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Theoretical and experimental investigation of photopolymer chirped gratings formation

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Abstract: In the work an analytical model of transmission holographic grating formation by light beams with non-uniform phase-amplitude profiles is presented. The model takes into consideration influence of light absorption of a photopolymer with dye-sensitizer and nonlinearity of photo-polymerization on the spatial profiles and diffraction properties of the chirped grating. The model of diffraction efficiency and selective properties of chirped gratings is also presented. The results of fitting experimental data and numerical ones are shown and discussed.

OCIS codes: (090.7330) Volume holographic gratings; (160.5470) Polymers; (050.1590) Chirping; (050.1950) Diffraction gratings.

Introduction

In the last years photopolymers find application in display holography, optical data storage, holographic optical elements manufacturing and optical interconnections including free-space interconnections [1-2]. One of the interesting application of photopolymer gratings is wavelength separation in POF WDM (polymer optical fiber with wavelength division multiplexing) systems. Light of two different wavelength will be combined and propagate along the fiber. The same kind of grating separates the two wavelengths at the end of the fiber. Since POF exhibit large numerical apertures, i.e. large angular spectrum of output radiation >10° – whereas polymer gratings have really narrow angular response ~2° at the level of –3 dB – our main task is to design a photopolymer grating with the angular bandwidth suitable for POF systems. Good solutions for the problem are chirped gratings. But photopolymers are complex non-linear holographic material that is why in order to find the optimum conditions of record it is necessary to create the theoretical model and carry out some experiments to check the model. Then using the model it is possible to find the optimum parameters of record for each special practical problem.

So the main task of the paper is creation an analytical model of record and readout chirped grating in absorbing photopolymers. And to check the model we need to carry out experimental investigation and compare the experimental data with the results of numerical simulation.
Analytical model

In photopolymer (PPM) under the light action the process of radical polymerization is occurred. The polymerization leads to record of a phase grating accordingly to the interference pattern of record beams. Let’s consider the case when two coherent light beams with amplitude and phase distributions $E_j(x,y)$ and $\phi_j(x,y)$ incident in transmission geometry on absorbing PPM(Fig.1a).

The interference pattern in this case can be described as:

$$I(x,y) = e^{-\alpha_0} |E_0(x,y) + E_1(x,y)|^2 = I_0(x,y)[1 + m(x,y) \cos \phi(x)],$$

where $E_j(x,y) = e_j E_j(x,y) e^{i\phi_j(x,y)}$, $\phi(x) = \{\phi_0 - \phi_1\}$, $k_j = \nabla \phi_j$ and $e_j$ – wave vector and vector of state of polarization of $j$–th beam, $j=0,1$; $\alpha$- PPM optical absorption coefficient, $I_0(x,y) = (I_0(x,y) + I_1(x,y)) e^{-\alpha_0}$, $m(x,y) = 2 \sqrt{I_0(x,y) I_1(x,y) \cdot (e_1 \cdot e_0) / (I_0(x,y) + I_1(x,y))}$, $I_j(x,y) = |E_j(x,y)|^2$.

The process of phase grating formation in PPM with dye-sensitizer can be described with the help of kinetics equations for monomer concentration $M$ and refraction index $n [3-4]$:

$$\frac{\partial M}{\partial t} = \text{div}(D(M) \text{grad } M) - K_g K_b^{-k} (\alpha_0 \beta(K) I(x,y) \tau_0)^k \cdot M,$$

$$\frac{\partial n}{\partial t} = \delta n_p K_g K_b^{-k} (\alpha_0 \beta(K) I(x,y) \tau_0)^k \cdot M / M_n + \delta n_i \text{div}(D(M) \text{grad } M / M_n),$$

$$D = D_m \exp[-s \cdot (1 - M / M_n)],$$

where $I(x,y)$ is defined by (1), $<K>$ is a dye concentration, $n = n(t,y,x)$ – refraction index, $M = M(t,y,x)$ is the monomer concentration, $M_n$ – initial monomer concentration, $\alpha_0$, $\beta$ is a photorefraction parameters, $K_g$ is a parameter describing rate growth of the polymeric chain, $K_b$ is a parameter describing a velocity of the break of the polymeric chain, $D_m$ is a monomer diffusion coefficient, $s$ - describes rate of decreasing $D_m$, $\delta n_p$ and $\delta n_i$ are the process parameters describing contributions of photo-polymerization process and monomer diffusion in refraction index grating amplitude respectively [3], $k$ – characterized the nonlinearity level of photopolymerization process [4].

The solutions of the kinetics equations we shall find in the form:

$$M(\tau) = M_0(\tau) + M_1(\tau) \cos \phi(x), \quad n(\tau) = n_{st} + n_0(\tau) + n_1(\tau) \cos \phi(x),$$

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where $M_0(\tau)=M_0(\tau, x, y)$, $n_0(\tau)=n_0(\tau, x, y)$ and $M_1(\tau)=M_1(\tau, x, y)$, $n_1(\tau)=n_1(\tau, x, y)$ – average values and amplitudes of gratings of $M$ and $n$, $n_s$ – the value of refractive index of PPM at $\tau=0$; $\tau=t/T_m$ – relative time; $T_m=1/(D_nK_1^2)$ – diffusion time, $K_1=\sqrt{\nabla \phi} = \varphi^\prime$ - the wave number of grating.

We shall use the Taylor series expansion for $\phi(x)=\phi_0+\phi^\prime x+0.5\phi^\prime\prime x^2$ and a restriction $0.5\phi^\prime\prime \Lambda<\phi^\prime$, where $\Lambda$ - the grating period. Substituting Eqs. 1, 4, 5 into Eqs. 2 and 3 and taking into account that $I_0(x,y)$, $m(x,y)$, $M_0(\tau)$, $M_1(\tau)$, $n_0(\tau)$, $n_1(\tau)$ are slow changing function of $x$ in comparison with $\cos(\phi^\prime x)$ and $M_1(\tau)<M_0(\tau)$, $n_1(\tau)<n_0(\tau)$, we use the property of orthogonality of spatial harmonics in order to find the equations for spatial harmonics:

$$\frac{\partial M_0(\tau)}{\partial t} = -M_0(\tau) \frac{2k}{b_s} \left[1 + m_s^2 k(k-1)/4\right], \quad \frac{\partial n_0(\tau)}{\partial \tau} = \delta n_p M_0(\tau) \frac{2k}{b_s} \left[1 + m_s^2 k(k-1)/4\right],$$

$$(6)$$

$$\frac{\partial M_1(\tau)}{\partial \tau} = -M_1(\tau)b(\tau) - \frac{2k}{b_s} \left[ M_0(\tau)km_s + M_1(\tau) \left(1 + \frac{3}{4} k(k-1)m_s^2\right) \right].$$

$$(7)$$

$$\frac{\partial n_1(\tau)}{\partial \tau} = -\delta n_p \frac{M_1(\tau)}{M_n} b(\tau) + \delta n_p \frac{2k}{b_s} \left[ \frac{M_0(\tau)}{M_n} km_s + \frac{M_1(\tau)}{M_n} \left(1 - \frac{3}{4} k(k-1)m_s^2\right) \right].$$

$$(8)$$

where $b(\tau) = b(\tau, x, y) = \exp[-s(1-M_0(\tau)/M_n)]$, $m_s = m(x, y)$, $b = b(x, y) = T_{ps}/T_{mx}$, $T_{ps} = T_p(x, y) = 1/[K_1 K_b \alpha_k \beta(x, y) \tau_0 K_1^{k+1}]$, $T_{mx} = T_m / (1 + x\phi^\prime / \phi^\prime)^2$ – local times of photopolymerization and monomer diffusion.

Integrating the kinetic equations for zero harmonics with initial conditions $M(\tau=0, x, y) = M_n$, $n(\tau=0, x, y) = n_s$, we obtain the following expressions for $M_0(\tau, y)$ and $n_0(\tau, y)$:

$$M_0(\tau, x, y) = M_n \cdot p(\tau, x, y), \quad n_0(\tau, x, y) = n_s + \delta n_p \left\{1 - p(\tau, x, y)\right\},$$

$$(9)$$

where $p(\tau, x, y) = \exp\left[-2^k \tau \cdot (1 + k(k-1)m_s^2/4)/b_s\right]$.

After substitution Eq. 9 into Eq. 7 and integrating we write the final expression describing a behavior of monomer concentration grating $M_1(\tau, x, y)$:

$$M_1(\tau, x, y) = -M_n \cdot f(\tau, x, y),$$

$$(10)$$

where $f(\tau, x, y) = (2^k km_s / b_s^2) \cdot e^{-2^k m_s \tau / b_s} \int_0^\tau p(\tau', x, y) \exp\left(2^k m_s \tau'/b_s - \frac{1}{2} b(\tau', x, y) dt'\right) dt'$, $m_{2s}=1+3k(k-1)m_s^2/8$.

From Eq. 8, using Eqs. 9 and 10, we determine a spatial-time distribution of amplitude of refraction index grating:

$$n_1(\tau, x, y) = n_{1p}(\tau, x, y) + n_{1i}(\tau, x, y)$$

$$(11)$$

where $n_{1p}(\tau, x, y) = \delta n_p \frac{2^k}{b_s} \int_0^\tau p(\tau', x, y) km_s - f(\tau', x, y) m_{2s} dt'$,

$$n_{1i}(\tau, x, y) = \delta n_i \int_0^\tau f(\tau', x, y) b(\tau', x, y) dt'.$$

So, Eq. 5 and Eq. 11 are the general analytical solution of non-linear diffusion-polymerisation process of transmission grating formation by the beams with non-homogeneous amplitude-phase profiles in absorbing photopolymers.
Presenting the grating profile Eq.2 in the view of spectrum of spatial frequencies and profiles of interacting beams $E_j(r)$ in the view of angular spectrums we can define the light distribution after the grating with the help of couple waves equations:

$$\begin{align*}
\frac{\partial E_0(\theta_0, y)}{\partial y} &= -iG_0 \delta n_p \int_{K_{\|}} E_1(\theta_0, K_{\|}, y) \cdot S_m(K_{\|}, y) \cdot e^{i\Delta K_0(\theta_0, K_{\|})y} dK_{\|}, \\
\frac{\partial E_1(\theta_1, y)}{\partial y} &= -iG_1 \delta n_p \int_{K_{\|}} E_0(\theta_0, K_{\|}, y) \cdot S_m(K_{\|}, y) \cdot e^{-i\Delta K_1(\theta_1, K_{\|})y} dK_{\|},
\end{align*}$$

(12)

where $S_m(K_{\|}, y) = \delta n_p^{-1} \int_{K_{\|}} e^{ik_{xy}y} \int_{n_{xy}} n_1(x, y) e^{ik_{xy}x^2/2} e^{i(K_{xy}+K_{\|})y} dxdy$; $E_j(r) = \int E_j(\theta_j, y) e^{-ik_jr} d\theta_j$,

$G = \pi/(\lambda \cos \theta_j), \quad j = 0, 1$; $r$ – radius-vector; $\theta_0(\theta_1, K_{\|}) = m_{1x}\theta_1 / m_{0x} - K_{\|} / m_{1x}$,

$\Delta K_0(\theta_1, K_{\|}) = -(m_1N_0)k_{\theta_1} / N_{0y} + N_{0x}K_{\|} / N_{0y}$, \quad $\theta_1(\theta_0, K_{\|}) = m_{0x}\theta_0 / m_{1x} - K_{\|} / m_{1x}$,

$\Delta K_1(\theta_1, K_{\|}) = -(m_0N_1)k_{\theta_1} / N_{1y} + N_{1x}K_{\|} / N_{1y}$, $k = k' -$ wave number, where we used series expansion of current wave vector $k$, near central direction $k = k' + km\Delta \theta_j$, where $\Delta \theta_j$ – angular deviation from the central direction $k' = kN_j$, subscript $x$ or $y$ in signs means a projection of the vector on the $x$ or $y$ axis.

Using initial conditions $E_0(\tau, y=0) = E_0(\theta)$, $E_1(\tau, y=0) = 0$ it is possible to find the solution of Eqs. 12 with the help of disturbance technique:

$$\begin{align*}
E_0(\tau, y) &= \sum_{k=0}^{\infty} E_{0,2k}(\tau, y)(\delta n_p / n_{st})^{2k}, \quad E_1(\tau, y) = \sum_{k=0}^{\infty} E_{1,2k+1}(\tau, y)(\delta n_p / n_{st})^{2k+1}, \quad (13)
\end{align*}$$

where $E_{0,2k}(\theta_0, y) = -iG_0 \int_0^\infty E_{1,2k-1}(\theta_0, K_{\|}, y)S_m(K_{\|}, y)e^{i\Delta K_0(\theta_0, K_{\|})y} dK_{\|}$,

$E_{1,2k+1}(\theta_1, y) = -iG_1 \int_0^\infty E_{0,2k}(\theta_0, K_{\|}, y)S_m(K_{\|}, y)e^{-i\Delta K_1(\theta_1, K_{\|})y} dK_{\|}$.

Diffraction efficiency we can write on the base of Eq. 13 in the view:

$$\eta_d(\theta, d) = \int |E_1(\theta, d)|^2 d\theta_1 / \int |E_0(\theta_0, 0)|^2 d\theta_0.$$  \quad (14)

So, Eq. 13 and Eq. 14 present the subsequence solution of diffraction problem at readout of chirped grating by light beam with any angular spectrum and enable to find distribution of light field after the grating.

**Numerical simulation and experimental results**

The schemes of experimental setups for record (a) and readout (b) of chirped gratings in acrylomide based photopolymer samples are presented in Fig.2. The record process (Fig.2a) was carried out by beams with Gaussian amplitude distribution. The record beams had plane phase fronts. One beam passing the lens (L) change the plane phase front to spherical one. The diameters of the beams on the front plane of the photopolymer sample (PPM) were equaled, i.e. the distance between the lens (L) and the PPM sample equaled double focus length (2F). Changing the diameters of the record beams ($d_0$) with the spatial frequencies filter (SF) and the focus length (F) we changed the bandwidth of angular spectrum of diverging beam, i.e. chirped grating vector deviation - $\varphi''$.

During the record process the diverging beam shut down for short time and a diffraction
efficiency of the grating is measured. The experimental curves of kinetics of the diffraction efficiency are shown in Fig. 3a. The solid curves in Fig.3a present the results of numerical simulation for the experimental conditions: the record angle $\theta_0=\theta_1=10^0$ (in air), the thickness of the PPM sample $d=85\mu$m, the record beams intensities $I_1/I_0=1$, the optical absorption of PPM sample $\alpha d=2$Nepers, and the material parameters: $\delta n_p=0.003$, $C_n=\delta n_d/\delta n_p=0.01$, $s=1$, $T_m=4.3$sec, $T_p=(10.7–21.4)$sec, $D_m=19.3\times10^{-15}$m$^2$/s. The material parameters were obtained from fitting experimental data and results of numerical simulation on the base of the model presented.

![Fig.2 Experimental setup](image)

C – collimator, SF- spatial filter, BS – beam splitter, M1,M2 – mirrors, L – lens, PPM – photopolymer sample on rotation mount, PhD1, PhD2 – photodiodes.

After record the lens (L) was removed and the beam after the mirror (M2) was shut down. Rotating the rotation mount with the photopolymer sample on the angle $\Delta\theta_0$ we measured the angular response of the chirped grating by the beam with plane phase front (Fig.2b). The experimentally obtained curves of normalized angular response of the chirped gratings are presented in Fig.3b. The solid curves in Fig.3b were numerically calculated for the experimental conditions and the material parameters corresponding to Fig.3a.

![Fig.3](image)

Experimentally obtained angular bandwidth of chirped gratins in degrees at the level -3dB for different focus length of the lens (L on the Fig.2a) $F$ and recording beams diameters $d_0$ are presented in the Table 1.

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<th>$d_0$</th>
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<th>16mm</th>
<th>20mm</th>
<th>40mm</th>
<th>Plane beams</th>
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<td>3mm</td>
<td>4.6</td>
<td>3 (2)</td>
<td>2.7</td>
<td>1.7</td>
<td>(1)</td>
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Table 1 Experimentally obtained angular bandwidth of chirped gratins.
Table 1

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<tr>
<td>4mm</td>
<td>7.1</td>
<td>5.7 (3)</td>
<td>-</td>
<td>1.7 (1)</td>
</tr>
<tr>
<td>5mm</td>
<td>9.4 (4)</td>
<td>7</td>
<td>-</td>
<td>1.7 (1)</td>
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Conclusion

In the work the analytical model of transmission holographic chirped grating formation by light beams with non-uniform phase-amplitude profiles is presented. The model takes into consideration influence of light absorption of PPM with dye-sensitizer and nonlinearity of polymerization on diffraction properties of the grating. The expressions for diffraction properties of chirped gratings are also presented. The results of experiments of kinetics of diffraction efficiency and angular response of chirped gratings are shown for different wave vector deviations. The results of fitting experimental data and numerical ones have shown good agreement between experimental and theoretical data that enables to say about adequacy of the model presented.

References