# Multiwave Mixing in Complex Molecular Media and Dynamic Modes of Light-Wave Transformation

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The mechanism of multiwave mixing realized in solutions of complex organic compounds (dyes), in conditions when higher-order nonlinearities are exhibited along with the cubic one, has been analyzed theoretically and tested experimentally. Nonlinear recording of dynamic holograms enables significant improvement of the potentialities of holographic methods used for image conversion due to switching from phase conjugation to smoothing and amplification of the wavefront spatial structure. It has been demonstrated that a nonstationary energy exchange between the interacting waves is the cause for transition from the optical bistability mode to the mode of intensity self-oscillations.

**Key words:** dynamic holograms, multiwave mixing, resonant media, higher-order nonlinearities

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## 1 Introduction

Extended potentialities of the diffraction methods for light wave transformation of laser radiation are determined by nonlinear recording of dynamic holograms, enabling multiwave mixing in media characterized by the fifth and higher-order nonlinearities. In a variety of such media, higher-order nonlinearities in resonant media are conditioned by the absorption saturation effect and transitions between different excited states of the molecules. Owing to the nonlinear relationship between a light-induced change in the refractive index of a resonant medium and the intensity, the groove profile of a holographic grating becomes distorted and ceases to be sinusoidal. The diffraction characteristics of such dynamic structures may be analyzed using Fourier expansion of phase and amplitude response of the medium in spatial harmonics of the grating [1]. Scattering at varying harmonics of the grating determines the second or higher-order diffraction. The grating's angular selectivity for volume dynamic holograms permits independent reconstructing of the waves diffracted into different orders, through a change in the propagation direction of the reading wave. In Bragg mode the reading beam guided at an angle that is associated with the M-th - order diffraction will be scattered from the corresponding spatial harmonic of the grating. In the process, diffracted wave $E_D$ is determined by nonlinear polarization $P = \chi^{(N-1)}(E_1 E_S^*)^M E_2$ (M is the diffraction order), and N-wave mixing ($N = 2(M+1)$) for the th - order nonlinearity takes place. The propagation direction of the diffracted wave $E_D$ is determined from the phase synchronism condition for wave vectors

$\vec{k}_D = M\vec{k}_1 - M\vec{k}_S + \vec{k}_2$ [2]. And the light beams diffracted from different components of the dynamic grating are distinguished not only by the propagation direction, but also by the wavefront spatial structure. Optimizing the conditions of a multiwave mixing allows for essentially nonlinear character of the energy exchange between the interacting waves and realization of optical multistability [3]. Multistable states of light fields differ both in the intensity of the diffracted wave and spatial distribution of the intensity for all waves within the nonlinear medium volume.

In the present work a theoretical model for nonlinear recording of dynamic holograms in complex molecular media has been developed and a mechanism of light field conversion upon multiwave mixing has been analyzed. The conditions for realization of effective energy exchange between the light beams have been established and different transitions to the mode of intensity self-oscillations have been studied.

## 2 A theory of multiwave mixing in resonant media

A light-induced change in the optical properties of resonant media within the interference field of interacting waves results in recording of dynamic diffraction gratings (holograms). Their formation may be due to changing of the refractive index and absorption coefficient on transition of the molecules to higher states and also due to thermal nonlinearity caused by heating of the medium. Along with transitions in the principal channel, transitions between the excited singlet and triplet molecular states may contribute considerably to a light-induced change of the optical properties. As it takes place, the light-induced response of a resonant medium may be characterized by a complex dependence on the intensity causing exhibition of higher-order nonlinearities. The experiments conducted with the use of gases [4-6], semiconductor glasses [7-10], dyes [10] as nonlinear media support the possibility for realization of multiwave mixing in the form of continuous laser radiation and nano- or picosecond pulses as well. To give an adequate theoretical description for the process of coherent light - wave interaction in the above-mentioned cases, one should take into consideration the second and higher-order components in series expansion of the nonlinear susceptibility in harmonics of the dynamic grating recorded by interacting waves within the medium volume.

To analyze expansion of the medium susceptibility in grating harmonics, first we represent the nonlinear susceptibility as a power series of the light field intensity

$$\chi_{nl} = \chi^{(3)}E_\Sigma^2 + \chi^{(5)}E_\Sigma^4 + \chi^{(7)}E_\Sigma^6 + ....., \qquad (1)$$

where $E_\Sigma^2 = (8\pi/cn_0)I_0(1+\gamma\cos(\vec{K}\,\vec{r}))$ is the total field of interfering waves, $\vec{K}$ is a wave vector of the grating, $I_0$ is an average field intensity within the medium, $\gamma$ is a modulation depth of the intensity, $n_0$ is the non-resonant component of the refractive index. Using the binomial formula expansion, we represent the nonlinear susceptibility as

$$\chi_{nl} = \sum_{m=1}^{\infty}\chi^{(2m+1)}(8\pi I_0/cn_0)^m \times \sum_{m=1}^{\infty}C_m^k\gamma^k\cos^k(\vec{K}\,\vec{r}), \qquad (2)$$

where $C_m^k = m!/k!(m-k)!$ are binomial coefficients. Then the components of the nonlinear susceptibility Fourier-series expansion in spatial harmonics of the grating

$$\chi_m = \frac{1}{2\pi}\int_{-\pi}^{\pi} \chi_{nl} \exp[-im(\vec{K}\cdot\vec{r})] d(\vec{K}\cdot\vec{r}), \quad (3)$$

may be written as follows:

$$\begin{aligned}
\chi_1 &= \gamma\chi^{(3)} I_0 + 2\gamma\chi^{(5)} I_0^2 + 3\gamma(1+\gamma^2/4)\chi^{(7)} I_0^3 \\
&+ 4\gamma(1+3\gamma^2/4)\chi^{(9)} I_0^4 + ..., \\
\chi_2 &= (\gamma^2/2)\chi^{(5)} I_0^2 + (3\gamma^2/2)\chi^{(7)} I_0^3 \\
&+ 3\gamma^2(1+\gamma^2/6)\chi^{(9)} I_0^4 + ..., \qquad (4) \\
\chi_3 &= (\gamma^3/4)\chi^{(7)} I_0^3 + \gamma^3\chi^{(9)} I_0^4 + \ldots,
\end{aligned}$$

Each member of the Fourier series describing the grating and hence the diffraction efficiency of dynamic holograms is determined by different orders of nonlinear susceptibility. Considering that expansion of the nonlinear susceptibility into a series in powers of the field (1) holds true for small values of optical nonlinearities when each nonlinearity is significantly lower in value than the preceding one, the main contribution into expression (4) is made by the first members of the series. Such a situation is true for e.g. a two-level medium model at intensities below the saturation intensity of a resonance transition. In these conditions the diffraction efficiency into the $M$-th order may be unambiguously related to nonlinear susceptibility $\chi^{(2M+1)}$, and independent measurements may be performed for each order of nonlinear susceptibility.

Further analysis will be performed for a three-level model of a resonant medium with due regard for radiative and nonradiative transitions in the principal and excited singlet channels ($S_0-S_1$ and $S_1-S_2$). Considering that experimental intensity of the interacting waves is, as a rule, appreciably lower than the saturation intensity in the excited channel, analysis is performed including saturation of the principal resonance transition but for linear absorption from the excited level. Using the solution for a system of kinetic equations describing the level populations and dispersion relations relating changes in the refractive index and absorption coefficient, we represent the overall nonlinear (resonance and thermal) susceptibility of the medium as [11]

$$\chi_{nl} = \frac{n_0\kappa_0}{2\pi}\left(\frac{\widehat{\theta}_{12}}{B_{12}} - \frac{\widehat{\alpha}I + b_T I^2}{1+JI}\right), \qquad (5)$$

where

$$\begin{aligned}
\widehat{\alpha} &= \frac{(\widehat{\theta}_{12} + \widehat{\theta}_{21} - \widehat{\theta}_{23})}{vP_{21}} - \sigma_T(1-\mu_{21}) \\
b_T &= \sigma_T \frac{B_{23}}{vP_{21}}(1-\mu_{32}) \\
J &= \frac{B_{12}+B_{21}}{vP_{21}}
\end{aligned}$$

$B_{12}(\omega)$ are the Einstein coefficients for stimulated transition $i-j$ ; $v$ is the speed of light in the medium; $P_{ij}$ is a total probability of spontaneous and nonradiative transitions in $i-j$ channel; $\widehat{\theta}_{ij}(\omega) = \theta_{ij}(\omega) + iB_{ij}(\omega)$ (coefficients $\theta_{ij}(\omega)$ are related by Kramers-Kronig relations $\theta_{ij}(\omega) = \frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{B_{ij}(\omega')}{\omega'-\omega}$ to Einstein's coefficients $B_{ij}(\omega)$), $\sigma_T = 2\omega(dn/dT)t_{pulse}/cC_\rho$, $t_{pulse}$ is the interaction time, $C_\rho$ is the unit volume heat capacity, $dn/dT$ is a thermooptic coefficient, $\mu_{ij}$ luminescence quantum efficiency in $i-j$ channel, $\kappa_0$ is a linear extinction coefficient.

A theoretical description of the multiwave mixing process is based on the approximation for slowly varying amplitudes making it possible to write a system of reduced wave equations for the interacting light fields [12]

$$\frac{\partial E_{1,S}}{\partial z} = \frac{i2\pi\omega}{cn_0}\left[\chi_0(\omega)E_{1,S} + \chi_{\pm1}(\omega)E_{S,1}\right], \quad (6)$$

$$\frac{\partial E_{2,D}}{\partial z} = -\frac{i2\pi\omega}{cn_0}\left[\chi_0(\omega)E_{2,D} + \chi_{\mp M}(\omega)E_{D,2}\right]. \tag{7}$$

where $\chi_m$ are the Fourier series expansion components of nonlinear susceptibility determined by relation (3). Proceeding from (5), Fourier series expansion components of $\chi_m$ may bee represented as

$$\chi_0(\omega) = \frac{n_0\kappa_0}{2\pi}\left(\frac{\widehat{\theta}_{12}}{B_{12}} + \frac{b_T}{J}I_\Sigma + \left(\widehat{\alpha}/J + b_t/J^2\right)\frac{1-A_0}{A_0}\right), \tag{8}$$

$$\chi_{\pm1}(\omega) = \frac{n_0\kappa_0}{2\pi}\left(\frac{b_t}{J}\sqrt{I_1I_S} - \left(\widehat{\alpha}/J + b_T/J^2\right) \times \frac{2J\sqrt{I_1I_S}}{A_0\left(1+JI_\Sigma+A_0\right)}\right)\exp\left[\pm i\left(\varphi_1-\varphi_S\right)\right], \tag{9}$$

$$\chi_{\pm2}(\omega) = \frac{n_0\kappa_0}{2\pi}\frac{4\left(\widehat{\alpha}J+b_T\right)I_1I_S}{A_0\left(1+JI_\Sigma+A_0\right)^2} \times \exp\left[\pm i2\left(\varphi_1-\varphi_S\right)\right], \tag{10}$$

.....

$$\chi_{\pm1}(\omega) = \frac{n_0\kappa_0}{2\pi}\frac{\left(\widehat{\alpha}J+b_T/J\right)\left(-2J\sqrt{I_1I_S}\right)^M}{JA_0\left(1+JI_\Sigma+A_0\right)^M} \times \exp\left[\pm iM\left(\varphi_1-\varphi_S\right)\right], \tag{11}$$

where $A_0 = \left(\left(1+JI_\Sigma\right)^2 - 4J^2I_1I_S\right)^{1/2}$, $I_\Sigma = I_1 + I_2 + I_S + I_D$ (approximation of a weak diffracted wave $I_D \ll I_1, I_2, I_S$).

The results obtained from numerical analysis of the energy efficiency of multiwave mixing are given in Fig. 1, where the diffraction efficiency $\xi = I_D(r=0)/I_2(r=L)$ is presented as a function of the intensity of hologram recording waves that is normalized to the saturation intensity of the principal resonance transition $I_{sat} = J^{-1}$. The calculations have been based on (a) four- and (b) six-wave mixing with regard both to pure resonance and combined (resonance and thermal) nonlinearities, for the parameters associated with single-pulse excitation conditions in complex molecular media: $t_{pulse} = 20\ ns$, $n_0 = 1.36$, $(dn/dT)C_\rho^{-1} = -10^{-4}J^{-1}\ cm^3$, $\lambda = 0.5\mu m$, $\Delta\lambda = 25\ nm$, ($\lambda$ and $\Delta\lambda$ represent the center and halfwidth of the absorption band), $\mu_{21} = 0.5$, $\mu_{32} = 0.005$ (luminescence quantum yields). As seen, in both cases a maximum of the diffraction efficiency is observed at intensities in the neighborhood of the saturation intensity of a resonance transition. And involvement of thermal nonlinearity (dashed line) has practically no effect on the efficiency of six-wave mixing (dashed and solid lines in Fig.1b are almost coincident) whereas the efficiency of four-wave mixing increases considerably at the intensities above the saturation one. This is also confirmed by analysis of expressions (8) - (10). Thermal nonlinearity introduces additional terms to the expressions for the zero- and first-order expansion components and affects only the factor in case of the second-order diffraction.

## 3 Experimental

Experimental study of the efficiency of multiwave mixing has been conducted with Rhodamine $6G$ solution in ethanol and YAG laser second-harmonic radiation ($\lambda = 532\ nm$, $t_{pulse} = 20\ ns$) falling into $S_0 - S_1$ absorption band of the dye. The selected diffraction scheme was associated with Bragg mode and the signal and reference waves propagating in the same direction (Fig.

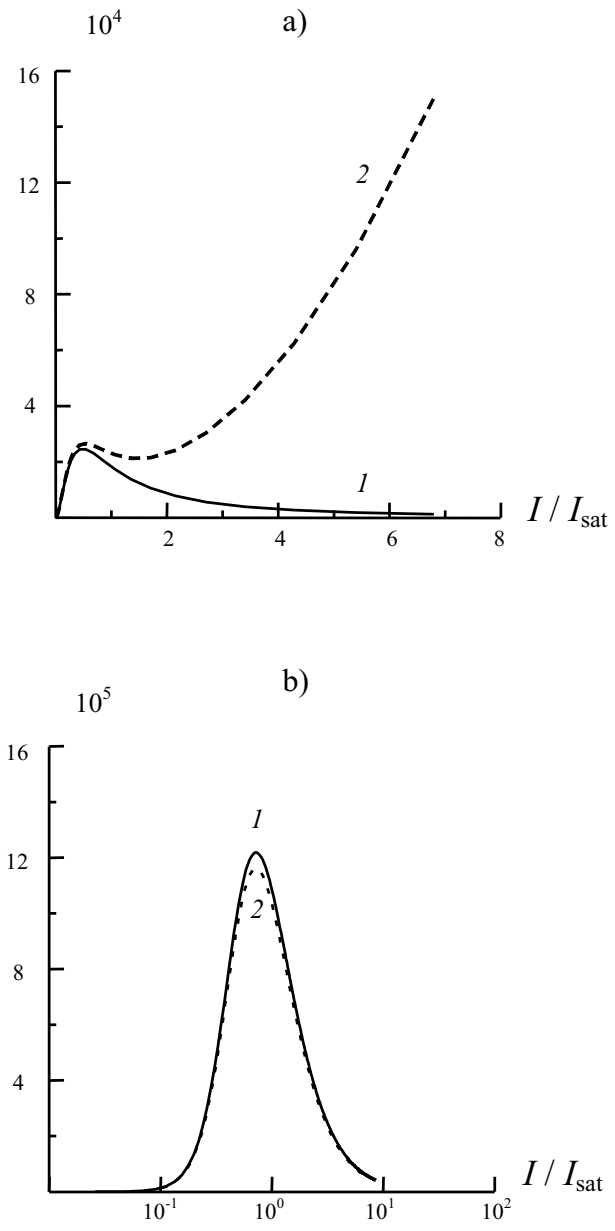


FIG. 1. Diffraction efficiency $\xi$ as a function of the hologram recording waves for (a) four- and (b) six-wave mixing with regard to (1) resonance only or (2) combined (resonance and thermal) nonlinearities.

2). Signal wave $E_S$ and reference wave $E_1$ were formed by mirrors 3, 7 and 8. Reading wave was directed into the nonlinear medium by mirror 4. This mirror was movable allowing for variation of the reading angle. An angle ($\alpha = 90\ mrad$ ) between the propagation directions of the reference and signal waves provided overlapping of the interacting waves over the whole length of the dye cell. The energy efficiency of multiwave mixing $\xi$ has been measured by a recording system 6, 9.

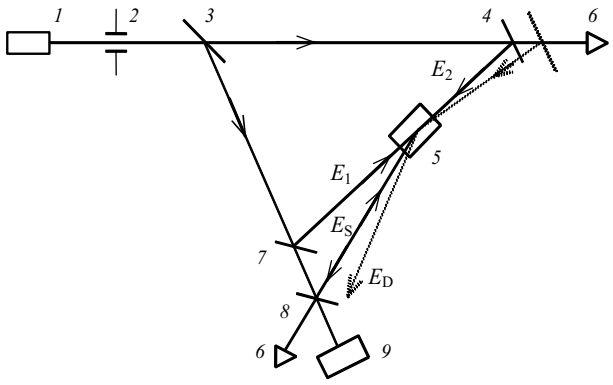


FIG. 2. Experimental setup: (1) laser; (2) diaphragm; (3, 4, 7, 8) mirrors; (5) cell with dye solution; (6, 9) measuring system.

Fig. 3 presents the dynamic hologram diffraction efficiency $\xi$ as a function of angle $\beta$ formed by the propagating waves $E_2$ (reading) and $E_1$ (reference). Two maxima appearing in the figure are due to the fact that 1) for the counter-propagating reading wave ($\beta = 0$ ) wavefront conversion is realized at four-wave mixing, when the polarization giving rise to $E_D$ wave is of the form $P \sim \chi^{(3)} E_1 E_2 E_S^*$ (phase synchronism condition $\vec{k}_D = \vec{k}_1 - \vec{k}_S + \vec{k}_2$), and 2) $\beta \approx \alpha/2$ is associated with a phase synchronism condition $\vec{k}_D = \vec{k}_1 - \vec{k}_S + \vec{k}_2$ for six-wave mixing ($P \sim \chi^{(5)} (E_1 E_2)^2 E_S^*$).

Fig. 4 gives the diffraction efficiency $\xi$ as a function of the intensity of hologram recording waves for both mixing types together with the intensity dependence of optical thickness of the dye solution. It is seen that in accordance with the above theoretical analysis, a maximum efficiency of six-wave mixing is realized at the intensities close to the saturation one. In case of four-wave

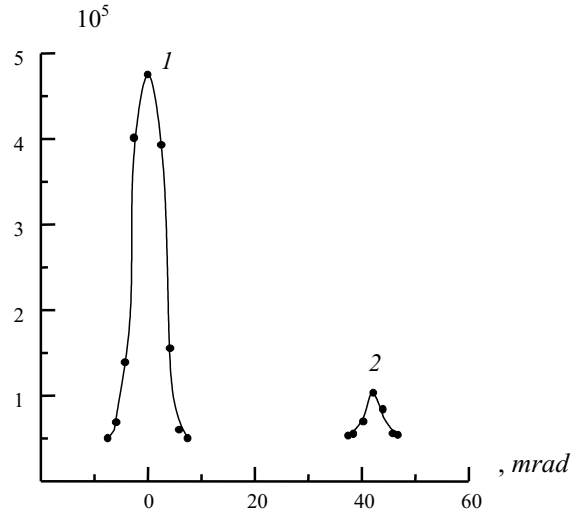


FIG. 3. Diffraction efficiency as a function of the reading-wave incidence angle $\beta$, for the following configurations: $I_2 = 2I_1 = 4I_S$ =2 MWcm$^{-2}$ (1), $I_2 = 2I_1 = 2I_S$ =2 MWcm$^{-2}$ (2), (the diffraction efficiency values for curve 2 are given with 10-fold increase).

mixing a considerable growth in the diffraction efficiency is observed at the intensities above the saturation one, due to thermal nonlinearity on absorption from the energy level.

It should be noted that the light beams diffracted from different spatial components of the dynamic grating in the process of four- and six-wave mixing are distinguished not only by the propagation direction but also by the wavefront spatial structure. Higher order diffraction results in greater phase distortions (a phase of a diffracted wave is a multiple of the phase of the signal one, i.e. $\varphi_D = -M\varphi_S$) and corresponding change in dimensions of the reconstructed image.

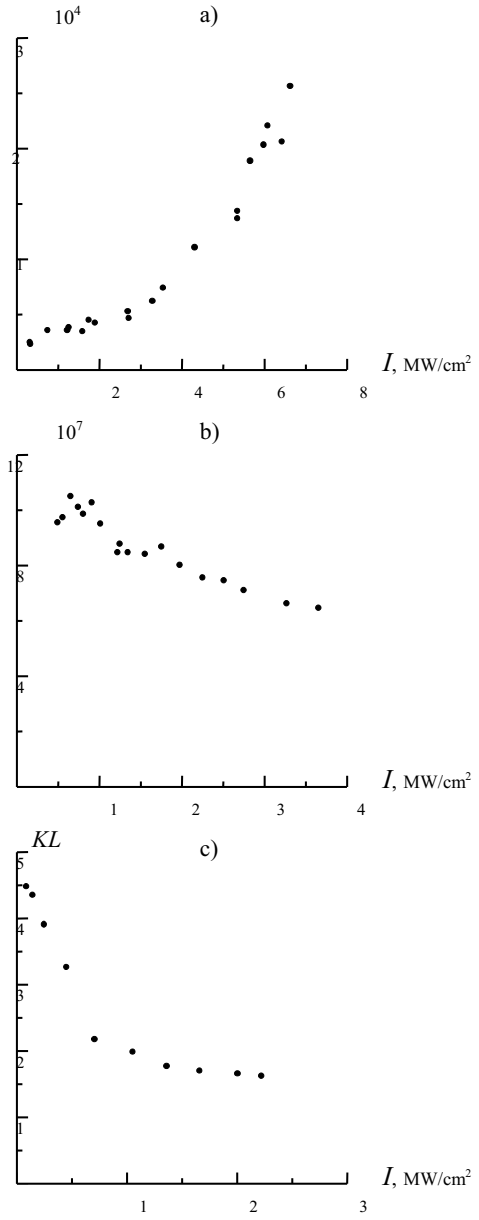


FIG. 4. Diffraction efficiency $\xi$ as a function of the intensity of hologram recording waves for (a) four- and (b) six-wave mixing and (c) optical thickness of a nonlinear layer versus the laser radiation intensity.

## 4 Optical bistability

Apart from thermal nonlinearity, a significant increase in the efficiency of muliwave mixing may be expected for recording of resonance dynamic phase gratings at a frequency tuned away from the absorption profile center [13]. Owing to optimization of the conditions of multiwave mixing, the efficiency of energy exchange between

the interacting waves may be improved. As it take place, intensities of the signal and diffracted waves are growing within the medium volume and become comparable to those of the reference and reading waves, making the energy exchange between the waves essentially nonlinear in character. In the process optical multistability may be exhibited in case when the same boundary conditions are responsible for realization of different stationary states. And multistability may be considered proceeding from classical conditions for observation of optical bistability and hysteresis [14]. The parametric feedback upon multiwave mixing is achieved through rescattering of the waves from the dynamic gratings formed by these waves within the volume of a nonlinear medium. Such a distributed feedback acts as mirrors, similar to the well-known patterns for realization of optical bistability in Fabry-Perot interferometer [15].

Fig. 5 represents characteristic bistable functions for the mode of four-wave mixing at a frequency tuned away from the absorption band center at varying intensities of the signal and reference waves incoming into the nonlinear medium. As seen, a bistable region is located near the saturation intensity and is greatly dependent on the ratio of input intensities. To realize optical bistability, intensity of the reference wave should be in excess of the signal wave intensity by the order of magnitude and more. In this case each of the bistable states is associated with its own spatial distribution of the intensities and phases characteristic for the waves interacting within the medium volume. An analysis for stability of the stationary solutions performed by varying the boundary conditions has demonstrated that an intermediate solution corresponding to the upper portion of the circular region is unstable. At small deviations of the intensities of interacting waves at the entrance to the medium, a system was switching from unstable state to one of the stable states associated with maximum or minimum diffraction efficiency. The dynamics of multiwave mixing is treated in greater detail in subsequent sections based on a nonstationary model of the interaction.

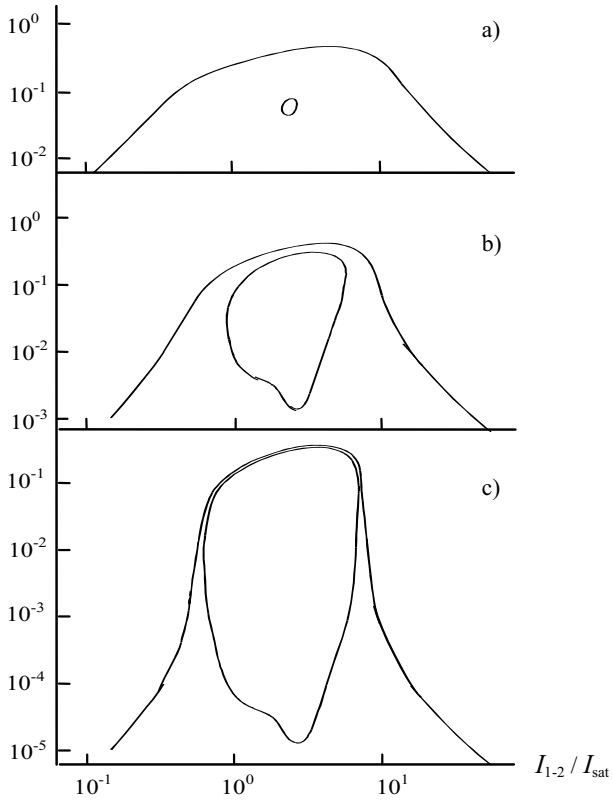


FIG. 5. Diffraction efficiency $\xi$ as a function of the intensities of reference and reading waves ($I_1(z=0) = I_2(z=L) = I_0$) with intensity detuning and the signal wave intensity $I_S(z=0)/I_0 = 10^{-1}$ (a), $10^{-2}$ (b), $10^{-4}$ (c).

## 5 Dynamic interaction modes

To describe the formation dynamics of diffraction structures in resonant media, we again use Fourier series expansion of the nonlinear suscep-

tibility in harmonics of the dynamic gratings, and interaction of the light fields may be defined by equations (6), (7). A nonstationary character of the interaction may be included by the use of a classical kinetic equation for the population of the excited energy level.

In this approach it is advantageous to use the representation of nonlinear medium susceptibility $\chi_{nonlin} = n_0\Delta n/2\pi$ relating its value to the number of molecules passing to the excited state

$$\chi_{nl} = \frac{\hbar n_0^2}{2\pi} N_\Sigma \left( \widehat{\theta}_{12} - \frac{N}{N_\Sigma}(\widehat{\theta}_{12} + \widehat{\theta}_{21}) \right), \quad (12)$$

where $N_\Sigma$ - is the number of molecules in the unit volume, $N/N_\Sigma$ - portion of molecules in the excited state.

In the approximation of a two-level medium model the kinetic equation for the population of the excited level is of the form

$$\frac{\partial N}{\partial t} = N_\Sigma B_{12} I/v - N((B_{12} + B_{21})I/v + P_{21}). \quad (13)$$

Considering that $I$ is characterized by a periodic redistribution within the medium volume, we perform spatial series expansion (similar to nonlinear susceptibility) of the population at the excited level within a resonant medium in harmonics of the grating

$$N = \sum_{m=-\infty}^{\infty} N_m \exp[im(\vec{K}\cdot\vec{r})], \quad (14)$$

where

$$N_m = \frac{1}{2\pi}\int_{-\pi}^{\pi} N \exp[-im(\vec{K}\cdot\vec{r})] d(\vec{K}\cdot\vec{r}).$$

Kinetic equation (13) is transformed by the use of expansion (14) into an infinite system of differential equations for the associated expansion components in the following way:

$$\frac{\partial N_0}{\partial \tau} = N_\Sigma \alpha_1 I_\Sigma - N_0(1+\alpha I_\Sigma) - N_1 \alpha S - N_1^* \alpha S^*,$$

$$\frac{\partial N_1}{\partial \tau} = N_\Sigma \alpha_1 S^* - N_0 \alpha S^* - N_1(1+\alpha I_\Sigma) - N_2 \alpha S,$$

$$\frac{\partial N_2}{\partial \tau} = -N_1 \alpha S^* - N_2(1+\alpha I_\Sigma) - N_3 \alpha S, \quad (15)$$

.....

$$\frac{\partial N_m}{\partial \tau} = -N_{m-1} \alpha S^* - N_m(1+\alpha I_\Sigma) - N_{m+1} \alpha S,$$

.....

where $\tau = t^* P_{21}$ is the interaction period normalized to the characteristic lifetime of the molecules in the excited state, $\alpha_1 = B_{12}/vP_{21}$ and $\alpha = (B_{12} + B_{21})/vP_{21}$- coefficients determined by the spectral characteristics of the medium, $I_\Sigma = I_1 + I_2 + I_S + I_D$ and $S = \sqrt{I_1 I_S} + \sqrt{I_2 I_D}\exp(i\Phi)$ are the total intensity and interference term, $\Phi = \varphi_1 + \varphi_2 - \varphi_S - \varphi_D$ is the value of phase detuning of interacting waves.

It should be noted that the above formulae derived without an approximation of a weak diffracted wave and are completely describing the dynamic mixing mode in conditions of a high-efficiency parametric energy exchange. The problems associated with the necessity to solve an infinite system of differential equations (15) may be resolved by analysis of the solutions derived with different number of the components used. As is seen from a numerical analysis, in practice the calculations of the diffraction efficiency may be limited to the second or third-order expansion component depending on the interaction conditions.

The results obtained during calculations of the energy efficiency of four-wave mixing as a function of time for different intensities of the signal and reference waves are given in Fig. 6. And the interaction parameters were the same as in the above case of stationary interaction (Fig. 5). As seen, for high intensity of the signal wave (Fig. 6a) the transition to the stationary value takes a period of time that is on the order of the lifetime of a molecule in the excited state. A nonmonotonic character of the transition is observed at intensities exceeding the saturation intensity corresponding to optimum conditions of the interaction.

The efficiency of a parametric energy exchange between the waves increases considerably as the intensity of a signal wave incoming into the medium is decreased, making the intensity of the diffracted wave at the output practically invariable (Fig. 6c).However, a character of the relationship between the reflection factor and time is changing. A stationary value of the diffraction efficiency is reached in steps. Initially the system is in the intermediate state associated with the upper portion of the ring-shaped region (Fig. 5c), and then a transition to the stationary state corresponding to a maximum value of the diffraction efficiency is realized. A step-wise transition to the stationary value is in accord with the above conclusion about instability of the intermediate solution.

Further decrease in the intensity of the signal wave results in the essential changing of the dynamic behavior. In the interval of parameters associated with the optical bistability mode (Fig. 5c), transition to the dynamic mode of interaction allows for realization of the mode of intensity self-oscillations (Fig. 6c) when the output intensity oscillations are observed at constant intensities of the waves incoming into the medium.

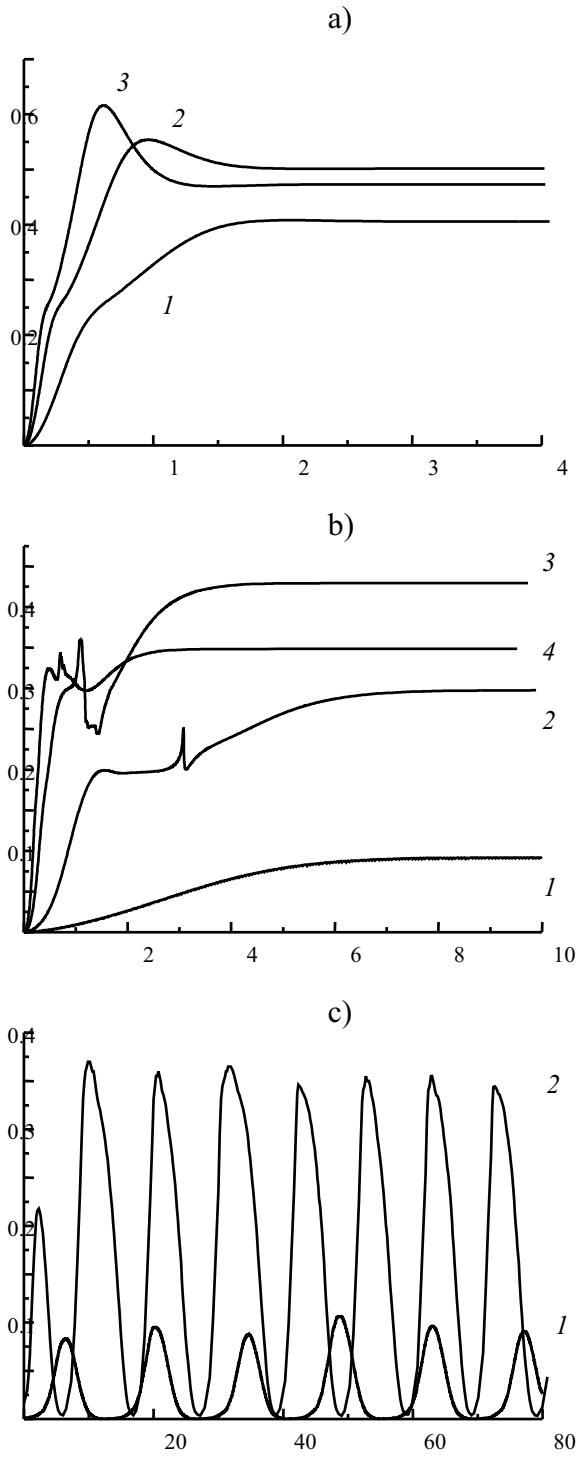


FIG. 6. Diffraction efficiency $\xi$ as a function of the interaction period with the signal wave intensity $I_S(z = 0)/I_0$ =$10^{-1}$(a), $10^{-2}$ (b), $10^{-4}$ (c) and

intensities of the reference and reading waves $I_1(z=0) = I_2(z=L) = I_0$:
(a) $I_0/I_{sat}$ = 1 (1), 2(2), 3 (3);
(b) $I_0/I_{sat}$ = 0.3 (1), 0.8 (2), 2 (3), 3.2 (4);
(c) $I_0/I_{sat}$ = 0.5(1), 2 (2).

And in the process the intensities are changing over the range determined by the boundaries of a bistable region. Varying the input intensities of light fields, one is enabled to control the amplitude and frequency of the oscillations.

As seen in Fig. 6, in conditions of an efficient parametric energy exchange the diffraction efficiency and hence the intensity of a diffracted wave is weakly dependent on the intensity of the signal wave incoming to the medium. The limiting case ($I_S(z=0) \to 0$) is related to the mode of parametric generation of counter-propagating waves. Because of this, intensity self-oscillations realized at low intensities of the signal wave ($I_S/I_1 \ll 1$) may be characterized as a pulse generation process of the counter-propagating waves.

## 6 Conclusion

Thus, the theoretical and experimental data obtained during analysis of multiwave interaction in complex molecular media point to the effective realization of four- and six-wave mixing in conditions when higher order nonlinearities are developing. It has been demonstrated that thermal nonlinearity associated with the induced absorption from the excited level results in a significantly improved efficiency of four-wave mixing, whereas six-wave mixing remains practically unaffected. In case of effective energy exchange at the dynamic phase gratings for a weak signal wave, at constant input intensities one can realize the mode of optical bistability transformable to the dynamic intensity self-oscillations mode of the interacting waves leaving the medium. As this takes place, intensity self-oscillations occur between the highest and lowest stationary values associated with the region of optical bistability.

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