Holographic recording of superimposed gratings by angle multiplexing in photopolymers with light induced changing of optical absorption

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ABSTRACT

The results of theoretical investigation of recording superimposed holographic gratings of transmitted type in optical absorbent photopolymeric materials are presented in the report. We consider the case of serial record of superimposed holographic gratings with angle multiplexing. The influence of photopolymer material parameters on diffraction effectiveness of superimposed holographic gratings is investigated in the work. It shows that the change of optical absorbent photopolymer material during record may strongly change integrated diffraction characteristics of the recorded holographic gratings.

Keywords: analytical model, photopolymer media, photo-induced absorption, superimposed transmitted gratings, angle multiplexing technique.

1. INTRODUCTION

Nowadays advanced technologies allow to create optical channel-shared elements in WDM optical de-multiplexing systems and realize optical memory storage. These technologies exhibit some advantages including smooth performance and relative low cost.

One of the technologies is usage of superimposed diffraction gratings in holographic materials, specifically photopolymers.

In the paper we present the calculation technique and the model of superimposed grating recording with predetermined diffraction characteristics as well as the influence of series material parameters on superimposed grating diffraction characteristics.

The results of numerical simulation of 6 superimposed gratings recording with equal diffraction efficiencies 10\% at initial optical absorption = 4 Nepers and final = 0.1 Nepers are shown in the paper.

Finally, experimental data and calculation of superimposed holographic grating recording are fitted.

2. GENERAL EQUATIONS

Free radical polymerization process initiated in photopolymeric material exposed to light has the following basic stages. At first, initiator photo decomposition with forming initial radicals occurs under action of light quantum, then, as the consequence, forming active polymeric radicals at interaction of initial radicals with monomer molecules and polymeric chain growth are observed. The last stage of free radical polymerization is conversion of polymeric radical into non-active form and forming final product called polymer molecule.

Let consider superimposed grating recording in photopolymer material in details. Assume that the photopolymeric layer of thickness $d$ is exposed with 2 light beams with the wave vectors $k_0$, $k_1$ and amplitudes $E_0$, $E_1$ at the angles $\theta + \Psi$.
and \( \theta - \Psi \) as shown in Fig. 1a. Fig. 1b shows the vector diagram of non-symmetrical record model (\( \Psi > 0 \)).

The interference pattern is formed in the result of laser beams interaction. The more intensive light exposition at the surface (Fig. 1a), the quicker polymerizing reaction.

Consequently, concentration gradient makes monomer diffuse from dark areas into light ones where diffused monomer is polymerized as pointers show in Fig.2. Intensity of light is non uniform within thickness of polymer sample as the result of polymer optical absorption. (Fig. 1a). The intensity achieves itself maximum when \( y=0 \) and itself minimum when \( y=d \). So, polymerizing reaction goes quicker in the lighter areas.

At start of hologram recording, the polymerizing reaction goes quicker on the surface of the material. The concentration gradient leads to monomer diffusion from dark areas. With increasing coordinate \( y \) the intensity of light decreases and the monomer diffusion from dark areas goes down (Fig. 2a). At the recording goes on, photopolymeric material becomes lighter, that increases monomer diffusion from dark to light areas within thickness of material. In the dark ranges monomer diffusion decreases at small \( y \) as the monomer is depleted (Fig. 2b).

Finally, holographic phase grating with the grating vector \( K \) is recorded in photopolymeric material. The grating vector \( K \) forms angle \( \Psi \) with \( z \) axis.

For mathematical description of the process of recording superimposed holographic gratings in photopolymers we take as the base the equations for monomer concentration \( M \) and refraction index \( n \) presented in 1. These equations describe the record of single holograms. The rate of change of monomer concentration \( M \) is described with the following relation:

\[
\frac{\partial M}{\partial \tau} = \text{div}(D_m \text{ grad } M) - K_g K_b^{-1/2} \sqrt{1/\tau_0 + \beta M} M^{3/2}, \tag{1}
\]
where \( I(r) \) – intensity of light in the point with radius-vector \( r \), \( D_m \) – the monomer diffusion coefficient, \( \alpha \) -the dye absorption coefficient, \( K \) – the dye concentration, \( \beta \) - parameter of photo-initiation reaction.

The monomer diffusion coefficient \( D_m \):

\[
D_m = D \cdot \exp \left[ -s \left( 1 - \frac{M}{M_0} \right) \right],
\]

(2)

where \( s \) – proportionality coefficient, \( M_0 \) – initial monomer concentration.

The solution (2) has a phenomenological character but this enables to take into consideration contribution of temporal changing \( D_m \).

Temporal changing \( M \) in some point is occurred in the result of polymerization process and monomer diffusion. Taking into account the both processes which change \( n \), we will have:

\[
\frac{\partial n_1}{\partial \tau} = \Delta n_p K_g K_b^{-1/2} \left[ \frac{\alpha \beta K I(r)}{1/\tau_0 + \beta M} \cdot \frac{M^{3/2}}{M_0} + \Delta n_l \text{div}(D_m \text{grad} \frac{M}{M_0}) \right].
\]

(3)

The system of equations (1)-(3) is the base for description of hologram recording in photopolymeric media.

Assume that the medium has the absorption coefficient \( \alpha \) and the record is realized by the plane waves with the equal amplitudes \( A_0 \) and \( A_1 \). Then the interference pattern of light field inside the polymer plate is:

\[
I(y) = I_0 \exp \left[ -\alpha y \right] \left( K \cdot y \right).
\]

(4)

where \( I_0 = A_0^2 + A_1^2 \) - the light field intensity, \( 0 \leq y \leq d \), \( K = |k_p - k_i| \) – module of grating vector.

The record of grating is occurred in accordance with light distribution (4). The diffraction of light beams on the grating results in change of this distribution. But we will assume, the diffraction efficiency is small and the change of distribution (4) is negligible. So description of the record process of holographic grating in absorbent photopolymeric media will be solved in approximation of set light field.

The technique of the solving the equations for description of dynamics of recording one hologram (1),(3) is presented in 1. There it was assumed the monomer diffusion time on the length of order \( l \approx 1/\alpha \) is more than record time. That is why the diffusion on this length was neglected. But the diffusion on the grating period was taken into account.

In 1 final relations for the first harmonic of monomer concentration \( M_{1f}(\tau, y) \) and refraction index \( n_{1f}(\tau, y) \) which was taken as the base for development of the mathematical model of the dynamics of superimposed holograms recording.

Assuming that readout of hologram is realized during record at wavelength on which the polymer dye does not interact with readout radiation. Therefore the optical attenuation for the readout wavelength is negligible. In this case the kinetics of diffraction efficiency can be describe with the following equation in according to Bragg conditions of readout:

\[
\eta_d(\tau) = \sin^2 \left[ \frac{d}{G_1} \int_0^d n_1(\tau, y)dy \right],
\]

(5)

where \( G_1 = \alpha (2c \cos \varphi) \); \( \varphi \) – angle between \( y \) axis and the vector \( k_i \) inside polymer layer of thickness \( d \). It is necessary to note that the absorption has been taken into consideration in distribution of \( n_1 \).

In 3 influence of the model parameters on dynamics of holographic grating record was researched. There the Eq. (5) was used for it.

It was shown in 3 that during record the dynamics of diffraction efficiency sufficiently different at various optical absorption of material. Its character depends on parameter \( b \) (ratio of polymerization time to diffusion time): \( b = T_p / T_m \).
4. ANALITICAL MODEL OF RECORD OF SUPERIMPOSED HOLOGRAMS

We shall consider the model consequent record of superimposed holograms by the way of the angular multiplexing. Each hologram is recorded under its angle $\Psi$ (Fig.3a) with the same convergence angle of laser beams $\theta$. The vector diagram of superimposed holograms record is shown in Fig.3b.

As a result, in photopolymeric material several phase grating are records with their own vector grating $K_1, K_2, \ldots, K_n$, forming with axis $z$ accordingly corners $\Psi_1, \Psi_2, \ldots, \Psi_n$.

In practice, for optical communication devices, all recorded holograms grating must have equal diffraction efficiency.

In this case the simulation technique of superimposed holograms record is the following: the first hologram (curve 1, Fig.4a) is recorded under angle $\Psi_1$ and at initial monomer concentration $M_{n1} = M_{n0}$, where $M_{n0}$ – initial monomer concentration before record. During record the monomer concentration decreases due to polymerization reaction. At achieving set diffraction efficiency $\eta_0$ record is stopped and record time is determined $t_1$ (Fig.4a) and the current monomer concentration $M_{n2} < M_{n1}$ is determined as well as. Then the second hologram (curve 2) is recorded under angle $\Psi_2$ but initial monomer concentration equals $M_{n2}$. At achieving set diffraction efficiency $\eta_0$ record is stopped.
and record time is determined $t_2$ and the current monomer concentration $M_{n3} < M_{n2}$ is determined as well as. Then the process is repeated needed times.

In Fig.4a, the dotted line show development of the dynamic diffraction efficiency of the hologram during time, for the case if record was not interrupted in time which corresponds to set diffraction efficiency.

On the base of record times determined, the dependence of diffraction beam intensity on relative Bragg mismatch from which the integral diffraction characteristics is determined (Fig.4b).

For mathematical description of dynamics of sequentially exposed superimposed holograms we will receive the Eqs. (1)-(3) as the base. Since record is sequentially that in the model (1)-(3) for each hologram the change of initial monomer concentration must be taken into account.

In models takes into account change of optical absorption of photopolymeric material during recording holograms as well as. Experimentally it was shown, that change of the optical absorption during record can lead to change of integral diffraction features of recorded hologram grating. In $^2$ approximating function taking into account change of the optical absorption photopolymeric material during record was received:

$$a(t) = \alpha_2 - \alpha_1 \cdot \ln\left[\frac{t}{T_\alpha} + 1\right],$$

where $\alpha_1, \alpha_2, T_\alpha$—coefficients of the approximation function.

To take into account change of the optical absorption in record process of the superimposed holograms, it was made the following assumption. During record of one grating the optical absorption is constant, but for each grating value of absorption is different. I.e. the first grating is recorded with constant optical absorption $\alpha_1$, the second grating with absorption $\alpha_2 < \alpha_1$ and etc, as shown in Fig.5.

![Fig.5](image)

Write the expressions for the first harmonic of the concentration of the monomer $M_i(\tau, y)$ and refraction index $n_i(\tau, y)$ for $i$-th hologram, with taking into account of the depletion of the monomer and change of the optical absorption:

$$M_{ii}(\tau, y) = -\frac{8 \cdot M_{ii} \cdot \exp\left[-\frac{\alpha_i}{y^2}\int_0^\tau [\exp(A_0(\tau', y) - M_{0i}(\tau', y))]^{3/2} d\tau'\right]}{\exp\left[\int_0^\tau A_0(\tau', y) \cdot 3\pi T_{pi} \cdot M_{ni}^{1/2} d\tau'\right]},$$

where $A_0(\tau, y) = K_p^2 \int_0^\tau D_{pi}(\tau', y) d\tau' + \frac{28}{\sqrt{M_{mi} \cdot 5\pi T_{pi}}} \cdot \exp\left[-\frac{\alpha_i}{y^2}\int_0^\tau [M_{0i}(\tau', y)]^{1/2} d\tau'\right].$
amplitude of zero harmonic: \[ M_0(y) = M_m \left[ 1 + \frac{2\tau}{\pi} \exp \left( -\frac{\alpha_y}{2} \right) \right]^2, \]

\[ \tau = \frac{T_{pi}}{T_{pi}^{-1}} = \frac{\sqrt{\alpha \beta_0(K)} M_{ni} I_0}{\sqrt{2K_b} \cdot K}, \]

\[ M_{ni} \] - initial monomer concentration at start moment of record of \( i \)-th hologram \( (M_{0i-1}(t_{i-1}, y = 0)) \);

\( T_{pi} \) - polymerization time at start moment of record of \( i \)-th hologram;

The first harmonic of refraction index \( n_{i1}(t, y) \) of \( i \)-th hologram is:

\[
\int_0^\infty \int_0^{\infty} \cdot \cdot + \frac{8}{3\pi T_{pi}} \left( \frac{M_{0i}(\tau, y)}{M_{ni}} \right)^{3/2} \cdot \cdot \cdot \cdot \cdot 5. \]

\[
\cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \n\]
Dependencies diffraction efficiency from number of the hologram are shown in Fig.6, for different optical absorption for two cases: a) \( b<1 \) and b) \( b>1 \). As it is seen from the graph, the more optical absorption, the more diffraction efficiency of the holograms. But this behavior occurs only at the record of the first several holograms.

This is explained with the fact that the polymerization time \( T_p \) increases during record that leads to stretching of the dependencies diffraction efficiency on time of its record. In this connection, at small times of record, features will differ not strongly. The differences will be essential only at greater times. 

At \( b>1 \) (Fig.7b), diffraction efficiency of all holograms increases. Since process radical polymerization occurs slowly, but diffusion of the monomer from dark areas in light is significant. As effect, growth of grating occurs slowly and reaches greater diffraction efficiency, than in the case at \( b<1 \) (Fig.7a), since diffusion processes leads to increase of the monomer concentrations in light area.

Dependencies diffraction efficiency from number of the hologram, at various parameter \( n_i \) are presented in Fig.8, and at various parameter \( n_p \) in Fig.9, for two cases: a) \( b<1 \) and b) \( b>1 \).

Increase of contribution into growth of grating for count of displacement of inert components and the polymerization reactions, leads to increase diffraction efficiency. But in the first case, this is observed close to completion of record, but in the second case - close to the begin of record. This is connected with the following: at the begin of record, the growth
of grating occurs as the result of polymerization basically, (see to explanations to Fig.2). At outflow of time, diffusion of the monomer all more checks the growth of a grating.

Dependence of diffraction efficiency from relative Bragg mismatch \( \Delta \) is shown in Fig.10a. Calculation was conducted for the case when initial absorption was 4 Nep and final - 0.1 Nep, for 6 superimposed holograms (\( \delta n_p = 10^{-2} \), \( \delta n_i = 10^{-2} \), \( s=1 \), \( d=40 \) мкм, \( \theta=20^\circ \), \( \Psi=2^\circ \)), at condition of equality of diffraction efficiency.

On base of normalized dependencies of diffraction efficiency of recorded grating on relative Bragg mismatch \( \eta_d(\Delta K) \), we shall define the following features (see to indications on Fig.10b): width of the main lobe at level -3\( \eta \) \( 2 \cdot \Delta K_{0.5} \) (for \( 2 \cdot \Delta K_{0.5} = 5.566 \) uniform profile) and the amplitude of the first side lobe \( \eta_{d\_SL} \) dB (\( \eta_{d\_SL} = -13.26 \) dB for uniform profile).

The dependences of diffraction efficiency on record time (\( \delta n_p = 10^{-2} \), \( \delta n_i = 10^{-2} \), \( s=1 \) \( d=40 \) мкм, \( \theta=20^\circ \), \( \Psi=2^\circ \)) with account of the change the optical absorption (a) and without this account (b) are presented in Fig.11. The dynamics of diffraction efficiency was calculated for 6-th superimposed holograms (the first hologram - an upper curve, the last – lower one). All grating have the same diffraction efficiency. It is Seen that at account of the change the optical absorption, times of record required for achievement given diffraction efficiency, greatly decrease.
Dependencies of the width of the main lobe at level -3 dB is presented in Fig.12 (a) and amplitude of the first side lobe (b) on number of hologram (1- without account of the change of optical absorption, 2 - with account of the change of optical absorption).

At presence of absorption, the amplitude of the first side lobe becomes less than in the case without absorption. And the less value of the absorption, the less width of the main lobe .

From Fig.11 and Fig.12 it is seen that account of change of optical absorption in model of serial record of superimposed holograms in photopolymeric material is necessary.

For determination degree of adequacy we carried out comparison of theoretical and experimental data. The experimental dependences were received in NIOCH SD RAS for two samples, when recording 10 and 16 superimposed holograms grating for the following condition record: a) \( \theta=20^\circ, \delta \Psi=7^\circ, t_{rec}=2\text{sec}; d=40 \mu\text{m} \); b) \( \theta=10^\circ, \delta \Psi=7^\circ, t_{rec}=1.25\text{sec}, d=100 \mu\text{m} \). Serial record was realized at equal times of the exposures.
Experimental and theoretical curves of dependences of diffraction efficiency on number of hologram are shown in Fig.13. Fitting of theoretical and experimental data was realized with the help of selection material parameter of photopolymer: (a) $\delta n_p=10^{-2}$, $\delta n_i=10^{-2}$, $b=10$, $\alpha=1.3$ Nep; (b) $\delta n_p=10^{-3}$, $\delta n_i=0.7 \times 10^{-2}$, $b=10^{-2}$, $\alpha=3$ Nep)

It is seen that mathematical model well describes the experimental results (Fig13a) in the field of value of the parameter $b=\frac{T_p}{T_m} > 1$, characterizing ratio of polymerization time to monomer diffusion time, and qualitative matches with them (Fig.13b) for $b<1$.

6. CONCLUSION

In the work we consider the mathematical model of serial record of superimposed gratings by the way of angle multiplexing.

We present the procedure of calculation and optimization of the record process of the superimposed grating in photopolymer, with set diffraction characteristics. We present results of the numerical simulation of record of 6 superimposed diffraction grating with equal diffraction efficiency (10%) with initial optical absorption 4 Nep and final - 0.1 Nep. The Comparison its with the known experimental results has shown the satisfactory agreement.

The influence of material parameter of photopolymeric material on diffraction efficiency superimposed hologram grating was researched.

It was shown that change the optical absorption of photopolymeric material during record can lead to change of integral diffraction features recorded hologram grating.

The results of the numerical modeling have shown the good consent with experiment and allow to use the theoretical model for determination of the optimum times of record of the superimposed holograms.

REFERENCES

