

Dynamics of two-wave mixing in a film of azocontaining photosensitive LC polymer

A N Simonov, M S Andreeva, V I Shmal'gauzen, V P Shibaev

Abstract. The dynamics of two-wave mixing in a film of azocontaining photosensitive polymer with liquid-crystal properties is studied experimentally and theoretically. A simplified theoretical model is proposed for the energy exchange between light beams in a polymer layer taking into account absorption, saturation, and diffusion in the polymer. The theoretical results obtained in the approximations of the chosen model are in good agreement with the experimental data. The dynamic characteristics of an interferometer based on two-wave mixing in a film of photosensitive polymer are studied and the possibility of adaptive compensation of slow phase fluctuations in one of its arms is demonstrated.

Keywords: azocontaining polymer, liquid crystal, two-wave mixing, adaptive interferometer.

1. Introduction

The application of photosensitive reversible polymeric materials in the dynamic holography and optical data processing is of considerable importance [1–4]. High values of nonlinear optical susceptibility of polymers containing chemically linked dye molecules make it possible to carry out efficient nonlinear transformations of light signals with low intensities ($I < 1 \text{ W cm}^{-2}$). The high sensitivity of photochromic polymers and their resistance to repeated light exposures combined with the possibility of controlling their dynamic parameters (with the help of external actions) make such media attractive for various applications [1, 4, 5].

Adaptive interferometry, which combines dynamic holography and adaptive optics, is a promising field for applications of photosensitive reversible organic materials [6–8]. The implementation of the dynamic holography methods using photosensitive reversible materials for mixing light beam makes it possible to considerably improve the parameters of interferometric systems and to simplify their construction [6–10]. In most cases, adaptive interferometers are made on photorefractive crystals (PRCs) with high

holographic parameters, but their production is costly and they require special operation conditions [9].

Apart from applied problems, adaptive interferometric systems can be also used for determining intrinsic characteristics of nonlinear optical materials. Experimental investigations of the dynamics of the system response upon multi-wave mixing provide information on the features of the nonlinearity mechanism of the material under study. As applied to organic compounds, such information is very important for determining the relation between the molecular structure of the medium and its resultant nonlinear-optical properties [11].

In this paper, we analyse theoretically the two-wave mixing in the films of azocontaining polymers with liquid-crystal (LC) properties. Examples of such materials are side-chain LC acrylic copolymers with cyanobiphenyl fragments in side groups [12–14]. The theoretical results are compared with the experimental data obtained using an adaptive interferometer.

2. Theoretical analysis

Like most photochromic organic compounds, the film samples of azocontaining LC polymers (ALCP) studied here belong to media with a local response to the external light exposure [15]. The energy transfer between coherent light beams without the frequency shift is possible only in nonstationary (transient) regimes [16]. Consider the features of transient processes in ALCP films.

Nonlinear optical properties of azocontaining polymer compounds are caused by reversible *trans*–*cis* photoisomerisation of fragments of azo dyes followed by molecular and structural rearrangements (Fig. 1b) [17]. Azo dyes may be in the form of *cis*- or *trans*-isomers (we do not consider here the intermediate electronic states). *Trans*-isomer is more stable than the *cis*-isomer: under standard conditions, the reverse *cis* → *trans* transition occurs spontaneously during a thermal reaction. The characteristic time T_r of thermal relaxation is determined by the temperature and chemical structure of the compound (the matrix type, etc). A polymer sample exposed to light undergoes *trans* → *cis* and *cis* → *trans* transitions. The transition rates depend on the effective absorption cross sections σ_i of isomers and on quantum yields γ_i of isomerisation processes. Hereafter, the subscript $i = 1$ corresponds to *trans*-isomers and $i = 2$ to *cis*-isomers of the azo dye. In the field of a light wave with the intensity I_0 , the rates of *trans* → *cis* and *cis* → *trans* transitions are equal to $a_i I_0$, where $a_i = \gamma_i \sigma_i / \hbar \omega$ are the rate constants and $\hbar \omega$ is the energy of a light quantum.

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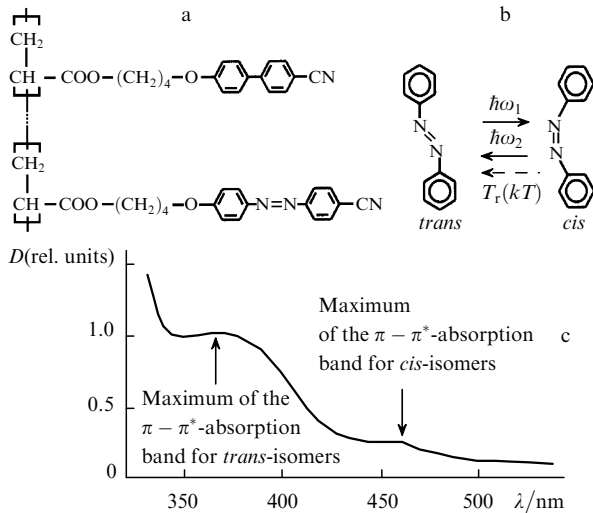


Figure 1. Chemical formula of the azocontaining LC polymer (a), scheme of configuration transformations in azobenzene (b), and spectrum of the linear absorption D of a polymer film ($\omega_{1,2}$ are the absorption frequencies for *cis*- and *trans*-isomers) (c).

Consider the energy transfer between light beams on an ALCP layer. The geometry of coupling is shown in Fig. 2a, where θ is the angle of convergence of the beams on the film; $E_{s,r}$, $k_{s,r}$ are the electric field strengths and the wave vectors of the signal and reference beams, respectively; L is the polymer film thickness. We assume that the wave fronts of interfering beams are plane and their polarisations are identical and perpendicular to the plane of incidence xz (s -polarised beams). The electric field strengths in light waves are connected with their complex amplitudes through the relations

$$E_s = \frac{A_s}{2} \exp(i\omega t - i\mathbf{k}_s \mathbf{r}) + \text{c. c.}, \quad (1)$$

$$E_r = \frac{A_r}{2} \exp(i\omega t + i\delta \sin \Omega t - i\mathbf{k}_r \mathbf{r}) + \text{c. c.},$$

where Ω and δ are the frequency and amplitude of the phase modulation of the reference beam. The intensities of the beams in these case are $I_{r,s} = |A_{r,s}|^2/2$. The light beams intersecting in the medium produce the interference pattern

$$I = W [1 + m \cos(Kx + \delta \sin \Omega t + \xi)], \quad (2)$$

where $W = I_r + I_s$ is the total intensity of light in the layer under study; $m = 2(I_r I_s)^{1/2}/(I_r + I_s)$ is the contrast of the interference pattern; $\xi = \arg A_r - \arg A_s$ is the phase shift (we assume that $\xi = 0$ for $z = 0$); $K = |\mathbf{k}_s - \mathbf{k}_r|$ is the modulus of the wave vector of the intensity grating. Note that I_s, I_r, m and ξ are functions of z in the general case.

In analogy with Refs [6, 18], the equation for the light-induced addition $\Delta n(z, t)$ to the refractive index in the chosen polymer layer can be represented in the form

$$\frac{\partial \Delta n}{\partial t} - D_x \frac{\partial^2 \Delta n}{\partial x^2} - (\Delta n_s - \Delta n) a_1 I + \Delta n \left(\frac{1}{T_r} + a_2 I \right) = 0. \quad (3)$$

Here, Δn_s is the phenomenologically introduced saturation; T_r is the characteristic time of thermal *cis* \rightarrow *trans* relaxation of isomers; D_x is the diffusion coefficient of *trans*-isomers of the dye along the vector of intensity grating. The value of Δn increases due to the direct *trans* \rightarrow *cis* photoisomerisa-

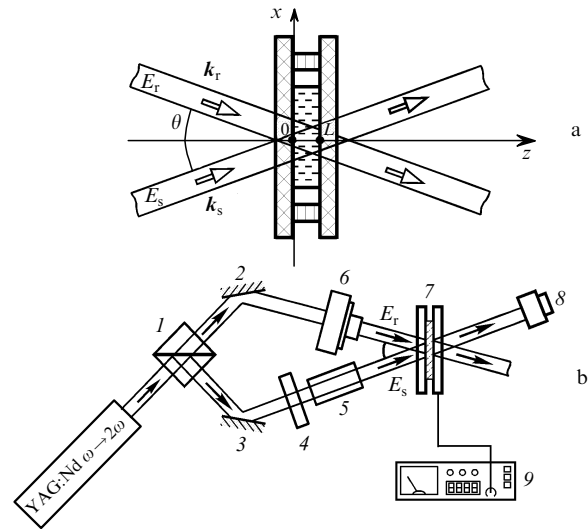


Figure 2. Geometry of two-wave mixing in an ALCP film (a) and schematic of experimental setup (see notation in the text) (b).

tion of azo chromophores, while the decrease in Δn is caused by the inverse reaction of photoisomerisation and thermal relaxation.

The Helmholtz equation for the resultant electric field strength $E = E_s + E_r$ of the light waves in the polymer has the form

$$\nabla^2 E + k_0^2 (1 + \Delta n/n_0)^2 E = 0, \quad (4)$$

where $k_0 = 2\pi n_0/\lambda$ is the wave number and n_0 is the mean (unperturbed) refractive index. Equation (3) together with (4) completely characterises the interaction of light waves in the ALCP. We will solve Eqn (4) using the method of slowly varying amplitudes, assuming that the condition $|\partial A_{r,s}/\partial z| \ll k_0 |A_{r,s}|$ is satisfied. We also use the approximation of low contrast ($m \ll 1$) of the interference pattern in view of the large difference in the intensities of interfering beams (which is typical of such experiments).

In the low-contrast approximation, we can assume that the refractive index grating formed in the polymer is harmonic:

$$\Delta n = n_0 \left\{ \Delta n^{(0)} + \frac{m \Delta n^{(1)}}{2} \times \exp[i(Kx + \delta \sin \Omega t)] + \text{c. c.} \right\}. \quad (5)$$

Here, $\Delta n^{(0)}(t)$ defines the mean value of Δn ; $\Delta n^{(1)}(t)$ characterises the complex amplitude of the refractive index grating at the spatial frequency K . Substituting expressions (2) and (5) into (3) and equating the coefficients for the same powers of the exponential factors, we obtain the system of equations for $\Delta n^{(0)}$ and $\Delta n^{(1)}$:

$$\frac{\partial \Delta n^{(0)}}{\partial t} + \Delta n^{(0)} \left(\frac{1}{T_r} + a_1 W + a_2 W \right) - \frac{\Delta n_s}{n_0} a_1 W = 0, \quad (6)$$

$$\frac{\partial \Delta n^{(1)}}{\partial t} + \Delta n^{(1)} \left(D_x K^2 + i\Omega \delta \cos \Omega t + \frac{1}{T_r} + a_1 W + a_2 W \right) + \Delta n^{(0)} (a_1 + a_2) W - \frac{\Delta n_s}{n_0} a_1 W = 0.$$

Consider two important special cases of coupling of light waves in the ALCP layer: a steady-state energy transfer and a transient process corresponding to the switching on of a strong reference beam without phase modulation at instant $t = 0$. In the former case, putting $\partial\Delta n^{(0)}/\partial t = 0$, we obtain from (6)

$$\Delta n^{(1)} = q \exp(-\gamma t - i\delta \sin \Omega t) \int_{-\infty}^t \exp(\gamma u + i\delta \sin \Omega u) du, \quad (7)$$

where

$$q = \frac{a_1 W \Delta n_s}{(1 + a_1 W T_r + a_2 W T_r) n_0};$$

$$\gamma = \frac{1}{T_r} + a_1 W + a_2 W + D_x K^2.$$

Expression (7) can easily be transformed to

$$\Delta n^{(1)} = q \sum_{n=-\infty}^{\infty} \sum_{l=-\infty}^{\infty} \frac{J_n(-\delta) J_l(\delta)}{\gamma - in\Omega} \exp[-i(n+l)\Omega t], \quad (8)$$

where J_n is the n th the order Bessel function. For small-amplitude phase perturbations (when $\delta \ll 1$), expression (8) gives

$$\Delta n^{(1)} \approx q J_0(\delta) J_1(\delta) \frac{\Omega}{\gamma(\Omega^2 + \gamma^2)^{1/2}} \exp\left(i\Omega t + i \arctan \frac{\Omega}{\gamma}\right). \quad (9)$$

In the case of a transient process ($\delta = 0$, $\Omega = 0$), the solution of the system of equations (6) gives

$$\Delta n^{(1)}(t) = g_1 \exp(-\gamma t) \int_0^t \{g_3 + g_2 \exp[(\beta - \gamma)t']\} \times \exp[\gamma t' + i\zeta(t')] dt', \quad (10)$$

where

$$\beta = D_x K;$$

$$g_1 = \frac{a_1 \Delta n_s W}{n_0(1 + a_2 W T_r + a_1 W T_r)(1 + a_2 I_s T_r + a_1 I_s T_r)};$$

$$g_2 = T_r(a_1 + a_2)[I_0 - n_0 I_s + (a_1 + a_2)(1 - n_0) W T_r I_s];$$

$$g_3 = 1 + I_s T_r(a_1 + a_2);$$

and I_s is the signal-beam intensity in the polymer for $t < 0$, i.e., prior to the switching on of the reference beam. Note that expression (10) is similar to the expressions obtained for the grating amplitude in a semiconductor [19] and in bacteriorhodopsin [6].

Consider the mixing of light waves E_s and E_r in the polymer film of a finite thickness. Taking into account the assumptions made above, we can obtain from (4) the equations for the complex amplitudes $A_r(z)$ and $A_s(z)$ of coupled waves in an ALCP layer:

$$\frac{\partial A_s}{\partial z} = -\frac{\alpha A_s}{2 \cos(\theta/2)} - iC_1 A_s - iC_2^* m A_r, \quad (11)$$

$$\frac{\partial A_r}{\partial z} = -\frac{\alpha A_r}{2 \cos(\theta/2)} - iC_1 A_r - iC_2 m A_s.$$

Here, $C_1 = k_0 \Delta n^{(0)} / \cos(\theta/2)$; $C_2 = k_0 \Delta n^{(1)} / [2 \cos(\theta/2)]$; α is the absorption coefficient of light. According to (6), coefficients C_1 and C_2 depend on the total intensity $W = (|A_s|^2 + |A_r|^2)/2$ of the beams in a complicated way. It is convenient to solve the system (11) by introducing new unknown functions \tilde{A}_r and \tilde{A}_s , which are related to the complex amplitudes A_r and A_s by the expressions

$$A_{r,s} = \tilde{A}_{r,s} \exp\left[-\frac{\alpha z}{2 \cos(\theta/2)}\right]. \quad (12)$$

Such a substitution makes it possible to eliminate from this equation the terms describing linear absorption. The modified system of equations (11) has the integral of motion $|\tilde{A}_s|^2 + |\tilde{A}_r|^2 = 2[I_s(0) + I_r(0)] = \text{const}$, where $I_s(0)$ and $I_r(0)$ are the intensities of light beams at the entrance to the medium ($z = 0$).

It was shown in Ref. [16] that in the case of weak non-linearity of the medium in the steady state, we can disregard the change in the mutual phase difference of coupled waves; i.e., we can assume that $\partial(\arg \tilde{A}_r - \arg \tilde{A}_s)/\partial z = 0$ in the entire layer. In this case, the equation describing the variation of the signal beam intensity $\tilde{I}_s(z)$ takes the form

$$\frac{\partial \tilde{I}_s(z)}{\partial z} = -\frac{2k_0 \text{Im}[\Delta n^{(1)}(W)]}{W_0 \cos(\theta/2)} \tilde{I}_s(z) [W_0 - \tilde{I}_s(z)], \quad (13)$$

where

$$W = W_0 \exp\left[-\frac{\alpha z}{\cos(\theta/2)}\right];$$

$W_0 = I_r(0) + I_s(0)$ is the total intensity of light at the entrance to the medium ($z = 0$); $\tilde{I}_s(z) = |\tilde{A}_s|^2/2$. Since the amplitude $\Delta n^{(1)}$ of the grating in (8) is a periodic function of time, the intensity $I_s(L)$ of the signal wave for $z = L$ is also a periodic function of time.

Let us define the frequency transfer function $F(\Omega)$ of an adaptive interferometer as the ratio of the first-harmonic amplitude of the signal wave (at the perturbation frequency) at the ALCP film exit to the product of the phase perturbation δ and the intensity $I_s(0)$ of the signal beam at the entrance to the polymer:

$$F(\Omega) = \frac{1}{\delta \pi I_s(0)} \left| \Omega \int_0^{2\pi/\Omega} I_s(L) \exp(i\Omega t) dt \right|. \quad (14)$$

In the approximation of low contrast ($m \ll 1$) and a small amplitude of the phase modulation ($\delta \ll 1$), the substitution of the approximate solution of (13) into (14) gives

$$F(\Omega) \approx \frac{1}{2} \int_0^L \frac{q|\Omega|}{\gamma(\Omega^2 + \gamma^2)^{1/2}} dz. \quad (15)$$

Here, we used the relation $J_n(\delta) \approx \delta^n / (2^n n!)$. The quantities q and γ depend on z (through W) in a complicated way. It is impossible to obtain a general analytic expression for $F(\Omega)$ from (14); for this reason, we calculated the transfer function numerically using the Maple V mathematical package (the theoretical curves are shown in Fig. 4d).

The phase mismatch between the beams cannot be neglected in an analysis of the time-dependent response of the polymer [16]. Taking into account the integral of motion, we can reduce the system of equations (11) to the form

$$\frac{\partial \tilde{I}_s}{\partial z} = 4 \frac{\tilde{I}_s (W_0 - \tilde{I}_s)}{W_0} \rho \sin(\xi - \eta), \quad (16)$$

$$\frac{\partial \xi}{\partial z} = \frac{2(W_0 - 2\tilde{I}_s)}{W_0} \rho \cos(\xi - \eta).$$

where $\xi = \arg \tilde{A}_r - \arg \tilde{A}_s$ is the phase mismatch of the beams; $\rho \exp(i\eta) \equiv k_0 \Delta n^{(1)} / [2 \cos(\theta/2)]$; ρ , η are the amplitude and phase of the $\Delta n^{(1)}$ grating, respectively. To simplify calculations, we will write expression (10) for the complex amplitude of the refractive index grating in the form of a system of differential equations:

$$\frac{\partial \rho}{\partial z} + \beta_2 \rho = f(t) \cos(\xi - \eta), \quad (17)$$

$$\rho \frac{\partial \eta}{\partial z} = f(t) \sin(\xi - \eta),$$

where $f(t) = g_1 \{g_3 + g_2 \exp[(\gamma - \beta)t]\}$.

The solution of the system of equations (16) and (17) allows us to analyse the dynamics of a transient process for the signal-beam intensity $I_s(t)$ at the polymer film exit ($z = L$) after the switching on of the reference beam I_r at the instant $t = 0$. It is convenient to represent the results of calculation in the form of the dynamics of ‘two-wave gain’ $\Gamma(t) = \ln[I_s(t)/I_s(0)]/L$, where $I_s(0)$ is the signal-beam intensity at the polymer exit before the switching on of the reference beam. The curves $\Gamma(t)$ obtained by the numerical simulation are presented in Fig. 4c. The results of simulation are considered and compared with experimental data in the next section.

In obtaining numerical estimates, we used the following values of material parameters of the ALCP ($\lambda \approx 532$ nm): $T_r = 10$ s, $\Delta n_s = 0.09$, $a_1 = 10^{-3} \text{ s}^{-1} \text{ W}^{-1} \text{ cm}^2$, $a_2 = 8 \times 10^{-4} \text{ s}^{-1} \text{ W}^{-1} \text{ cm}^2$, $D_x = 1.25 \times 10^{-10} \text{ cm}^2 \text{ s}^{-1}$, $\alpha = 180 \text{ cm}^{-1}$. The rate constants a_1 and a_2 , the thermal relaxation time T_r , and Δn_s for the ALCP at $T \approx 85^\circ \text{C}$ were determined from the measurement of the light-induced addition to the refractive index [20]. In our model, the value of D_x was used as a fitting parameter.

3. Experiment. Comparison with the theory

We studied the two-wave mixing in SPK-4/4 LC polymer samples with a 20%-molar concentration of the azo dye [12,13]. The chemical formula and the linear absorption spectrum of the polymer are shown in Fig. 1. The polymer film of thickness $L = 50 \mu\text{m}$ was studied in a special cell whose construction permitted heating and thermal stabilisation of the ALCP as well as the application of a homeotropically orienting electric field to the sample. The ac electric field $E_a \approx 10^4 \text{ V cm}^{-1}$ of frequency 350 Hz, which was required for maintaining the homeotropic orientation of the ALCP layer, was applied to the cell in all experiments [21, 22].

The scheme of the experimental setup (adaptive interferometer) is shown in Fig. 2b [23]. The 532-nm beam emitted by a diode-pumped, frequency-doubled Nd:YAG laser was incident on a beamsplitter cube (1). Light beams reflected from mirrors (2) and (3) intersect in the plane of the polymer sample (7). The angle of convergence of the beams in air is $\theta \approx 15^\circ$. Phase modulation of the reference beam was obtained using a LiNbO₃ electrooptical modulator (6).

The intensity of the signal beam was controlled by a neutral filter (4). A stack of glass plates (5) in the signal arm of the interferometer was used to level out the optical paths of the interacting beams. This made it possible to attain the maximum contrast of the interference pattern on the polymer film. The signal beam at the polymer cell exit was detected by photodiode (8). The general control of the experiment and data acquisition were performed through the interface (9) and an ADC/DAC converter using a PC.

The interferometer response $I_s(t)$ to the pulsed phase perturbation ($\Delta\varphi \approx 2.7$ rad) of the reference beam is shown in Fig. 3a. The experiments were performed at $T = 85^\circ \text{C}$, the intensity ratio for the incident beams was $I_r/I_s = 11$, and the total intensity was $I_r + I_s = 330 \text{ mW cm}^{-2}$. One can see from Fig. 3a that the interferometer strives to automatically compensate phase perturbations, i.e., to adapt to the change in the operating conditions. The transient process for a step phase perturbations $\Delta\varphi \approx 1$ rad is shown in Fig. 3b for various temperatures of the polymer. One can see that polymer heating accelerates the transient process and enhances the response signal.

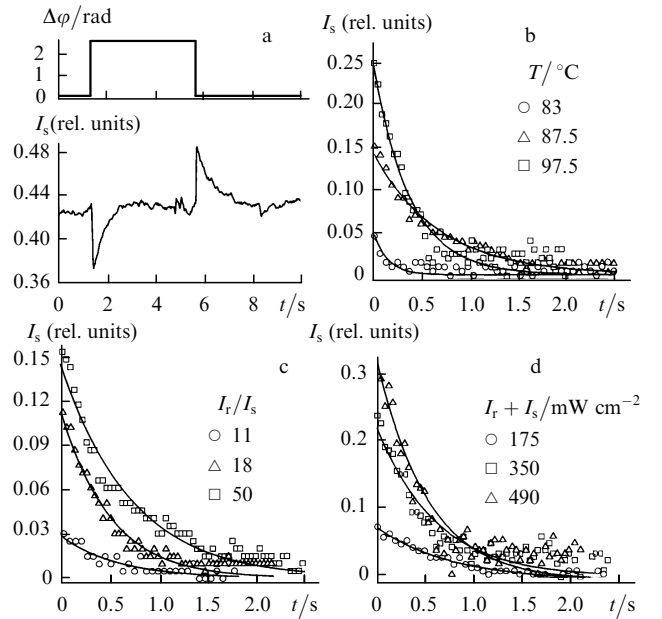


Figure 3. Response I_s of the adaptive interferometer to the pulsed phase perturbation $\Delta\varphi$ of the reference beam (a) and the dynamics of the transient process for various temperatures T of the polymer film ($I_r/I_s = 11$, $I_r + I_s = 175 \text{ mW cm}^{-2}$) (b), various intensity ratios I_r/I_s of coupled beams ($T = 87.5^\circ \text{C}$, $I_r + I_s = 175 \text{ mW cm}^{-2}$) (c), and various total intensities $I_r + I_s$ ($T = 87.5^\circ \text{C}$, $I_r/I_s = 11$) (d). Solid curves are approximations of the initial parts of the dependences.

The transient process is shown in Fig. 3c for various intensity ratios I_r/I_s . In this case, the rate of the response remains almost unchanged since $I_r + I_s = \text{const}$, but the output signal increases with decreasing contrast due to the change in the contrast of the interference pattern affecting the phase-grating amplitude $|\Delta n^{(1)}|$. Fig. 3d shows the dependences $I_s(t)$ for a step variation of the phase and for various values of the total input intensities $I_r + I_s$ of the beams. One can see that the increase in $I_r + I_s$ leads to a decrease in the time of the transient process induced by the step perturbation of the phase.

The dynamics of the transient process after the switching on of the reference light beam at the instant $t_0 = 2$ s is presented in Figs. 4a, b, and c. No additional phase shift was introduced between the beams interfering in the polymer film. The two-wave gain $\Gamma(t) = \ln[I_s(t)/I_s(0)]/L$ was calculated from the measured values of I_s . The curves presented in Fig. 4a were obtained for different temperatures of the ALCP film. One can see that heating considerably accelerates the response and enhances the signal. The dependences $\Gamma(t)$ obtained for various intensity ratios I_r/I_s of the incident beams are shown in Fig. 4b. In this case, an increase in the ratio I_r/I_s results in an increase in $\Gamma(t)$.

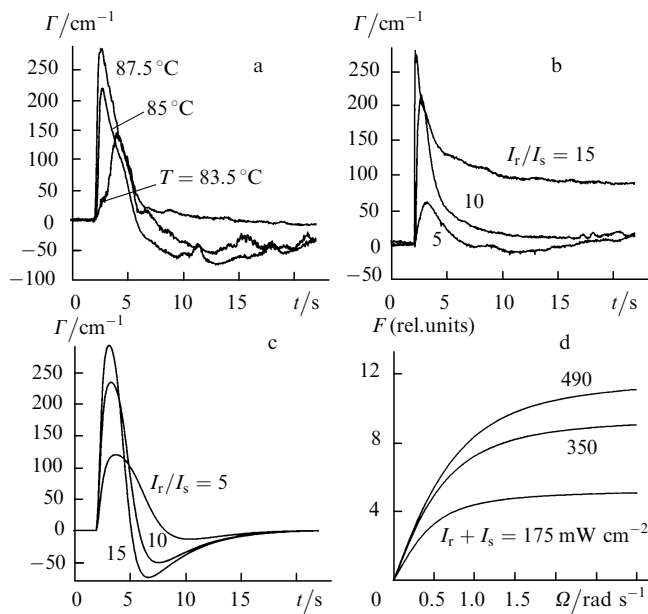


Figure 4. Transient process after the switching on of the reference beam at the instant $t_0 = 2$ s: dependences $\Gamma(t)$ for various temperatures T of the film (a), various intensity ratios I_r/I_s of coupled beams (b), the theoretical dependence $\Gamma(t)$ for $I_r + I_s = 330$ mW cm $^{-2}$ and various values of I_r/I_s (c), and the theoretical frequency transfer function $F(\Omega)$ of the adaptive interferometer for $\delta = 0.1$ rad, $I_r/I_s = 11$, and various values of $I_r + I_s$ (d).

Fig. 4c shows the theoretical curves $\Gamma(t)$ obtained by the numerical solution of Eqns (16) and (17). One can see that for $t \leq 5$ s, the theoretical curves are in good agreement with the experimental data. The discrepancies for $t > 5$ s may be due to the fact that our model neglects the nonlinear absorption in the ALCP. In addition, the initial equation (3) is approximate for photoisomerisation of azo chromophores because it does not describe the angular redistribution of the azo dye isomers [21]. For this reason, the initial increase and subsequent relaxation of $\Gamma(t)$ in the theoretical analysis are determined by the same time scale. Finally, Fig. 4d depicts the frequency transfer function of an adaptive interferometer calculated from Eqns (13) and (14) for various values of the total intensity of the incident light. The shape of the curves corresponds to the transfer function $F(\Omega) \sim \Omega/(\Omega^2 + \gamma^2)^{1/2}$ of the differentiating circuit. The differentiation of the phase perturbation was also observed in experiments (see Fig. 3a).

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