of cyclohexane-fluorinated biphenyls and fluorinated terphenyls with a chiral agent 4-cyano-4'-(2-methylbutyl)-biphenyl.^{34,35} The incident light beams have wavelength $\lambda = 532$ nm, ordinary and extraordinary refractive indices of the mixture TL205/CB15 are $n_o = 1.527$ and $n_e = 1.744$, respectively.¹² Additionally, using expressions describing the refractive indices for the waves with the opposite circular polarization³² we estimate the parameters in eq. (32) $n^{(1)} - n^{(2)} \approx 0.005$ and $n^{(1)} + n^{(2)} \approx \sqrt{2(n_o^2 + n_e^2)}$. We also can set $\cos(2\delta) \approx 1$.

In Fig. 5 the gain coefficient versus the grating spacing, $\Gamma(\Lambda)$, is presented at different values of the parameters D , A , B , and K . As it is seen from eq. (32), the gain coefficient increases with an increase of D , its dependence on the grating spacing is shown in Fig. 5a for several values of the parameter D setting parameters A , B , and K to be constant and equal to the values obtained in Ref. 26. The gain coefficient has a maximum approximately at a grating spacing $\Lambda \approx 2.5 \mu\text{m}$ and can reach values that are greater by one order of magnitude or more than those obtained in solid photorefractive inorganic crystals.



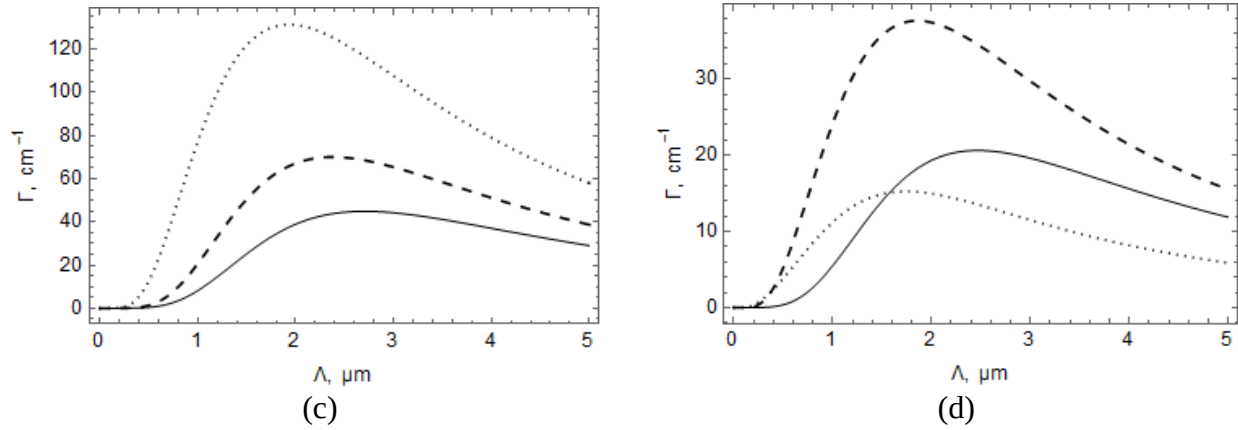


FIG. 5. Gain coefficient versus grating spacing for several values of parameters D , A , B , and K . (a) D (in $10^{-12} \text{V}^{-1} \text{m}^2$) = 3 - solid line, 5 - dashed, 6 - dotted, $A = 1.6 \times 10^2 \text{Jm}^{-3}$, $B = 10^3 \text{Jm}^{-3}$, $K = 8 \times 10^{-12} \text{N}$; (b) B (in Jm^{-3}) = 10^2 - solid line, 10^3 - dashed, 10^4 - dotted, $A = 1.6 \times 10^2 \text{Jm}^{-3}$, $K = 8 \times 10^{-12} \text{N}$, $D = 6 \times 10^{-12} \text{V}^{-1} \text{m}^2$; (c) K (in N) = 10^{-12} - solid line, 5×10^{-13} - dashed, 10^{-13} - dotted, $A = 1.6 \times 10^2 \text{Jm}^{-3}$, $B = 10^3 \text{Jm}^{-3}$, $D = 6 \times 10^{-12} \text{V}^{-1} \text{m}^2$; (d) A (in Jm^{-3}) = 1.6×10 - solid line, 1.6×10^2 - dashed, 1.6×10^3 - dotted, $B = 10^3 \text{Jm}^{-3}$, $K = 8 \times 10^{-12} \text{N}$, $D = 6 \times 10^{-12} \text{V}^{-1} \text{m}^2$.

Plots in Figs. 5b, 5c, and 5d show the influence of parameters B , K , and A on $\Gamma(\Lambda)$, respectively. The gain coefficient increases with both an increase of parameter B (Fig. 5b) and a decrease of parameter K (Fig. 5c). It can be understood that because the translation of the fibrils in a more elastic polymer network (i.e. polymer network with greater parameter B) and in a less elastic CLC (i.e. in CLC with smaller parameter K) induces greater perturbations of the director field (see Figs. 4c and 4d). However, the influence of parameter A (Fig. 5d) is more complex: dependence of the gain coefficient on parameter A is nonmonotonic. At first, $\Gamma(\Lambda)$ increases with an increase of A , then after reaching maximum value $\Gamma(\Lambda)$ decreases with a further increase of A . This is connected with a competition of two processes in the behavior of the director grating magnitude $\varphi(z)$: with an increase of parameter A the maximum of the function $\varphi(z)$ increases, but at the same time the area of considerable values of $\varphi(z)$ decreases (see Fig. 4b). Note that as calculations show the gain coefficient increases with an increase of the CLC pitch, although the influence of the pitch is weak.

4. Conclusions

We have developed a theoretical model describing the energy gain of a weak signal beam interacting with a strong pump beam at a diffraction grating in a hybrid photorefractive-cholesteric cell stabilized in the planar state by the polymer network. Two incident interfering light beams induce the space-charge field in the photorefractive substrate. This field penetrates into the CLC cell and interacts with the charged movable fibrils of the polymer network. Translation of the fibrils under the spatially periodic space-charge field leads to the spatially

periodic director deformation (director grating). We show that the dominating contribution into the director grating magnitude arises from the fibril translation at the photorefractive substrate. Direct interaction of the photorefractive field with the charged fibrils in the CLC bulk gives negligibly small contribution, because of the fast decrease of the field with distance from the substrate.

Coupling of the light beams on the induced director grating leads to the energy gain of the weak signal beam. We calculated the gain coefficient versus the grating spacing and analyzed the influence of the polymer network parameters and the CLC elasticity on the gain. In particular, it is shown that the polymer network with stronger elasticity and the CLC with less elasticity promotes an increase of the gain coefficient. We found out that the dependence of the gain coefficient on the director anchoring with the polymer network has a nonmonotonic character and an optimal value of the anchoring energy. Finally, we show that the gain coefficient in the CLC in the planar state can reach values, which are at least one order of magnitude higher than those obtained in solid photorefractive crystals.

Acknowledgements

This work has been partially supported by STCU Grant P649a and COST Action CA17139.

References

- 1G. P. Wiederrecht, B.A. Yoon, and M. R. Wasielewski, *Science* **270**, 1794 (1995).
- 2I. C. Khoo, B. D. Guenther, M. V. Wood, P. Chen, and M.-Y. Shih, *Opt. Lett.* **22**, 1229 (1997).
- 3H. Ono and N. Kawatsuki, *J. Appl. Phys.* **85**, 2482 (1999).
- 4L. Solymar, D. J. Webb, and A. Grunnet-Jepsen, *The Physics and Applications of Photorefractive Materials* (Clarendon Press, Oxford, 1996).
- 5G. Cook, C. J. Finnan, and D. C. Jones, *Appl. Phys. B: Lasers Opt.* **68**, 911 (1999).
- 6E. V. Rudenko and A. V. Sukhov, *JETP Lett.* **59**, 142 (1994).
- 7E. V. Rudenko and A. V. Sukhov, *JETP* **78**, 875 (1994).
- 8I. C. Khoo, H. Li, and Y. Liang, *Opt. Lett.* **19**, 1723 (1994).
- 9A. Brignon, I. Bongrand, B. Loiseaux, and J. P. Huignard, *Opt. Lett.* **22**, 1855 (1997).
- 10F. Kajzar, S. Bartkiewicz, and A. Miniewicz, *Appl. Phys. Lett.* **74**, 2924 (1999).
- 11S. Bartkiewicz, K. Matczyszyn, A. Miniewicz, and F. Kajzar, *Opt. Commun.* **187**, 257 (2001).
- 12G. Cook, J. L. Carns, M. A. Saleh, & D. R. Evans, *Mol. Cryst. Liq. Cryst.* **453**, 141 (2006).
- 13D. R. Evans, & G. Cook, *J. Nonlinear Opt. Phys. Mater.* **16**, 271 (2007).
- 14D. R. Evans, G. Cook, V. Yu. Reshetnyak, C. M. Liebig, S. A. Basun, and P. P. Banerjee, "Inorganic-Organic Photorefractive Hybrids", in: Pierre-Alexandre Blanche (Ed.), *Photorefractive Organic Materials and Applications* (Springer, 2016), pp. 223-247.
- 15V. Yu. Reshetnyak, I. P. Pinkevych, G. Cook, D. R. Evans, and T. J. Sluckin, *Phys. Rev. E* **81**, 031705 (2010).
- 16N. V. Tabiryan and C. Umeton, *J. Opt. Soc. Am. B* **15**, 1912 (1998).
- 17D. C. Jones and G. Cook, *Opt. Commun.* **232**, 399 (2004).
- 18V. Yu. Reshetnyak, I. P. Pinkevych, G. Cook, D. R. Evans and T. J. Sluckin, *Mol. Cryst. Liq. Cryst.* **560**, 8 (2012).
- 19V. Yu. Reshetnyak, I. P. Pinkevych, T. J. Sluckin, G. Cook, and D. R. Evans, *J. Appl. Phys.* **115**, 103103 (2014).
- 20R. Bao, C. M. Liu and D.-K. Yang, *Appl. Phys. Express* **2**, 112401 (2009).

- 21S. S. Choi, F. Castles, S. M. Morris and H. J. Coles, Appl. Phys. Lett. **95**, 193502 (2009).
- 22J. Chen, S. M. Morris, T. D. Wilkinson and H. J. Coles, Appl. Phys. Lett. **91**, 121118 (2007).
- 23H. Ren, D. Fox, P. Andrew Anderson, B. Wu and S. T. Wu, Opt. Express **14**, 8031 (2006).
- 24S. Y. Lu and L. C. Chien, Appl. Phys. Lett. **91**, 131119 (2007).
- 25H. Nemati, D.-K. Yang, K.-L. Cheng, C.-C. Liang, J.-W. Shiu, C.-C. Tsai and R. S. Zola, J. Appl. Phys. **112**, 124513 (2012).
- 26H. Nemati, S. Liu, R. S. Zola, V. P. Tondiglia, K. M. Lee, T. White, T. Bunning and D-K. Yang, Soft Matter **11**, 1208 (2015).
- 27P. Yeh, *Introduction to Photorefractive Nonlinear Optics* (Wiley, New York, 1993).
- 28H. Nemati, S. Liu, A. Moheghi, V. P. Tondiglia, K. M. Lee, T. J. Bunning, D-K. Yang, J. Mol. Liq. **267**, 120 (2018).
- 29M. E. McConney, V. P. Tondiglia, J. M. Hurtubise, L. V. Natarajan, T. J. White, T. J. Bunning, Adv. Mater. **23**, 1453 (2011).
- 30O. Francescangeli, S. Slussarenko, F. Simoni, D. Andrienko, V. Reshetnyak, and Y. Reznikov, Phys. Rev. Lett. **82**, 1855 (1999).
- 31Y. Kurioz, V. Reshetniak, and Y. Reznikov, Mol. Cryst. Liq. Cryst. Sci. Technol., Sect. A **375**, 535 (2002).
- 32P. G. de Gennes and J. Prost, *The Physics of Liquid Crystals* (Clarendon Press, Oxford, 1993), Ch 6.
- 33H. Kogelnik, Bell Syst. Tech. J. **48**, 2909 (1969).
- 34J. K. Gupta, N. L. Abbott, Langmuir. **25**, 2026 (2009).
- 35T. A. Kumar, K. V. Le, S. Aya, S. Kang, F. Araoka, K. Ishikawa, S. Dhara & H. Takezoe, Phase Transitions **85**, 888 (2012).