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Inharmonic theoretical models for photopolymers gratings formation

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Abstract: We present an analytical model of the dynamics of spatial harmonics of holographic grating profiles, taking into consideration record mechanisms (polymerization and components diffusion), absorption, interference pattern contrast and optional photopolymerization nonlinearity. On the basis of numerical simulation we compare the model presented with the non-local polymerization driven diffusion model. The results of fitting experimental data with the model results are also presented.

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

Introduction

Photopolymeric diffraction gratings have found many new possible fields of application, i.e.e arrays with μm sizes. The rapid speed of formation, uniformity, good domain-size control, switchability, and the addition of optically-active components make the H-PDLCs (holographic polymer-dispersed liquid crystals or H-PDLCs) an attractive polymeric/organic platform for optically-active PBGs and organic nanophotonics interesting for optical communication as commutation units for wavelength division multiplexing systems [1].

Holographic patterning allows rapid fabrication (~seconds) and high design flexibility. Such patterning generally falls into two categories: lithography and photopolymerization. The photopolymerization process typically involves holographically initiating polymerization of a monomer within a syrup. Non-reactive components in the syrup phase, such as liquid crystals, separate from the regions of rapid molecular weight increase. This enables one-step fabrication of electro-optic structures such as electric-field-switchable transmission and reflection structures. The inherent flexibility of the photopolymerization process allows fabrication of switchable multidimensional structures as well as reflective Bragg stacks that are not directly accessible with lithography. [1]

All these issues demand a theory capable of predicting the behaviour of the harmonics for different parameters of photopolymer and holographic record conditions. That is why the aim of this work is to analyse the dynamics of forming inharmonic spatial distribution of photopolymeric gratings, using analytic techniques to solve coupled kinetic equations governing the spatial harmonics (component amplitudes) and comparing the results of this model with those of an existing model [2].

General equations

We assume the photopolymerization equations describing record process in photopolymer media with dye sensitizer [3] are:

$$\frac{\partial M}{\partial t} = \text{div}(D_m \text{grad } M) - h[I(\mathbf{r})]^k M, \quad \frac{\partial n}{\partial t} = \delta n_p h[I(\mathbf{r})]^k \frac{M}{M_n} + \delta n_i \text{div} \left[D_m \text{grad} \frac{M}{M_n} \right], \quad (1)$$

where $h = K_g K_b^{-1/2} (\alpha_0 \beta \tau_0 <K>)^k$, $M = M(\mathbf{r}, t)$ – monomer concentration, $n = n(\mathbf{r}, t)$ – refraction index, $I(\mathbf{r}, t)$ – intensity of interference pattern, $\mathbf{r}$ – radius vector of spatial point; D_m – the diffusion coefficient; δn_p , δn_i – change of n due to polymerization and diffusion of the components of material, respectively; α_0 – a dye molecule absorption coefficient, $<K>$ – a dye concentration responsible for initiation of photo-polymerization, τ_0 – the excited conditions relaxation time, K_g – chain growth parameter, K_b – chain break parameter.

We shall assume that the exposure/recording is realized using plane waves. Therefore the interference pattern, taking into consideration of absorption, can be described as:

$$I(\mathbf{r}) = I_0(y) \exp(-\alpha y) (1 + m_y \cos(K_1 x)), \quad (2)$$

where $I_0(y) = I_y^0 + I_y^1$; $m_y = m(y) = 2\sqrt{I_y^0 I_y^1} \cdot (\mathbf{e}_1 \cdot \mathbf{e}_0) / (I_y^0 + I_y^1)$ – contrast of interference pattern; $I_y^j = I(y) = I_j \exp(-\alpha y / \cos(\theta_j))$, $I_j = |E_j|^2$, $j = 0, 1$; $K_1 = |\mathbf{K}_1| = |\mathbf{k}_0' - \mathbf{k}_1'|$, $\mathbf{K}_1 = \mathbf{k}_0 - \mathbf{k}_1$ – grating vector, $\mathbf{r}$ – radius-vector, E_j – amplitudes, $\mathbf{k}_j$ – wave vectors and $\mathbf{e}_j$ – polarization vector of recording waves, respectively

Analytical model

We shall be finding a solution of Eqns (1) using the harmonics expansions:

$$M(\tau, x, y) = \sum_{i=0}^m M_i(\tau, y) \cos(i \cdot K_1 x), \quad n(\tau, y) = n_{st} + \sum_{i=0}^m n_i(\tau, y) \cos(i \cdot K_1 x) \quad (3)$$

where $M_j(\tau, y) = \frac{1}{2\pi} \int_{-\pi}^{\pi} M(\tau, x, y) \cos(j K_1 x) dK_1 x$, $n_j(\tau, y) = \frac{1}{2\pi} \int_{-\pi}^{\pi} n(\tau, x, y) \cos(j K_1 x) dK_1 x$ –

harmonic amplitudes of monomer concentration and refraction index respectively; $\tau = t/T_m$ – relative time; $T_m = 1/(D_m K_1^2)$ – characteristic diffusion time, n_{st} – refraction index of the polymer sample before the record.

To obtain kinetics equations for M_j , n_j from Eqns. (1) we use the Taylor series expansion of the function $(I(\mathbf{r}))^k = I_0^k(y) (1 + m_y \cos(K_1 x))^k$, taking into consideration three first terms:

$$I^k(\mathbf{r}) \approx I_0^k(y) \left[1 + \frac{k}{m_y^{-1}} \cos(K_1 x) + \frac{k(k-1)}{2m_y^{-2}} \cos^2(K_1 x) \right]. \quad (4)$$

The error in using the approximation in Eqn (4) over the area of parameters values given by $0.75 < m_y < 1$ and $0.1 < k < 0.75$, is typically between 1.5% and 3%, and in general less than 1.5% for the area of parameters values given by $m_y < 0.75$ and $0.75 < k < 1$.

Substituting Eqns. (2)-(4) into Eqn. (1), and using property of orthogonality for harmonics in Eq. (3), we obtain a system of coupled kinetic equations:

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The second theoretical model used here is referred to as the non-local polymerization driven diffusion model (NPDD) and some recent results obtained using this mode are presented in [2]. For the sake of conciseness we do not present the expressions governing this model here, we simply compare the predictions of the two models.

Numerical simulation and comparison results

In order to compare the models we should compare the parameters (terminology, notations) used in the two models. In the model presented above the main parameters are: $b = T_p/T_m$, where $T_p \sim hI_0^{-k}$, $T_m = 1/(K^2 D_m)$, and $m(y) = 2\sqrt{m_0}/(1 + m_0)$, where, $m_0 = I_0/I_1$ – record beams intensities ratio, k – non-linearity parameter. The main parameters appearing in the NPDD model are $R = D_m K^2 / F_0$ where $F_0 = \kappa I_0^\gamma$, $\gamma = k$, $\kappa = h$, i.e. $R = b$, and σ – non-local parameter. The rest of the terminology used is less significant as we will compare the normalized curves.

In Fig.1 we present the dependences of normalized amplitudes of harmonics of refraction index gratings on record time. At the left side we shown the results of NPDD model (a, c), and at the right side – of the model presented here (b, d). The parameters are the following: $m_0 = 0.3$, $k = \gamma = 0.85$, $\sigma = 1/32$ (for NPDD), $R = b = 0.1$ (a, b) and $R = b = 5$ (c, d).

Figure 1

From this comparison it can be seen that time dependence of curves are very similar, but the amplitudes of harmonics are different for the 1st and 2nd. For the 0th harmonics we have the same behavior for all parameters. For the 3rd harmonic the difference increase with increase of $R = b$. The differences appear to be primarily caused by the inclusion of the non-local effect in the NPDD model. Note in particular that for higher harmonics the NPDD model amplitudes decrease in comparison with the predictions of the model presented here. However it must be emphasized that there is general agreement regarding the behavior of the two sets of results.

Numerical simulation and experimental results

The photopolymer composition used in our experimental research had a large light absorption and the NPDD model used does not take into account this fact. To proceed we fit our experimental results with the model presented in the paper.

The schemes of experimental setup for readout of gratings in acrylamide based photopolymer samples are presented in Fig.2a. After recording the beam power, after the mirror (M2), was shut down. Rotating the mount, with the photopolymer sample, through the angle $\Delta\theta_0$ we measured the angular response of the grating recorded. The experimentally obtained curves of normalized angular response of the grating are presented in Fig.2b. The solid curves in Fig.2b were numerically calculated for the experimental conditions: using the record angle $\theta_0 = \theta_1 = 10^\circ$ (in air), the thickness of the PPM sample $d = 85\mu\text{m}$, the record beams intensities $I_1/I_0 = 0.3$, the optical absorption of PPM sample $\alpha d = 2\text{Nepers}$ and the material parameters: $k = 0.85$, $\delta n_p = 0.006$, $C_n = \delta n_i/\delta n_p = 0.012$, $T_m = 2.5\text{sec}$, $T_p = 82\text{sec}$. The material parameters were obtained by fitting the experimental data using numerical software based on the model presented above.

C – collimator, SF- spatial filter, BS – beam splitter, M1,M2 – mirrors, PPM – photopolymer sample on rotation mount, PhD0 , PhD1 , PhD2 – photodiodes for the diffracted beams in zero, first and second diffracted orders.

Figure 2

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.

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Conclusions

In the work the analytical model of transmission holographic grating formation with optional number of spatial harmonics 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 comparison of numerical simulation results of the model presented with the results of NPDD model shown qualitative agreement and the same behavior with changing model parameters. But some differences in amplitudes of spatial harmonics were also observed. We think that the differences can be caused by non-local effect, which is taken into account by the NPDD model.

The results of fitting experimental data and numerical ones have shown good agreement between experimental and theoretical data that indicating the usefulness of the model presented.

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