

Modulation of the characteristics of complex holographic gratings under an additional laser pulse

M.G. Kucherenko, A.P. Rusinov, D.S. Fedorov

Abstract. Recording of elementary holographic gratings in polymer films and solutions coloured with organic dyes has been investigated. Possible mechanisms of modifying a recorded grating by an additional laser pulse are considered. A theoretical model is proposed to describe the processes of recording/relaxation and modification of gratings recorded on triplet states of photochrome molecules; the predictions of this model are found to be in good agreement with the experimental data.

Keywords: triplet holographic grating, amplitude–phase relief, modulation of diffraction efficiency.

1. Introduction

Holographic methods are promising for studying processes occurring in condensed media. They can be used to investigate not only optical but also thermal, structural, and other characteristics of such systems. An interesting possibility is holographic study of molecular processes, such as the dynamics of motion of macromolecules in polymer systems, diffusion of photochrome molecules in a matrix, kinetics of radiative and nonradiative photoprocesses in condensed media, etc. [1–4].

In the latter case an observed signal can be formed using a system in which the laser effect is initiated by a holographic grating recorded on excited metastable (triplet) states of photochrome (referred to as a triplet grating below). The advantages of this method are the integral character of signal (when weak, spatially distributed processes are coherently amplified), high lifetime of gratings (due to which the kinetics of molecular processes can be observed for rather long times), etc. [5].

This study showed that the informativeness of the method proposed can be significantly increased by exposing recorded gratings to an additional laser pulse with a spatially uniform intensity distribution. This approach allows one to analyse laser-induced processes in an object through their influence on the characteristics of elementary gratings not only when the holographic structure relaxes but also when it is modified by an additional pulse. The latter is of interest for such practical applications as the development of laser methods to control grating characteristics.

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We experimentally investigated the processes of recording and modifying elementary holographic gratings in polyvinyl alcohol (PVA) polymer films, coloured with organic dyes, and performed theoretical analysis of these processes based on a simple mathematical model of a binary grating containing a surface (relief–phase) and volume (amplitude–phase) components, shifted in space by some distance Δx .

2. Experimental

Elementary gratings were recorded by pulses of a solid-state YAG: Nd³⁺ laser (LQ 529B, $\lambda = 532$ nm, $\tau = 12$ ns, $W_{\text{pulse}} = 10$ mJ) (Fig. 1). The beam convergence angle was varied in the range of 4°–8°, and the interference field period was 4–6 μ m. The holograms were read by a He–Ne laser beam ($\lambda = 632.8$ nm, $P = 50$ mW) in the first-order ($m = 1$) diffraction maximum. Additional irradiation of the system in the activated state was performed by another solid-state YAG: Nd³⁺ laser (LQ 125, $\lambda = 532$ nm, $\tau = 10$ ns, $W_{\text{pulse}} = 5$ mJ). A synchroniser was used to shift the additional laser pulse respect to the recording pulse by an arbitrary time in the range of 1 μ s–10 ms, with an error of 0.1 μ s. The instability of the synchroniser time characteristics did not exceed 10 ns.

The pump beams, which formed the interference recording field, and the additional laser beam were focused in the sample plane so as to provide their diameters of about 1 mm. The reading laser beam passed through the centre of the initiation region and was focused into a spot 0.3–0.5 mm in diameter. In this case, one can neglect the radial (Gaussian) intensity distributions of the recording and modifying light fields and assume them to be uniform within the reading beam.

The emphasis of this study is on the characteristics of triplet gratings. In this case, the main observable quantity is the time-dependent diffraction signal from the recorded structure: $\eta(t)$. A stationary grating was formed in a sample exposed to several (5–10) pump pulses to give rise to a stationary diffraction signal η_{st} , which exceeded several times the time-dependent one. The time-dependent diffraction signal, which is caused by recording a triplet grating, did not change much; however, to record it with a high accuracy, it was necessary to exclude the stationary diffraction component η_{st} . In this case, we recorded the difference signal $\Delta\eta(t) = \eta(t) - \eta_{\text{st}}$, which became negative at $\eta(t) < \eta_{\text{st}}$ (this particular remark is important for further consideration). The diffraction efficiency (DE) of stationary gratings was measured in their first-order diffraction maximum as a ratio of the probe laser beam intensity in the first-order maximum to the total intensity of the beams in all diffraction orders.

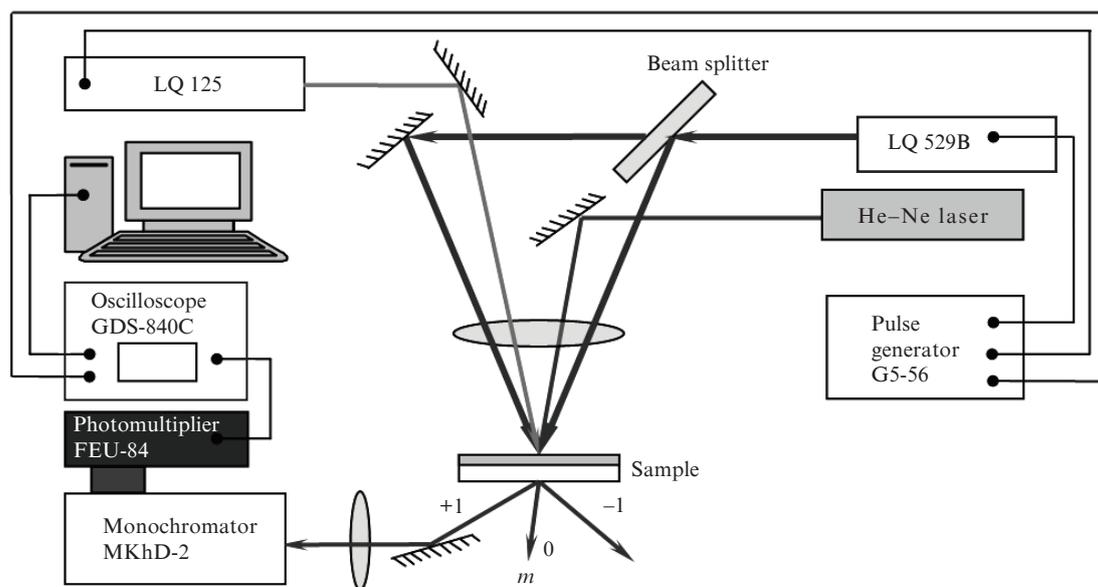


Figure 1. Schematic of the experimental setup.

The results of these experiments showed that, depending on the intensity of recording beams, one can select the following three modes of grating formation. (i) Destruction of the polymer film (polymer burning or/and ablation) initiates an amplitude-phase grating with a relatively low DE (less than 1%). This process occurs at pump intensities exceeding 10^8 W cm^{-2} and dye concentrations in the polymer higher than $10^{-2} \text{ mol L}^{-1}$. (ii) At pump intensities from 10^7 to 10^8 W cm^{-2} and dye concentrations of $(1-10) \times 10^{-3} \text{ mol L}^{-1}$ a relief thermoplastic grating is formed on the polymer film surface. This relief arises due to the nonuniform heating of the film by the light field, which generally leads to the formation of a phase grating with a fairly high DE (5%–10%). (iii) At pump intensities below 10^7 W cm^{-2} and dye concentrations $(1-5) \times 10^{-4} \text{ mol L}^{-1}$, a triplet grating is recorded against the stationary-grating background; this triplet structure serves as an amplitude grating when reading at a wavelength of 632 nm [5].

In the latter case the triplet character of the grating is confirmed by the complete reversibility of the process, exponential decay of the diffraction signal, and the proximity of the grating decay time to the lifetime τ_T of the dye triplet state (Fig. 2).

The additional laser irradiation led to significant changes in the time dependences of the diffraction signal. We observed basically different responses of the system to the primary and additional laser irradiations, depending on the total number of activating pulses. An important characteristic of this process is the total exposure time in the recording stage.

The character of the diffraction response of the grating in the sample exposed to an additional laser pulse ('2-pulse') was determined by the presence or absence of a stationary periodic structure in it. Prior to its formation the primary laser pulse induced a triplet grating characterised by an exponential decay (Fig. 2). The additional pulsed irradiation led to a sharp decrease in the diffraction signal (Fig. 3), which we believe to be caused by two mechanisms: (i) decrease in the population of T_1 states of dye molecules as a result of quasi-irreversible absorption $T_1 \rightarrow T_n$ during the additional irradiation and (ii) decrease in the diffraction efficiency of a triplet grating due to the reduced modulation depth of the number

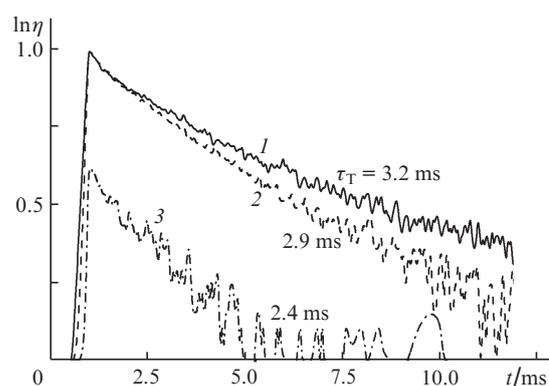


Figure 2. Kinetics of diffraction signals from triplet gratings into the first-order maximum at different lifetimes of gratings formed in PVA films coloured with (1) eosin G and (2) eosin H and (3) in a gelatine film coloured with eosin G.

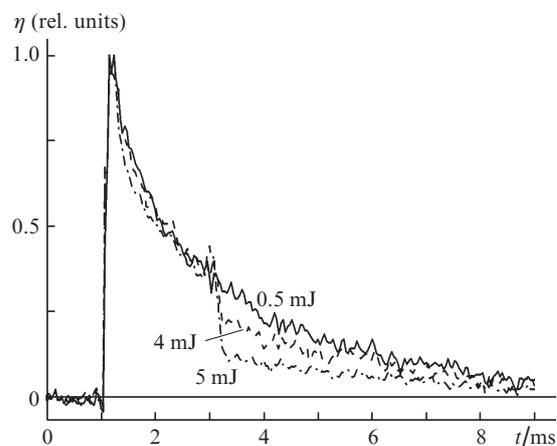


Figure 3. Diffraction signal from a triplet grating in a PVA film coloured with eosin G at different energies of additional laser irradiation at the instant $t = 2 \text{ ms}$.

of activated centres with a general increase in the population of T states.

The first mechanism of the observed ‘dips’, induced by the additional pump pulse, was discussed in [6, 7]. The only difference is that the delayed-fluorescence and phosphorescence signals from polymer films coloured with an organic dye were recorded as observables in [6, 7], where a step decrease in the luminescence response under additional laser irradiation was also observed. However, the holographic technique for detecting ‘dips’ induced by the laser 2-pulse turned out to be much more sensitive than the luminescence method. This is explained by the advantages of measurement of light signals satisfying the conditions of high spectral selectivity (recording and activation at the wavelength $\lambda = 532$ nm and reading the grating at $\lambda = 632$ nm) and spatial orientation of the recorded light flux. In our opinion, specifically this mechanism plays a key role in the sharp decrease in the triplet-hologram diffraction efficiency.

However, the ‘dips’ in the diffraction curves may have another nature, related to the decrease in the modulation depth of the triplet grating by the additional pulse. Since the triplet grating at the wavelength $\lambda = 632$ nm is an amplitude one, a sufficiently large amplitude of diffraction signal can be obtained only on a strongly modulated grating. In this case, the triplet-level population in the maxima of recording interference field is close to maximum; i.e., almost all photochrome molecules in this region are in the triplet state. The additional activation of the system by the 2-pulse (homogeneous over the entire illuminated area) causes a spatially inhomogeneous increase in population in the film. In particular, this increase is minimal in the maxima of the triplet grating and maximal in the ‘dark’ regions. As a result, the difference in the T populations in the illuminated and ‘dark’ regions is levelled out and the modulation depth of the amplitude grating and, correspondingly, the diffraction response signal are reduced.

During the formation of a stationary periodic structure with a stationary diffraction signal component η_{st} (after 10–20 pulses with an energy ~ 10 mJ) (Fig. 4), the diffraction signal from the time-dependent grating radically changes. After the exposure of the recorded structure to an additional pump pulse, the diffraction signal $\eta(t)$ becomes smaller than η_{st} , and their difference $\Delta\eta(t) = \eta(t) - \eta_{st}$ is described by a

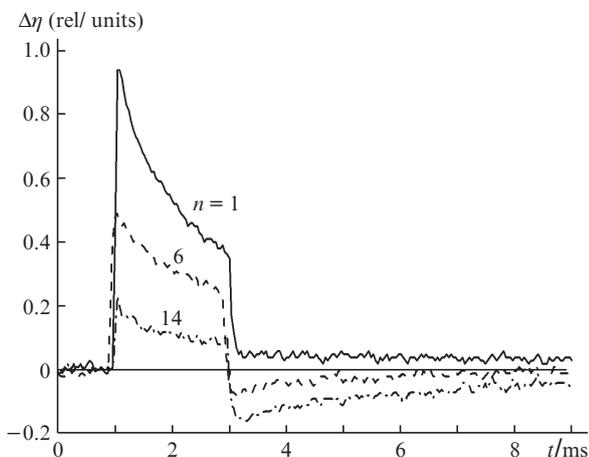


Figure 4. Change in the diffraction signal from a complex grating in a PVA film coloured with eosin G under double-pulse irradiation, depending on the number of recording pulses n .

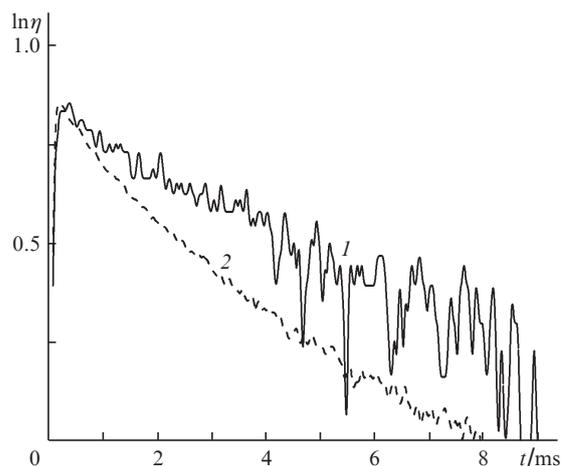


Figure 5. Dynamics of (1) recovery of the stationary grating and (2) damping of a triplet grating (the characteristic times are 4.7 and 3.15 ms, respectively).

curve that drops to negative values and then increases to zero. The observed diffraction-signal inversion can be related to the temporal decrease in the modulation depth of some optical characteristic of the grating and its subsequent recovery with a characteristic time of 4–5 ms (Figs 4, 5).

Detection of diffraction response signals from the gratings under consideration in the reflection geometry suggests that the stationary grating is recorded mainly due to the formation of a spatial relief on the polymer film surface. The effect of reversible inversion of the triplet grating diffraction signal by an additional pulse is absent in the reflection regime. Hence, one can conclude that it is related to the processes occurring specifically in the polymer bulk.

3. Model of light diffraction from a complex structure composed of a stationary and relaxing holographic gratings

We described the effects observed based on the model of complex gratings (Fig. 6). For example, the m th-order time-dependent DE $\eta_m(t)$ of a transparent phase grating with a period Λ can be written as

$$\eta_m(t) = \frac{1}{\Lambda^2} \left\{ \int_{-\Lambda/2}^{\Lambda/2} \exp \left[-i\Phi(x, t) - im \frac{2\pi x}{\Lambda} \right] dx \right\}^2; \quad (1)$$

note that at a small phase shift ($\Phi(x, t) \ll 1$) $\exp[i\Phi(x, t)] \approx 1 + i\Phi(x, t)$. The total phase shift $\Phi(x, t)$ in the case of time-dependent spatial modulation of the refractive index, $n_0 + \Delta n(t) \cos(Kx - \delta_0)$, of a coloured film of thickness l and a stationary surface relief $\Delta \cos Kx$ contains the corresponding contributions $\Delta\varphi_{ph}(x, t)$ and $\Delta\varphi_{rel}(x, t)$:

$$\begin{aligned} \Phi(x, t) &= \Delta\varphi_{ph}(x, t) + \Delta\varphi_{rel}(x, t) = \Delta n(t) k l \cos(Kx - \delta_0) \\ &\quad - (n_0 - 1) k \Delta \cos Kx. \end{aligned} \quad (2)$$

Here, $\Delta n(t) = \Delta n_0 \exp(-t/\tau_T)$ is the relaxing modulation depth of the film refractive index n_0 ; $K = 2\pi/\Lambda$; $k = 2\pi/\lambda$; and $\delta_0 = K\Delta x$ is the phase shift of the induced and stationary gratings.

The phase relations between the gratings are determined at the recording stage; they depend both on the sign of Δn_0 for

the volume phase grating (depending on the choice of dye, polymer, and reading laser wavelength, the sample refractive index in the maximum of illuminance may either increase or decrease) and on the sign of Δl of the relief grating (i.e., if the maxima of the interference pattern correspond to the maximum relief in the case of polymer expansion or to the minimum relief when the polymer is ablated). Obviously, in this situation the gratings can be shifted by $\delta_0 = 0$ or π , which corresponds to their in-phase or out-of-phase recording. Then $\Phi_0(x, t) = [(1 - n_0)\Delta l \pm \Delta n(t)l] \cos Kx$ and, based on expression (1) for the diffraction efficiency $\eta_m(t)$ of a complex grating, we have

$$\eta_m(t) = J_m^2\{k[(1 - n_0)\Delta l \pm \Delta n(t)l]\}, \quad (3)$$

where $J_m^2(z)$ a squared m th-order Bessel function.

However, along with the above-considered phase shift at the recording stage, there may be other mechanisms of grating shifting. The reason is the asymmetric arrangement of the pump beams with respect to the normal to the film surface and a deviation of the additional pump beam or the reading laser beam from the normal to the surface [3, 4]. In the latter case the grating shift is most pronounced.

Let us consider the reading beam transmission through a complex relief-phase grating with modulated relief, $\Delta l(x) = \Delta l_0 \cos(2\pi x/\Lambda)$, and refractive index, $\Delta n(x) = \Delta n_0 \cos(2\pi x/\Lambda)$, at a small angle α to the normal (Fig. 6). The phase shift on a relief grating with a relief height of several tens of nanometres, is independent of the angle α and can be written as $\Delta\varphi_{\text{rel}}(x) = (n_0 - 1)k\Delta l(x)$. For a volume phase grating the situation is different: when the reading beam deviates from the normal, the grating refractive index changes along the reading beam propagation direction; therefore, the expression for calculating the phase shift $\Delta\varphi_{\text{ph}}(x) = kl\Delta n(x)$ is invalid. In this case, to determine the effective phase shift, one must integrate over the reading beam trajectory:

$$\Delta\varphi_{\text{ph}}(x, \alpha) = k \int_0^l \Delta n(x + x') ds = k \int_0^l \Delta n(x + z \tan \alpha) \frac{dz}{\cos \alpha}.$$

Having substituted $\Delta n(x)$ into this expression and performed the corresponding transformations, we obtain

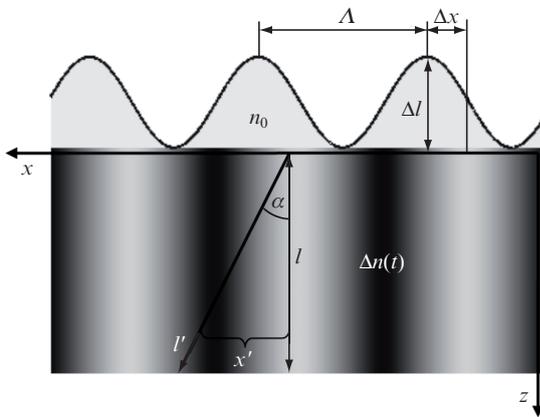


Figure 6. Complex relief-phase grating: Δl is the modulation depth of the relief grating profile; $\Delta n(t)$ is the relaxing modulation depth of the refractive index of the film n_0 ; Λ and Δx are, respectively, the period and spatial shift of the surface and volume gratings; and l' is the reading beam direction at an angle α to the normal of the film surface.

$$\begin{aligned} \Delta\varphi_{\text{ph}}(x, \alpha) &= \frac{kn_0}{2\pi \sin \alpha} \left\{ \sin \left[\frac{2\pi}{\Lambda} (x + l \tan \alpha) \right] - \sin \frac{2\pi}{\Lambda} x \right\} \\ &= \frac{kn_0 \sin \delta}{2\pi \sin \alpha} \cos \left(\frac{2\pi}{\Lambda} x + \delta \right), \end{aligned}$$

where $\delta = \pi l \tan \alpha / \Lambda \approx \pi \alpha l / \Lambda$ is the shift of the effective volume phase grating with respect to the initial one and, correspondingly, with respect to the relief. Thus, the phase shift is determined not only by the deviation angle of the reading beam from the normal but also by the l/Λ ratio. In our study the ratio of the film thickness to the recorded-structure period was varied within 10–15; therefore, small angles α corresponded to significant grating shifts.

It can be shown that additional laser irradiation of a recorded complex structure changes the relative shift of gratings, along with their other parameters. In our opinion, this change has a thermal nature: uniform heating of the polymer matrix reduces its refractive index and, accordingly, increases the reading angle α and the film thickness l . In this case, the refractive index modulation acquires an additional dependence on time:

$$n(t) = n_0 + \Delta n(t) \cos(Kx - \delta_0(t)).$$

We assume that the temporal dynamics of the grating shift is described by an exponential law, which is characteristic of both thermal relaxation processes and viscoelastic motions of polymer:

$$\delta(t) = \delta_0 \exp(-t/t_r),$$

where δ_0 is the amplitude of the grating shift initiated by the 2-pulse and t_r is the characteristic relaxation time.

When an induced grating is formed as a result of modulation of the film absorption coefficient, the field amplitude $E_1(\varphi, t)$ in the Fraunhofer diffraction pattern, when a wave passes through a grating region with a length of one period Λ , is the Fourier transform of the amplitude transmittance $\tau(x, t)$ of the film:

$$E_1(\varphi, t) \sim \int_{-\Lambda/2}^{\Lambda/2} \tau(x, t) \exp[-i\Phi(x, t) - ikx \sin \varphi] dx, \quad (4)$$

$$\tau(x, t) = \exp[-\varepsilon_T N_T(x, t)l].$$

Here, as previously, k is the reading radiation wave number. With allowance for the low intensity of the probe beam, the absorption can be considered as Bouguer. Then the diffracted beam at the hologram output should carry information both about the relaxing population profile $N_T(x, t)$ of the dye T level [through the transmittance $\tau(x, t)$ and refractive index modulation] and about the shift due to the formation of the film surface relief. The parameter l in (4) is the absorbing-layer thickness and ε_T is the extinction coefficient of the $T_1 \rightarrow T_n$ electronic transition.

With allowance for (4), the m th-order diffraction efficiency of a combined (phase-amplitude-relief) lattice structure can be written as

$$\begin{aligned} \eta_m^{(c)}(t) &= \frac{1}{\Lambda^2} \\ &\times \left\{ \int_{-\Lambda/2}^{\Lambda/2} \exp \left[-\varepsilon_T N_T(x, t)l - i\Phi(x, t) - im \frac{2\pi x}{\Lambda} \right] dx \right\}^2. \quad (5) \end{aligned}$$

Expressions (1)–(3) and (5) were used to calculate the diffraction efficiencies of complex lattice structures formed in a polymer sample. A numerical analysis of the diffraction efficiencies of different combined holograms consisting of gratings of two and three types was performed. The results obtained show that the proposed model is in good agreement with the experimental data.

4. Influence of additional laser irradiation on amplitude (triplet) holographic gratings

Formation and decay of a time-dependent amplitude grating is determined by the kinetics of the average volume concentration of relatively long-lived states of photosensitive centres. The level populations can be described within the balance scheme, where three active levels of dye with the energies E_0 , E_S , and E_T are selected; these levels correspond to the states 0 (the ground state), S (the first excited singlet), and T (the lower-energy triplet). This model was described in detail in [5].

Let us represent the light affecting the system in the form of rectangular pump pulses with a constant intensity and width t_0 . Since an elementary grating is recorded by the interference field of laser beams, the spatial profile of the pump field can be written as

$$I(x) = I_{01}[1 + a\cos(2\pi x/\Lambda)],$$

where I_{01} is the pump intensity, a is the interference-pattern contrast, $\Lambda = \lambda/[2\sin(\vartheta/2)] \approx \lambda/\vartheta$ is the spatial period of the interference pattern, ϑ is the beam convergence angle, and λ is the pump wavelength.

Let us denote the absorption cross section of the $0 \rightarrow S$ transition as σ , the spontaneous deexcitation rates of S and T levels as τ_S^{-1} and τ_T^{-1} , and the $S \rightarrow T$ intersystem crossing rate as K_{ST} , we can write the following system of balance equations for the populations:

$$\begin{aligned} \frac{\partial N_0}{\partial t} &= -\sigma I(x,t)N_0(x,t) \\ &+ [\sigma I(x,t) + \tau_S^{-1}]N_S(x,t) + \tau_T^{-1}N_T(x,t), \\ \frac{\partial N_S}{\partial t} &= \sigma I(x,t)N_0(x,t) - [\sigma I(x,t) + \tau_S^{-1} + K_{ST}]N_S(x,t), \\ \frac{\partial N_T}{\partial t} &= K_{ST}N_S(x,t) - \tau_T^{-1}N_T(x,t). \end{aligned}$$

Here, N_0 , N_S and N_T are the populations of the 0, S, and T levels.

The condition of conservation of the number of photochrome molecules has the form

$$N_{\text{all}} = N_0 + N_S + N_T.$$

The above system can be solved within the quasi-stationary mode of populations in the subsystem of S levels (see [5]). The total quantum yield of dye in the triplet state is determined as

$$\Phi_T(x) = \frac{K_{ST}}{\tau_S^{-1} + K_{ST} + 2\sigma I(x)}.$$

The expression for the diffraction efficiency of a thin amplitude triplet grating can be written as

$$\eta_T(m) = \frac{1}{\Lambda^2} \left\{ \int_{-\Lambda/2}^{\Lambda/2} \exp[-\varepsilon_T N_T(x')l] \exp\left(-\frac{2\pi i}{\Lambda} x' m\right) dx' \right\}^2.$$

Here, Λ is the spatial period and $N_T(x')$ is the grating concentration profile.

The additional pump radiation will induce transitions from T_1 to T_n with some rate $\sigma_{TT} I_{\text{add}}(t)$, where $I_{\text{add}}(t)$ is the additional-pulse intensity. Obviously, some part of T_n states relax back to T_1 , either directly or through the S_1 state, and only some part of photoactive centres that underwent the $T_n \rightarrow S_1 \rightarrow S_0$ transition will correspond to the additional depletion of the triplet level. However, in the general case, taking into account that the relaxation transitions rates between levels are sufficiently large, one can introduce an effective induced deexcitation rate for the T_1 level: $\sigma_{TT}^* I_{\text{add}}(t)$. Then, the solution of the balance kinetic equations for the populations in the system of three-level photoactive centres with allowance for this term yields the following expression for the triplet-level population [5]:

$$\begin{aligned} N_T(x,t) &= N_{\text{all}} \left\{ 1 - \exp\left[-\sigma \int_0^t \Phi_T(x,t') I(x,t') dt'\right] \right\} \\ &\times \exp\left[-\frac{1}{\tau_T} t - \sigma_{TT}^* \int_0^t I_{\text{add}}(\tau) d\tau\right]. \end{aligned}$$

Modelling for the additional rectangular pulse

$$I_{\text{add}}(t) = I_{02}[\theta(t - \tau_{12}) - \theta(t - \tau_{12} - \Delta)]$$

$[\theta(t)$ is the Heaviside function, τ_{12} is the delay of the additional pulse with respect to the recording one, and Δ is the 2-pulse width] shows very good agreement with the experiment (Figs 7, 8).

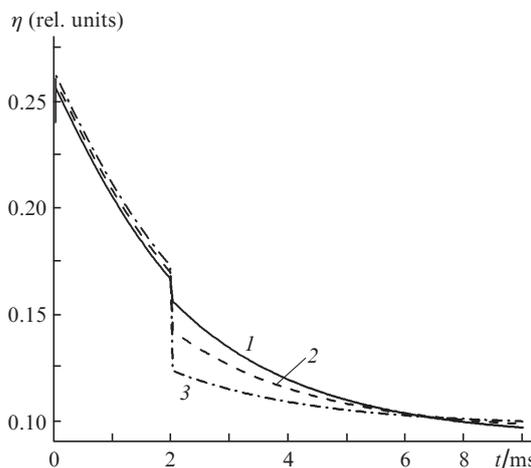


Figure 7. Kinetics of the diffraction signal from a triplet grating in a PVA film coloured with eosin G, at additional-pulse intensities $I_{02} = (1) 10^6$, $(2) 4 \times 10^6$, and $(3) 10^7 \text{ W cm}^{-2}$; $I_{01} = 2 \times 10^7 \text{ W cm}^{-2}$.

5. Conclusions

We investigated the mechanisms of recording and relaxation of simple holographic gratings in polyvinyl alcohol and gelatine films, coloured with organic dyes [eosin G (H)] and revealed the main recording regimes. A theoretical model was proposed to describe the diffraction from a complex ampli-

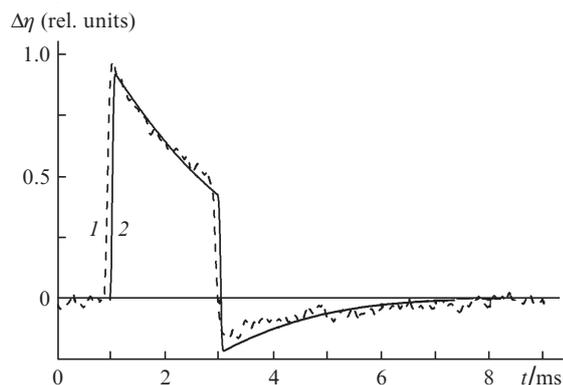


Figure 8. Kinetics of change in the diffraction signals from a complex grating in a PVA film coloured with eosin G, under additional exposure to a laser pulse: (1) experiment and (2) calculation

tude–phase–relief grating and a theoretical analysis of diffraction response signals was performed.

In addition, we carried out experiments on modulation of the recorded-grating parameters by an additional laser pulse and obtained the main regularities of this process. Within the proposed theoretical model we compared a series of experimental curves with the calculated ones. The results obtained make it possible to clarify the mechanisms and establish features of recording elementary holographic gratings in polymer films.

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