Holographic formation of phase diffractive elements for light beams conversion with photo-induced absorption coefficient changing in PDLCs

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Abstract. A theoretical model of the holographic formation of diffractive optical elements for the Gaussian light beams conversion into Bessel-like ones in polymer-dispersed liquid crystals (PDLCs) is developed. The model is based on the solution of the kinetic equations of photopolymerization-diffusion processes of holograms recording in PDLCs by light beams with inhomogeneous amplitude and phase spatial distributions. The model additionally takes into account the photo-induced changings in the absorption coefficient of the PDLC. It is shown that the absorption changings decreases the rate of the refractive index’s spatial profile decline over the thickness of the material. In this case, when the characteristic time of the absorption changing is much less than the characteristic polymerization time (the absorption changes more rapidly than the photopolymerization takes place), the diffractive element can be characterized by a homogeneous distribution of the refractive index over the thickness.

1. Introduction

As it’s known, any monochromatic collimated beam will travel in free space without significant spreading over the Rayleigh distance’ [1]. Looking for types of waves, which propagate at the speed of light and remain focused, has been held by scientists since the early 1980s. Some solutions were found [2, 3], but they had infinite energy, remained theoretically interesting but physically unreal.

Then Ziolkowski [4] found new, exact solutions to the free space wave equation for waves, that could be launched with finite energy from a complex antenna and propagates non-diffraacting. Later [5] he proved it experimentally with acoustic waves in water. In 1987, Durnin [6, 7] developed a non-diffracting zero-order Bessel function solution to the free space scalar wave equation and then published experimental evidence for the nearly-non-diffracting nature of these waves, claiming propagation distances far exceeding the normal Rayleigh range before significant beam spreading was observed. Beams of such waves were called Bessel beams, and they still find a lot of applications.

Non-diffracting Bessel-like beams could be generated in several ways: using optical systems [8], holographic methods [9, 10] fixed-length Fabry-Perot cavity [11, 12], etc. The most common method is the transformation of the Gaussian wave’s wavefront by the devices of axicon optics [13, 14], as well as by diffraction elements [15, 16].

Polymer-dispersed liquid crystals (PDLCs) with a dye sensitizer, used as a photosensitive medium for holographic diffraction elements recording [17-20, etc.], can potentially be used to form the light beam conversion elements. The aim of this work is to study the holographic formation of diffraction elements based on the PDLCs for the conversion of light beams.
2. Main equations

Holographic formation of a diffractive optical element (DOE) is performed by two beams of quasi-monochromatic linearly polarized laser radiation with a wavelength lying in the region of the maximum of the dye’s photosensitivity. In the general case, in view of the photo-induced changes in the absorption coefficient, the beams can be described by the following expressions:

\[
E_0(r, t) = e_0 \cdot E_0(r) \cdot e^{i(\omega - k_0 \cdot r - \varphi_0(r)) - \alpha(r, t)|N_0 \cdot r|}, \quad E_1(r, t) = e_1 \cdot E_1(r) \cdot e^{i(\omega - k \cdot r - \varphi_1(r)) - \alpha(r, t)|N_1 \cdot r|},
\]

where \(e_0, e_1\) - unit polarizations vectors of the beams; \(E_0(r), E_1(r)\) - spatial amplitude distributions; \(\varphi_0(r), \varphi_1(r)\) - spatial phase distributions; \(k_0, k_1\) - wave vectors; \(N_0, N_1\) - wave normals; \(\alpha(r, t)\) - absorption coefficient; \(\omega = 2\pi/\lambda, \lambda\) - wavelength of the recording radiation.

In this case, the spatial distribution of the formation field’s intensity is determined by the following expression:

\[
I(r, t) = I^0(r, t) \cdot [1 + m(r, t) \cos(K \cdot r + \varphi_0(r) - \varphi_1(r))],
\]

where \(m(r, t) = 2\sqrt{I_0(r, t) \cdot I_1(r, t) \cdot \left(\frac{e_0 \cdot e_1}{I_0(r, t) + I_1(r, t)}\right)}\) - local contrast of interference pattern;

\[
I_{0,1}(r, t) = \left|E_{0,1}(r, t)\right|^2; \quad I^0(r, t) = I_0(r, t) + I_1(r, t); \quad K = k_0 - k_1.
\]

In the framework of this paper, the process of holographic formation of DOEs for the Gaussian beams into Bessel-like conversion processes is investigated. The wave vectors of the forming beams lie in the \(xOz\) plane of the laboratory coordinate system. The recording scheme is assumed to be symmetric, i.e. the angles of the beams incidence are related by expression \(\theta_0 = -\theta_1\). The PDLC sample is perpendicular to the \(xOz\) plane, with the \(xOy\) plane coinciding with the lateral face of the sample. The polarization of the recording beams corresponds to the own extraordinary waves in the PDLC sample. Vectors \(e_0, e_1\) are parallel to each other and \(Oy\)-axis. Amplitude distributions of the beams are assumed to be uniform \(E_0(r) = E_1(r) = 1\). The reference beam has a plane wave front \(\varphi_0(r) = 0\), while the wavefront of the signal beam is formed by a successively located phase screw and an axicon and is described by the following expression:

\[
\varphi_1(x, y) = \arctg \left(\frac{y}{x}\right) + s \cdot \sqrt{x^2 + y^2}, \quad (3)
\]

where \(s\) – the steepness parameter of the forming axicon’s cone.

When the light falls on the PDLC, the absorbed quantum leads to the excitation of dye molecules, which interact with the initiator to form the primary radical. After the completion of this stage, the dye molecules do not participate in further chemical reactions, passing into a colorless leucoform. Thus, the concentration of the photosensitive dye during the formation of the DOE decreases, which causes a photo-induced decrease in the absorption coefficient [19]:

\[
\alpha(r, t) = \alpha_{sub} + \alpha_0 K_{d0} \cdot \exp\left(-\beta_{sub} \cdot \alpha_0 \times (I_0(x) / \cos \theta_0 + I_1(x) / \cos \theta_1) \right) \cdot t, \quad (4)
\]

where \(\alpha_{sub}\) – substrate’s absorption coefficient; \(\alpha_0\) – absorption of one dye molecule; \(K_{d0}\) – initial concentration of the dye; \(\beta_{sub}\) – quantum yield of the dye.

Below on Fig. 1 are the images of the wavefront of the signal beam (Fig. 1a) in the case under consideration, as well as the corresponding spatial distribution of the intensity of the recording field (Fig. 1b). The shown images are received for the described recording geometry and at following values of parameters: \(\lambda = 633\ nm; \theta_0 = -\theta_1 = 2^\circ; s = \sqrt{2}\pi/0.001\).
Holographic formation of diffractive elements in the PDLC is described by the kinetic equations of the monomer concentration and the refractive index of the material changings [21, 22]:

\[
\frac{\partial M(r,t)}{\partial t} = \text{div} \left[ D_M \text{ grad } M(r,t) \right] - K_g \left[ \alpha_0 \beta K_d \tau_0 I(r,t) \right]^{0.5} M(r,t), \tag{5}
\]

\[
\frac{\partial n(r,t)}{\partial t} = \delta n_p \cdot K_g \left[ \alpha_0 \beta K_d \tau_0 I(r,t) \right]^{0.5} \frac{M(r,t)}{M_n} + \delta n_{lc} \text{ div} \left[ D_{LC} \text{ grad } \frac{M(r,t)}{M_n} \right], \tag{6}
\]

where \( M_n \) – initial concentration of the monomer; \( K_g, K_p \) – parameters of the rate of growth and breakage of the polymer chain, respectively; \( \alpha_0 \) – absorption of one dye molecule; \( \beta \) – parameter of initiation reaction; \( K_d \) – concentration of the dye; \( \tau_0 \) – the lifetime of the excited state of the dye molecule; \( D_M, D_{LC} \) – the diffusion coefficients of the monomer and liquid crystal, respectively; \( \delta n_p, \delta n_{lc} \) – weight coefficients of the contribution of photopolymerization and diffusion processes; \( I(r,t) \) – intensity distribution (2).

Weight coefficients \( \delta n_p \) and \( \delta n_{lc} \) from (6) are found from the Lorentz-Lorentz formula [23]:

\[
\delta n_p = \frac{4\pi}{3} \left( \frac{n_p^2 + 2}{6n_n^2} \right) \left( \alpha_M + \frac{\alpha_p}{l} \right) \frac{\rho_M}{W_M}, \tag{7}
\]

\[
\delta n_{lc} = \frac{4\pi}{3} \left( \frac{n_p^2 + 2}{6n_{st}^2} \right) \left( \alpha_M \frac{\rho_M}{W_M} + \alpha_{LC} \frac{\rho_{LC}}{W_{LC}} \right), \tag{8}
\]

where \( \alpha_M, \alpha_p, \alpha_{LC} \) – polarizability of monomer, polymer, and liquid crystal molecules, respectively; \( \rho_M, \rho_{LC} \) – density of the monomer and liquid crystal, respectively; \( W_M, W_{LC} \) – molecular weights; \( l \) – average length of polymeric chains; \( n_{st} \) – the refractive index of the composition prior to the start of the recording process, determined by the Lorentz-Lorentz formula from the refractive indices of the monomer and the LC [23]:

\[
n_{st} = M_n \cdot \frac{n_M^2 - 1}{n_M^2 + 2} + L_n \cdot \frac{n_{LC}^2 - 1}{n_{LC}^2 + 2}, \tag{9}
\]
where \( n_M, n_{LC} \) – monomer and liquid crystal refractive indices; \( M_n, L_n \) – their initial concentrations.

Equations (5), (6) are differential equations of parabolic type with respect to two spatial coordinates. In the general case, their analytical solution is difficult. Within the framework of the problem under consideration, by dividing the entire \( Oy \)-axis into \( N \) segments \( \Delta y \), such that \( \Delta y \ll \Lambda \), where \( \Lambda \) - the intensity distribution period (see Fig. 1b), the diffusion of the material components along the \( Oy \)-axis can be neglected and the problem can be considered as quasi-two-dimensional. So, its solution can be found in the form of a set of all distributions of the refractive index along the \( Oy \)-axis, defined for each value \( y = y_0 + j \cdot \Delta y \), where \( y_0 \) - the initial value, \( j = 0...N \) - the segment number.

Then the solution of equations (5), (6) for each \( y_j \) in accordance with the procedure [21, 22] can be found as a sum of spatial harmonics of the monomer concentration and the refractive index:

\[
M^j(r, \tau) = \frac{H}{i} \sum_{i=0}^{H} M_i^j(r, \tau) \cos \left[ i \cdot (K_x \cdot x + \varphi_i^j(x)) \right]
\]

\[
n^j(r, \tau) = n_{sl} + \frac{H}{i} \sum_{i=0}^{H} n_i^j(r, \tau) \cos \left[ i \cdot (K_x \cdot x + \varphi_i^j(x)) \right]
\]

(10)

where \( M^j(r, \tau) = M(x, y_j, z, \tau) \); \( n^j(r, \tau) = n(x, y_j, z, \tau) \); \( \tau = t/T_m \) – relative time; \( T_m = \frac{1}{D_M K_x^2} \) – characteristic diffusion time; \( K_x = 2k_0 \cdot \sin \theta_0 \); the phase shift \( \varphi_i^j(x) \) of each harmonic for each \( y_j \) is determined by the following expression:

\[
\varphi_i^j(x) = \varphi_i(x, y_j).
\]

Consequently, the solution of the problem of holographic formation of DOE is reduced to finding the space-time distributions of the harmonic amplitudes \( M_i^j(r, \tau) \) and \( n_i^j(r, \tau) \).

3. **Analytical solution**

Amplitudes of spatial harmonics in accordance with the technique [21, 22] can be found from the solution of the system of \( H+1 \) kinetic equations for the monomer concentration and the refractive index:

\[
\frac{\partial M_i^j(r, \tau)}{\partial \tau} = -i^2 M_i^j(r, \tau) + \sum_{l=0}^{H} a_{i,l}^j(r, \tau) M_i^j(r, \tau)
\]

\[
\frac{\partial n_i^j(r, \tau)}{\partial \tau} M_n = \delta n_{le} \cdot i^2 M_i^j(r, \tau) - \delta n_p \cdot \sum_{l=0}^{H} a_{i,l}^j(r, \tau) M_i^j(r, \tau)
\]

(12)

where the coefficient matrix \( a_{i,l}^j(r, \tau) \):
where \( b^i_j = b^i_j(r, \tau) = \frac{T_p(r, \tau)}{T_m(r)} \) - the ratio of the characteristic polymerization and diffusion times;

\[
T_p(r, \tau) = e^{\frac{\alpha(r, \tau)}{r}} \left[ \frac{2K_b}{\alpha_0 |K_{d}^\tau_0|} \right]^{0.5} - \text{characteristic polymerization time.}
\]

In the framework of the problem under consideration, due to the heterogeneity of the phase distribution of the recording field, the characteristic diffusion time, firstly, depends on the coordinate \( x \) and, secondly, is determined for each \( y_j \):

\[
T_m^i(r) = T_m \left( 1 + \frac{d\phi^i(x)}{dx} \cdot \frac{1}{K_s} \right)^2
\]

For the analysis it is convenient to represent the parameter \( b^i_j(r, \tau) \) as:

\[
b^i_j(r, \tau) = b \left( 1 + \frac{d\phi^i(x)}{dx} \cdot \frac{1}{K_s} \right)^2 e^{\frac{\alpha(r, \tau)}{r}}, \quad b = \frac{T_p}{T_m}, \quad T_p = \frac{1}{K_s} \left[ \frac{2K_b}{\alpha_0 |K_{d}^\tau_0|} \right]^{0.5}.
\]

The ratio of the rates of photopolymerization and diffusion (expressed through a parameter \( b \)) determines the character of the formation process and the distribution of the refractive index of the material [21, 22]. In particular, the predominance of the photopolymerization process can lead to the formation of DOE, the spatial profile of which differs significantly from the interference pattern [24]. The study of the kinetics of the amplitude variation of the spatial harmonics of the refractive index will make it possible to trace the transformation of the DOE profile throughout the entire formation process.

For the analysis of the system of equations (12), it is convenient to introduce the coupling coefficients \( c_{ij}^p(r, \tau) = a_{ij}^p(r, \tau) - i^2 \delta_{ij} \) (\( \delta_{ij} \) - the Kronecker symbol), which characterize the connection of the \( i \)-th harmonic with the \( l \)-th harmonic. The difference between the coupling coefficients \( c_{ij}^p(r, \tau) \) and \( a_{ij}^p(r, \tau) \) characterizes the contribution of diffusion of the monomer to the recording process and is proportional to the square of the harmonic number. The increase in the diffusion contribution with the number of the harmonic reflected by the term \( i^2 \delta_{ij} \) is due to a decrease in the period of the structure and, accordingly, the diffusion time for a given harmonic with respect to the characteristic diffusion time adopted for the first harmonic.

To solve the system of coupled differential equations (12) initial conditions are introduced:

\[
M_0^j(r, \tau = 0) = M_n, \quad M_1^j(r, \tau = 0) = 0 \ldots M_h^j(r, \tau = 0) = 0,
\]
\[ n_{ij}^0 (r, \tau = 0) = 0, \quad n_{ij}^1 (r, \tau = 0) = 0 \ldots n_{ij}^H (r, \tau = 0) = 0 \] (17)

and the operator method is applied similarly to [21, 22]. Then the general solution for the spatial and temporal amplitude distributions of the harmonics of the monomer concentration will be written as:

\[ M_{ij}^j(r, \tau) = M_n \frac{H}{\lambda_j^0} \sum_{i=0}^{H} A_{ij}^j(r, \tau) \exp[i \lambda_j^0(r)] \] (18)

where the functional dependences of the coefficients \( \lambda_j^0(r) \) are defined as the roots of the characteristic equation \( |c_{ij}^j(r, \tau) - \lambda_j^0(r)| = 0 \). Analysis shows that \( \lambda_j^0(r) \) are real, different and negative. The coefficients \( A_{ij}^j(r, \tau) \) are found from (12), taking into account the initial conditions (14) as solutions of \((H+1)\) systems of linear algebraic equations

\[
egin{bmatrix}
1 & 1 & 1 & 1 \\
\lambda_0^1 & \lambda_2^1 & \lambda_3^1 & \lambda_H^1 \\
\lambda_0^2 & \lambda_2^2 & \lambda_3^2 & \lambda_H^2 \\
\lambda_0^3 & \lambda_2^3 & \lambda_3^3 & \lambda_H^3 \\
\lambda_0^4 & \lambda_2^4 & \lambda_3^4 & \lambda_H^4 \\
\lambda_0^H & \lambda_2^H & \lambda_3^H & \lambda_H^H
\end{bmatrix}
\begin{bmatrix}
A_{ij}^0 \\
A_{ij}^1 \\
A_{ij}^2 \\
A_{ij}^3 \\
A_{ij}^4 \\
A_{ij}^H
\end{bmatrix}
= M_n
\begin{bmatrix}
\delta_{ij} \\
c_{ij}^0 \\
\sum_{i=0}^{H} c_{ij}^j c_{ij}^j \\
\sum_{i=0}^{H} c_{ij}^j c_{ij}^j \\
\sum_{i=0}^{H} c_{ij}^j c_{ij}^j \\
\sum_{i=0}^{H} c_{ij}^j c_{ij}^j
\end{bmatrix}
\]

where \( \lambda_j^0 = \lambda_j^0(r), \quad A_{ij}^j = A_{ij}^j(r, \tau), \quad c_{ij}^j = c_{ij}^j(r, \tau) \).

The general solution for the amplitude of the \( i \)-th harmonic of the refractive index for each \( y_j \) has the form [23]:

\[ n_{ij}^j(r, \tau) = n_{ij}^j(r, \tau)|_p + n_{ij}^j(r, \tau)|_{lc}, \] (19)

where \( n_{ij}^j(r, \tau)|_p = \delta n_p \cdot \sum_{i=0}^{H} a_{ij}^j(r, \tau) \sum_{g=0}^{H} A_{ij}^j(r, \tau) \frac{\exp[i \lambda_g^j(r)] - 1}{\lambda_g^j(r)} \),

\[ n_{ij}^j(r, \tau)|_{lc} = \delta n_{lc} \cdot i^2 \sum_{g=0}^{H} A_{ij}^j(r, \tau) \frac{\exp[i \lambda_g^j(r)] - 1}{\lambda_g^j(r)} \).

Thus, the expressions (10), (18) and (19) represent a general solution of the equations of holographic formation of DOE in a PDLC with a dye-sensitizer and determine the kinetics of the spatial distribution of the monomer concentration and the refractive index.

4. Numerical simulation

To determine the kinetics of the amplitudes of the spatial harmonics of the refractive index of the PDLC \( n_{ij}^j(x_0, \tau) = n_l (x_0, y_0, z = 0, \tau), \quad x_0 = y_0 = 0 \) (the center of the sample, absorption is not taken into account), the numerical simulations were made by expressions (10), (18) and (19) for \( \delta n_{lc} / \delta n_p = 0.5 \) and for two values of the parameter \( b = 0.25 \) and \( b = 5 \).

Figure 2 shows the results of numerical simulation of the amplitudes of the refractive index harmonics kinetics obtained from expression (19) for the intensity distribution from Fig. 1b.
Figure 2. Kinetics of refractive index harmonics for $b = 0.25$ (a) and $b = 5$ (b)

Fig. 2 shows that in the case of predominance of the photopolymerization formation process over the diffusion (Fig. 2a), a significant contribution to the change in the refractive index is made by higher spatial harmonics. In particular, the second one in absolute magnitude exceeds the first harmonic by 0.08 rel. units, starting from $\tau = 1$ and up to the completion of the recording process. On the other hand, if the diffusion process predominates (Fig. 2b), the formation process is close to linear and the main (first) harmonic contributes the most to the change in the distribution of the refractive index.

Fig. 3 shows the results of numerical simulations of the spatial distributions of the refractive index of the PDLC after the completion of all the formation processes obtained from expression (10) for the interference pattern (2), (Fig. 1b).

Figure 3. Spatial distributions of refractive index for $b = 0.25$ (a) and $b = 5$ (b)

Fig. 3 shows that if the diffusion process predominates (Fig. 3b), the spatial distribution of the refractive index is close to the interference pattern (see Fig. 1b). This allows us to conclude that the holographic DOE has a minimum of distortion (which is extremely important for the efficient conversion of light beams), if the composition of the PDLC and the formation conditions are selected in such a way that the ratio of the photopolymerization and diffusion rates meets the requirement $b \gg 1$.

Taking into account the absorption in describing the process of DOE’s holographic formation leads to the dependence of the amplitudes of the spatial harmonics on the $z$ coordinate (along the thickness of the sample). In this case, the photo-induced changing of the absorption coefficient causes additional time dependence. Fig. 4 shows the results of numerical simulations of the refractive index harmonics amplitudes as a function of the $z$ coordinate for $b = 5$, after the completion of the DOE formation processes for two cases: steady-state attenuation (Fig. 4a) and its photo-induced changing (Fig. 4b). Parameters of photo-induced absorption changing were taken from [19].
Figure 4. Dependence of harmonics’ amplitudes along the samples’ thickness for steady-state absorption (a) and for its photo-induced changings (b).

Fig. 4 shows that the changing absorption decreases the rate of decline of the spatial profile of the refractive index over the thickness of the sample. In this case, when the characteristic time of the change in absorption is much less than the characteristic polymerization time (the absorption changes more rapidly than the photopolymerization takes place), the diffraction element can be characterized by a homogeneous distribution of the refractive index over the thickness.

5. Conclusion

Thus, the paper presents a theoretical model of holographic formation of diffractive optical elements (DOE) for the conversion of light beam fields into Bessel-like in PDLCs with a sensitizer dye. The model allows numerical simulation of the course of the formation of DOE depending on the experimental conditions and parameters of the prepolymer composition. Based on the simulation results, it is shown that the ratio of the diffusion and photopolymerization rates during the DOE formation process should be ensured to the level $b \gg 1$. In addition, it was shown that the effect of the photo-induced changing of the absorption coefficient causes a nearly uniform distribution of the amplitude of the change in the refractive index along the thickness of the sample.

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