FREQUENCY CONVERSION OF COHERENT IMAGES ON INTRACAVITY MULTIWAVE MIXING

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ABSTRACT

The schemes for recording and reading of dynamic holograms in conditions of nondegenerate four- and six-wave mixing in a nonlinear Fabry-Perot interferometer have been analyzed theoretically. It has been demonstrated that there is a possibility for a considerable improvement in the diffraction efficiency and angular selectivity of dynamic gratings in the interferometer compared to the off-cavity interaction. A method for the frequency conversion of coherent images with simultaneous phase conjugation has been realized experimentally.

Key words: dynamic holography, frequency conversion of images, nondegenerate multiwave mixing, nonlinear interferometer

1. INTRODUCCION

Extension of a theory for the formation and interaction of light beams in distributed nonlinear systems (e.g., nonlinear Fabry-Perot interferometer) is of great importance for the basic and applied research. The results of the related studies may be used for the development of new optical switchable devices, high-speed data processing systems, optical memories, etc. [1].

The use of an optical cavity in the schemes for recording of stationary [2] and dynamic [3, 4] holograms in semiconductor materials [5], solutions of complex organic compounds [6, 7], amplifying media [8] has demonstrated the possibility for a considerable improvement in the light-field transformation efficiency in conditions of Bragg diffraction. The advantages of optical cavities are particularly important for realization of the frequency-degenerate intracavity light-wave mixing [9] in the case when the intracavity feedback is effected for a reading light beam.

This paper presents a theoretical and experimental study into the efficiency of the coherent-image frequency conversion processes on multiwave interactions in Fabry-Perot interferometer, with the solutions of complex organic compounds (dyes) used as a nonlinear medium.

2. THEORETICAL MODEL FOR FREQUENCY-NONDEGENERATE MULTIWAVE INTERACTIONS IN NONLINEAR INTERFEROMETER

A theoretical model describing the nondegenerate multiwave mixing is based on a system of coupled wave equations for the complex amplitudes of light waves in the conditions when dynamic gratings are recorded by the signal \( E_s \) and reference \( E_1 \) waves at the frequency \( \omega \), whereas reading is performed by the wave \( E_2 \) at the doubled frequency \( 2\omega \). Depending on the propagation direction of a reading wave, one can realize four- (Figure 1a) or six-wave (Figure 1b) mixing.

In the first case, the nonlinear polarization responsible for the formation of the diffracted wave \( E_d \) at the frequency \( 2\omega \) is of the form \( P \propto E_s E_1 E_2 \), the phase synchronism condition \( \vec{k}_1 + \vec{k}_2 = \vec{k}_s + \vec{k}_d \) being associated with a decrease of an angle between the diffracted and reading beams relative to that formed by the hologram recording waves. Nonlinear polarization for the scheme of six-wave mixing is represented as \( P \propto (E_s E_1 E_2)^2 \), and the diffracted wave \( E_d \) propagation direction is determined by the phase synchronism condition \( \vec{k}_d = 2\vec{k}_s - 2\vec{k}_1 - \vec{k}_2 \).

Note that in terms of dynamic holography, the matter concerns recording and reading of linear (four-wave mixing) and quadratic (six-wave mixing) dynamic holograms [10]. With opposite propagation directions of the plane reference \( E_1 \) and reading \( E_2 \) waves \( (\vec{k}_1 + \vec{k}_2 = 0) \), the diffracted wave \( E_d \) has characteristics of a phase conjugate to the signal wave \( E_s \), because it is propagating exactly in the counter direction \( (\vec{k}_d = -2\vec{k}_s) \) at the conjugate wave front \( (\phi = -2\phi_s) \). So in this interaction geometry one can realize frequency conversion of complex wave fronts enabling visualization, e.g., of infrared (IR) images [11].
To describe theoretically the energy efficiency of interaction, let us assume that recording of dynamic gratings is realized at the frequency coincident with the absorption line center $S_0 - S_1$ of a dye solution. The medium is absorbing radiation at the frequency $\omega$, being transparent at the doubled frequency $2\omega$. Forming of the wave $E_D$ is determined by diffraction of the reading wave $E_2$ from a thermal dynamic grating recorded by the signal and reference waves.

Taking into consideration the absorption saturation effect in the principal spectral channel $S_0 - S_1$ and induced absorption from the excited channel $S_1$ characteristic for solutions of complex organic compounds (dyes), one can derive expressions for the nonlinear susceptibility of the medium at the fundamental and doubled frequency as follows [12]:

$$\chi(\omega) = \frac{n_0 \kappa_0}{2\pi} \left( \Theta_{l_1} \frac{\hat{a}^2 I - b_s I^2}{1 + JI} \right),$$

$$\chi(2\omega) = \frac{n_0 \kappa_0}{2\pi} \left( a_r I + b_s I^2 \right).$$

Here $\hat{a} = a + i\alpha = (\hat{\Theta}_{l_2} + \hat{\Theta}_{l_1} - \hat{\Theta}_{l_2})/\sqrt{P_{l_1}} - a_r$, $b_s = \sigma_r B_{l_1} (1 - \mu_{l_2})/\sqrt{P_{l_2}}$, $a_r = \sigma_r (1 - \mu_{l_1})$, $J = (B_{l_1} + B_{l_2})/\sqrt{P_{l_2}}$.

In these expressions $\Theta_{l_1}(\omega) = \Theta_{l_1}(\omega) + iB_{l_1}(\omega)$, where the coefficients $\Theta_{l_1}(\omega)$ are related by Kramers-Kronig relations to Einstein’s coefficients for the induced transitions $B_{l_1}(\omega)$, $\kappa_0$ – initial extinction coefficient, $n_0$ – initial refractive index; $\nu = c/n_0$ – speed of light in the medium, $P_{l_1}$ – total probability of spontaneous and nonradiative transitions between the levels $S_0 - S_1$; $\sigma_r = 2\omega (dn/dT) \tau / cC_\rho$, $\tau$ – interaction duration, $C_\rho$ – unit volume thermal capacity, $dn/dT$ – thermo optical coefficient, $\mu_{l_1}$ – luminescence quantum efficiency in the channel $k - l$.

In the approximation of slowly varying light-field amplitudes, the equations for complex amplitudes may be written in the following way:

Figure 1 - Schemes for transformation of light beams on the frequency-nondegenerate four-(a) six-wave (b) mixing.
ORMACHEA, ROMANOV AND TOLSTIK

\[
\begin{align*}
\frac{\partial E_{1s}}{\partial z} &= \frac{i2\pi\omega}{cn_0} \left( \chi_0(\omega) E_{1s} + \chi_{1s}(\omega) E_{2s} \right), \\
\frac{\partial E_{2s}}{\partial z} &= -\frac{i4\pi\omega}{cn_0} \left( \chi_4(2\omega) E_{2s} + \chi_{4s}(2\omega) E_{2s} \right),
\end{align*}
\]

where the components of the nonlinear-susceptibility series expansion in terms of spatial harmonics of a holographic grating are calculated using the Fourier transform \( \chi_n = \frac{1}{2\pi} \int \chi(\xi) \exp[-i(m\xi)]d\xi, \) \( \xi = (\xi_k - \xi_s) \cdot \hat{r}. \) With this system of equations one can describe the process of frequency-degenerate four-wave mixing for \( m=1 \) and also the process of nondegenerate six-wave mixing at \( m=2. \)

Considering the explicit form of expressions for the medium’s nonlinear susceptibility Fourier-components \( \chi_{1s,2s}, \) a system of coupled wave equations (3) may be transformed to

\[
\begin{align*}
\frac{\partial E_{1s}}{\partial z} &= i \frac{k_s}{2} f_{1s} E_{1s}, \\
\frac{\partial E_{2s}}{\partial z} &= -ik_s \left( \psi^{(1)} E_{2s} + \phi^{(1)} E_{2s} \exp[\mp im(\phi_1 - \phi_2)] \right),
\end{align*}
\]

where the following notation is used:

\[
\begin{align*}
f_{1s} &= \frac{\hat{\phi}_{1s} + b_s(I_{1s} + 2I_{2s})}{B_{1s}} - \frac{\hat{\phi} + b_s/J}{J} \left( \frac{1 - 1/A_s}{A_s(J + 1/J + A_s)} \right), \\
\psi^{(1)} &= b_s(I_1 + I_s)/J + (a_s - b_s/J)(1 - 1/A_s)/J, \\
\psi^{(2)} &= (a_s - b_s/J)(1 - 1/A_s)/J, \\
\phi^{(0)} &= \frac{b_s}{J} \sqrt{J_1 I_s} + \frac{2(a_s - b_s/J) I_1 I_s}{A_s(1 + J(I_1 + I_s) + A_s)}, \\
\phi^{(1)} &= -\frac{4(a_s - b_s/J) I_1 I_s}{A_s(1 + J(I_1 + I_s) + A_s)^2}, \\
A_s &= \left[ 1 + 2J(I_1 + I_s) + J^2(I_1 - I_s)^2 \right]^{1/2}.
\end{align*}
\]

The coefficients \( f_{1s} \) and \( f_{2s} \) include the absorption factor and refractive index modulation in the interference field as well as self-diffraction of the reference and signal waves; self-modulation is described by the expressions \( \psi^{(1)} \) and \( \phi^{(0)} \) and \( \phi^{(1)} \) is used to describe the parametric coupling of the reading and diffracted waves in the case of four - \( (m=1) \) and six-wave \( (m=1) \) \( (m=2) \) mixing.

To realize effective reading of the dynamic gratings recorded by the waves \( E_5 \) and \( E_1 \) at the frequency \( \omega \), it is proposed [9] to use the cavity feedback for the reading \( E_2 \) and diffracted \( E_D \) waves at the doubled optical frequency \( 2\omega \).

With due regard for multiple reflections from mirrors of the Fabry-Perot interferometer, an amplitude of the reading wave \( E_2 \) at the output may be found as a sum of the waves representing a geometrical progression

\[
E_{2r} = E_{20}\sqrt{1-R_1} \exp(i\Phi_2) \sqrt{1-R_2} \times \left[ 1 + V\sqrt{R_1R_2} \exp(i2\Phi_2) + V^2R_1R_2 \exp(i4\Phi_2) + \ldots \right],
\]

where \( E_{20} \) – amplitude of the reading wave at the cavity input, \( R_1 \) and \( R_2 \) - reflection factors of the cavity mirrors, \( \Phi_2 = \frac{2\pi}{\lambda} nL \cos \theta_2 \) – phase shift of the wave \( E_2 \) propagating at the angle \( \theta_2 \) to the cavity axis \( z, L \) – cavity length, \( V \) – coefficient determining intensity losses for the wave \( E_2 \) in one pass of the cavity.
And an intensity of the wave \( E_z \) at the cavity output may be determined as

\[
I_{zt} = cn_r |E_{zt}|^2 / 8\pi = I_{20} \frac{(1-R_z)(1-R_i)V}{(1-\sqrt{R_zR_i}V)^2 + 4\sqrt{R_zR_i}V\sin^2(\Phi_r)},
\]

where \( I_{20} = cn_r |E_{20}|^2 / 8\pi \) – intensity of the input reading wave. In case there is no absorption at the frequency \( 2\omega \), losses of the reading wave are governed by its diffraction from the dynamic grating recorded within the interferometer \( V = 1 - \xi_a \), \( \xi_a = I_o(z=0)/I_{20} \) being the diffraction efficiency of dynamic gratings in one pass of a nonlinear layer.

In a similar way, summing up the wave amplitudes due to diffraction of the wave \( E_z \) from the intracavity grating in every pass of the cavity, one can find the diffraction efficiency at the interferometer output as

\[
E_{ot} = E_o(z=0)\sqrt{1-R} \times \left\{1+V\sqrt{R_zR_i}\exp(2i\Phi_o) + V^2\left(R_zR_i\exp(i4\Phi_o) + \ldots\right)\right\}, \tag{7}
\]

where \( E_o(z=0) \) is the amplitude of the diffracted wave in one pass of a nonlinear layer determined by solving a system of equations (4). \( \Phi_o = \frac{2\pi}{\lambda} nL\cos\theta_o \) is the phase shift of the diffracted wave propagating at the angle \( \theta_o \) relative to the cavity axis \( z \). The one-pass diffracted wave amplification is determined by the parameter \( V' = 1 + \xi_a \).

Based on expression (7), the diffraction efficiency of the reading wave \( E_z \) at the intracavity grating may be as

\[
\xi = \left| \frac{E_{ot}}{E_{20}} \right| = \xi_a \frac{(1-R_i)}{(1-\sqrt{R_zR_i}V')^2 + 4\sqrt{R_zR_i}V'\sin^2(\Phi_o)}. \tag{8}
\]

As follows from analysis of expression (8), a maximum gain in the diffraction efficiency at a symmetric configuration of the cavity \( (R_1 = R_2 = R) \) may be attained when the interferometer is tuned to a maximum transmission \( (\Phi_o = 0) \) and, in the region of small \( \xi \), may be evaluated from a simple relation \( \xi = \xi_a / (1-R) \). Thus, due to the cavity coupling for the wave \( E_z \), a dynamic grating may be reread many times with the increased energy contribution from the wave \( E_z \) that is transformed to the diffracted wave \( E_{ot} \).

Numerical modeling for the equations of (4) with regard to expressions (6) - (8) has been performed, using the following spectral and thermo optical characteristics of the medium associated with 3274U dye in ethanol solution, in the assumption of laser excitation into the absorption line center \( S_0 - S_1 \) [14]: \( \lambda = 1064 \text{ nm, pulse length } \tau = 15 \text{ ns, } n_0 = 1.36, \quad dn/dT(C_<0>) = -2 \cdot 10^{-4} \text{ J}^{-1} \text{ cm}^3, \quad B_23 / B_21 = 0.43 \), with Stokes shift assumed to be equal to the absorption line halfwidth \( \Delta \lambda = 100 \text{ nm, luminescence quantum efficiency } \mu_{L12} = 0.01, \mu_{L22} = 0.0001 \).

The calculation results for the diffraction efficiency of dynamic gratings at the frequency \( 2\omega \) are shown in Figure 2 depending on the intensity of the hologram recording waves at different reflection factors of the cavity mirrors. The functions have been obtained when the Fabry-Perot interferometer was tuned to a maximum transmission of the waves \( E_z \) and \( E_D \) \( (\Phi_{z,D} = 0) \). Note a significant improvement in the diffraction efficiency of intracavity dynamic gratings as compared to the classical scheme without a cavity, particularly in the region of low intensities, where the use of high-reflection mirrors \( R \approx 0.9 \) leads to a diffraction efficiency increase approximating the above estimate \( \xi = \xi_a / (1-R) \).

Another feature of using the intracavity schemes for dynamic hologram recording is an enhancement of their angular and spectral selectivity [6] because of the increasing effective length of the interacting light beams. Let us compare the dynamic hologram selecting properties for the above-mentioned schemes of intracavity and off-cavity four- and six-wave mixing. A system of wave equations (4) in the case of minor deviation from the phase synchronism conditions may be rewritten as
where the deviation of a reading wave from the exactly fulfilled Bragg diffraction condition is determined by the parameter

\[
\Delta k = \frac{2\omega}{c} \theta \Delta \theta.
\]

Figure 3 presents the diffraction efficiency of dynamic gratings as a function of the angle of the reading wave deviation from the Bragg resonance conditions \(\Delta \theta\) obtained from numerical solution of a system of wave equations (9) for the cases of four- and six-wave mixing in off-cavity (a) and intracavity (b) geometries. All the functions are normalized to their maximum values. The calculations are performed using the interferometer parameters \((L = 500 \mu m, R_1 = R_2 = 68\%)\) involved subsequently in the experimental studies.

The following features of the functions obtained are noticeable. First, an angular width of the resonance peak for the case of six-wave mixing \((\Delta \theta_{3s} \approx 10 \text{ mrad})\) is only a half of that in the process of four-wave mixing \((\Delta \theta_{3s} \approx 20 \text{ mrad})\) (Figure 3a) due to the doubly decreased period of the volume dynamic hologram participating in the process of Bragg diffraction upon the associated change in the diffraction order. With the parameters used \((\Lambda \approx 10 \mu m, L = 500\mu m)\), the derived values are in a good agreement with the well-known expression for the angular selectivity of
holographic gratings: \( \Delta \theta_s \approx \frac{\Lambda}{L} \) (\( \Lambda = \frac{\lambda}{2 \sin \theta} \) - grating period) that is valid for small deviations in propagation of the reading wave from Bragg condition and for lacking of absorption at a frequency of the reading wave [14]. Second, the use of the interferometer with the reflection factors of mirrors \( R_1 = R_2 = 68\% \) results in a practically two-fold narrowing of the angular selectivity peak for dynamic gratings (Fig.3b) caused by the increased effective interaction length of light beams following from expression (6): \( L_{\text{eff}} \approx L_0 \frac{1}{\sqrt{R_1 R_2}} \). On the one hand, this situation may lead to the degraded transformation quality of complex wave fronts but, on the other hand, it may be employed to realize multiplex recording of dynamic holograms.

### 3. EXPERIMENTAL STUDY OF THE PROCESS OF IMAGE FREQUENCY CONVERSION

An experimental setup used to study the frequency-nondegenerate interaction in a nonlinear Fabry-Perot interferometer is demonstrated in Fig. 4a. The study was performed using the ethanol solution of 3274U dye, the fundamental- (\( \lambda = 1064 \) nm) and second (\( \lambda = 532 \) nm) harmonic radiation of yttrium-aluminum garnet laser \( I \) (\( \tau = 15 \) ns). The absorption band of 3274U dye is exhibited at the fundamental lasing frequency (saturation

![Experimental setup](image)

**Figure 4** - (a) – experimental setup; (b) – diffraction efficiency versus intensity of the waves recording dynamic gratings; (1) – off- and (2) – intra -cavity four-wave mixing.
intensity $I_{\text{Sat}} = 13$ Mw/cm$^2$, molecular lifetime in the excited singlet state $\tau_{s} \approx 10$ ps [15]), the dye being practically transparent at the second-harmonic frequency and enabling one to record dynamic holograms in the IR spectral region with their reconstruction in the visible. The selected diffraction scheme of laser radiation in the Bragg mode is realized with the co-propagating signal and reference waves. Diaphragm 2 cuts the spatially homogeneous portion of radiation from the yttrium-aluminum garnet laser. The signal $E_S$ and reference $E_1$ waves are formed by mirrors 3, 8, and 9. Owing to the angle $2\theta = 90$ mrad formed between propagation directions of the reference and signal beams, the interacting waves may be overlapping over the whole cell length. With the help of mirror 4, the reading wave $E_2$ is directed into the nonlinear medium 7. The energy efficiency of the interacting light beams is measured by recording system 5, 11. Light filters 6, 10 are used to change the power of laser radiation.

The main task of this experimental study is comparison between the diffraction efficiencies of dynamic gratings when nondegenerate multiwave mixing is realized in the off-cavity scheme as well as with the use of a Fabry-Perot interferometer. To this end, we use a cell with the thickness $L=500\mu$m and Fabry-Perot interferometer with the same cavity base and mirror reflection factors $R_1 = R_2 = 68\%$ at a wavelength of 532 nm. An optical thickness of the 3274U dye solution at $\lambda = 1064$ nm in both cases comes to $k_L L = 1$. Note that the interferometer is intended for the radiation wavelength $\lambda = 532$ nm, whereas (signal $E_s$ and reference $E_1$) waves of the fundamental lasing frequency are transmitted through the nonlinear medium without the cavity feedback. The diffraction efficiency of holographic gratings is measured using for recording the spatially homogeneous light beams of the same intensity.

Owing to the performed study, the prospects for improvement of the dynamic-grating diffraction efficiency have been demonstrated experimentally using the schemes of intracavity four- and six-wave mixing. Figure 4b shows the diffraction efficiency ($\xi = I_o(z = 0)/I_o(z = L)$) as a function of the intensity of the dynamic-grating recording waves $E_s$ and $E_1$ in case of four-wave mixing. As seen, an increase in the diffraction efficiency with growing intensity is practically quadratic, amounting to a few tens percent. In the process, the use of intracavity interactions (curve 2) enables one to double or treble the diffraction efficiencies compared to the off-cavity case (curve 1), making it possible to attain experimental diffraction efficiencies $\approx 13.5\%$.

Solid lines in Figure 4b represent the results of numerical modeling based on a system of equations (4), (8) with the radiation and nonlinear interferometer parameters meeting the experimental conditions. By the introduction of the effective interaction time ($\tau_{\text{eff}} = \tau/\sqrt{3}$) during the calculations one can take account of the fact that the numerical solution gives a value of the diffraction efficiency by the end of a pulse, while the experiment provides the pulse-averaged diffraction efficiency. By this assumption it is possible to have a reasonable agreement between the experimental data and theoretical results.

Fairly high experimental diffraction efficiencies make it possible to realize frequency conversion of the coherent images formed when the amplitude transparencies are brought about into the signal beam. With the use of both configurations for the light beam interactions (four- and six-wave mixing), the IR-to-visible conversion of images has been realized experimentally, Figure 5. A dynamic hologram of the image (1) formed at the wavelength $\lambda = 1064$ nm is recorded by the reference $E_1$ and signal $E_s$ light beams. Depending on the interaction geometry, reading of the hologram by the beam $E_1$ at $\lambda = 532$ nm enables reconstruction of the first- (2) or second-order (3) diffraction, in the latter case with simultaneous phase conjugation at the doubled optical frequency.

![Figure 5](image-url) - Visualized IR image (1) in the case of four- (2) and six-wave (3) mixing.
4. CONCLUSIONS

The theoretical and experimental studies performed demonstrate that using of the schemes for intracavity light-beam interactions permits the diffraction efficiency of intracavity dynamic gratings to be improved considerably as a result of a constructive interference on multiple diffraction of the reading wave in a nonlinear layer of the cavity. High diffraction efficiency (up to 13.5%) with simultaneous infrared-to-visible frequency conversion of coherent images has been experimentally obtained by intracavity nondegenerate four-wave mixing.

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