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Four-wave mixing in microresonators with resonance and thermal nonlinearities

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ABSTRACT

The results of theoretical and experimental investigations into four-wave mixing in microresonators with resonant and thermal nonlinearities are presented. Theoretical models of dynamically-induced optical gratings have been derived in two cases of the light beams interaction: when a nonlinear medium is incorporated into Fabry-Perot interferometer and for the off-cavity configuration. Experimental realization of intracavity four-wave mixing performed using ethanol solution of Rodamine 6G as a nonlinear medium. It is shown that the use of nonlinear interferometer increases significantly an efficiency of dynamic optical gratings. The potentialities for obtaining of high efficiency conversion of optical information owing the spectroscopic properties of multilevel resonant media are discussed.

Keywords: Four-wave mixing, dynamic gratings, microresonators, nonlinear interferometer, resonant media, dye solutions.

1. INTRODUCTION

Nonlinear interferometers are typical elements of optical data processing systems, exhibiting various self-organization effects due to nonlinearity of interaction between laser radiation and intracavity medium[1-3]. They provide the magnification of nonlinear optical phenomena that can be used, e.g., in dynamical holography[4-6].

In this work experimental and theoretical studies of an intracavity four-wave mixing in complex molecular media have been performed when a nonlinear system is represented by Fabry-Perot microresonator. For theoretical description of typical experimental situations we used the round-trip model of nonlinear interferometer adapted for the geometry of degenerate four-wave mixing[7, 8], which can be realized in the scheme of symmetrical oblique incidence of pump, signal and probe beams to the front and back mirrors of cavity. The model includes the consideration of resonant and thermal nonlinearity of organic dye solutions that gives rise to light-induced dynamic gratings of absorption coefficient and refractive index. The conditions of magnification of dynamic gratings efficiency due to contribution from interference of pumping and reading light beams have been studied experimentally and by means of theoretical modeling.

2. NONLINEAR RESPONSE OF ORGANIC DYE SOLUTIONS ON MONO-PULSE LASER EXCITATION

Further analysis will be performed taking two- and three-level models for the resonant medium that includes nonradiative and radiative transitions both in the principal and excited singlet channels ($S_0 - S_1$ and $S_1 - S_2$). As the experimental intensity of interacting waves is, as a rule, considerably lower than the saturation intensity of the excited channel, we allow for saturation of the main resonance transition but in conditions of linear absorption from the excited level. Based on a system of kinetic equations for the level population and dispersion relations relating changes in the

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refractive index and absorption coefficient, the total nonlinear (resonance and thermal) susceptibility of the medium for the systems under consideration may be given in the following forms[9, 10]

$$\chi_{nl} = \frac{n_0 \kappa_0}{2\pi}\left(\frac{\hat{\theta}_{12}}{B_{12}} - \frac{\hat{\alpha}(\omega) I}{1 + JI}\right), \tag{1}$$

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

where $\hat{\alpha}(\omega) = \frac{\left(\hat{\theta}_{12}(\omega) + \hat{\theta}_{21}(\omega)\right)}{vP_{21}}$ for a two-level model of resonant medium (1) and $\hat{\alpha}(\omega) = \frac{\left(\hat{\theta}_{12}(\omega) + \hat{\theta}_{21}(\omega) - \hat{\theta}_{23}(\omega)\right)}{vP_{21}} - \sigma_T(1 - \mu_{21})$, $b_T = \sigma_T \frac{B_{23}}{vP_{21}}(1 - \mu_{32})$, $J = \frac{B_{12} + B_{21}}{vP_{21}}$ for a three-level model that takes into account linear absorption form the excited level S_1 and thermal nonlinearity due to nonradiative transitions. In these equations $B_{kl}(\omega)$ – Einstein coefficients for the induced transition $k - l$; v - light velocity in the medium, κ_0 - initial extinction coefficient, n_0 – refractive index; P_{kl} – total probability of spontaneous and nonradiative transitions in channel $k - l$, $\hat{\theta}_{kl}(\omega) = \theta_{kl}(\omega) + iB_{kl}(\omega)$ (coefficients $\theta_{kl}(\omega)$ are related by Kramers-Kronig relations with Einstein coefficients $B_{kl}(\omega)$), $\sigma_T = 2\omega(dn/dT)\tau / cC_\rho$, τ – interaction duration, C_ρ – heat capacity for the unit volume, dn/dT – thermo-optical coefficient, μ_{kl} – luminescence quantum yield in channel $k - l$.

3. THEORETICAL MODEL OF INTRACAVITY FOUR-WAVE MIXING

Let us consider now the scheme of intracavity four-wave mixing (FWM) upon normal incidence of pump beams on the mirrors of Fabry-Perot interferometer and oblique incidence of signal beam. Under these conditions the coupled-mode equations have the form[11]

$$\frac{\partial E_{1,2}}{\partial r} = \pm \frac{i2\pi\omega}{cn_0}\left\{\chi_{0,0} E_{1,2} + \chi_{\pm1,\pm1} E_{2,1} + \chi_{0,\pm1} E_{S,D} + \chi_{\pm1,0} E_{D,S}\right\}, \tag{3}$$

$$\frac{\partial E_{S,D}}{\partial r} = \pm \frac{i2\pi\omega}{cn_0}\left\{\chi_{0,0} E_{S,D} + \chi_{\pm1,\mp1} E_{D,S} + \chi_{0,\mp1} E_{1,2} + \chi_{\pm1,0} E_{2,1}\right\}, \tag{4}$$

where $\chi_{m,n} = \frac{1}{(2\pi)^2}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\chi(\zeta_1,\zeta_2)\exp[-i(m\zeta_1 + n\zeta_2)]d\zeta_1 d\zeta_2$, $\zeta_1 = \vec{k}_1 \cdot \vec{r}$, $\zeta_2 = \vec{k}_2 \cdot \vec{r}$. The components of Fourier expansion of nonlinear susceptibility $\chi_{\pm1,\pm1}, \chi_{\pm1,\mp1}$ are responsible for self-diffraction of the fields E_1, E_2, and E_S, E_D, respectively; the components $\chi_{0,\pm1}, \chi_{\pm1,0}$ take into account the processes of self- and cross-modulation, and $\chi_{0,0}$ are responsible for nonlinear absorption. The boundary conditions for the coupled-mode equations in geometry of interaction read as follows:

$$\bar{E}_1(0) = \bar{E}_{10}\sqrt{1-R_1} + \bar{E}_2(0)\sqrt{R_1},$$
$$\bar{E}_S(0) = \bar{E}_{20}\sqrt{1-R_1},$$
$$\bar{E}_2(L) = \bar{E}_1(L)\sqrt{R_2} + \bar{E}_{20}\sqrt{1-R_2},$$
$$\bar{E}_D(L) = 0, \tag{5}$$

where $\bar{E}_j = \frac{1}{2}\bar{A}_j(z)\exp[i(\bar{k}_j\bar{r} - \omega t + \varphi_j)] + c.c.$ Taking into account the expressions for Fourier components of nonlinear susceptibility for two-level resonant medium (1) or for three-level medium (2), the system of coupled-mode equations can be transformed to the system for real amplitudes and phases of the light fields. After, the solution can be obtained from numerical modeling with the use of standard numerical procedures (e.g. Runge-Kutt method for solving nonlinear differential equations and shooting method for adjustment of solutions on opposite boundaries of cavity).

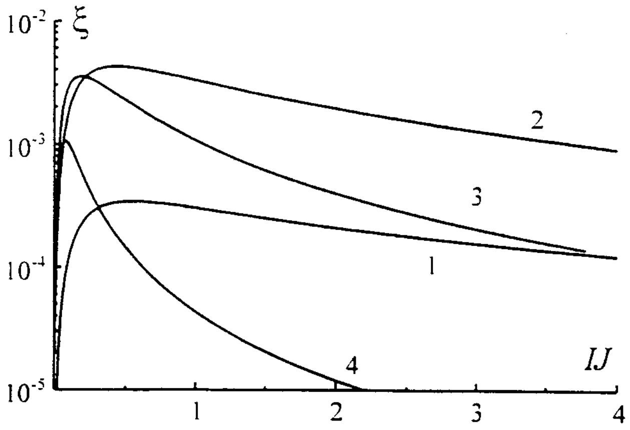


Fig. 1. Diffraction efficiency ξ versus the intensity of pump wave for intracavity four-wave mixing. $\eta = 0$. $k_0L = 0.1$. initial detuning from resonance peak of interferometer $\Phi_0 = 0$. reflection coefficients of cavity mirrors $R = 0(1), 0.1(2), 0.5(3), 0.75(4)$.

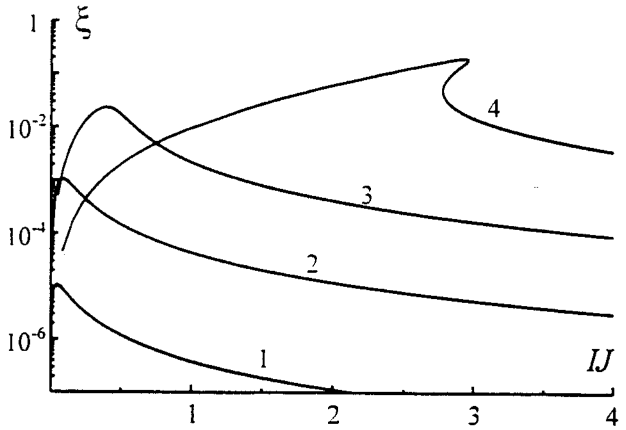


Fig. 2. Diffraction efficiency ξ versus the intensity of pump wave for intracavity four-wave mixing. $\eta = 0$. $k_0L = 0.01(1), 0.1(2), 0.5(3), 2(4)$. $R = 0.75$. $\Phi_0 = 0$.

Fig. 1 – 3 present the results from numerical analysis of coupled equations (3), (4) taking account of expansions for nonlinear susceptibility for two-level resonant medium. In case of intracavity four-wave mixing a considerable increase in the diffraction efficiency $\xi = I_D(r=0)/I_2(r=L)$ is observed (Fig.1) at the reflection coefficient of mirrors that correspond to the case of bad-finesse cavity (curves 2 – 3), which seems to be due to the increased efficiency of intracavity dynamic gratings of absorption coefficient. For high-finesse cavity (curve 4) the efficiency of intracavity four-wave mixing is decreased for high intensity level due to narrowing of transmission peak of interferometer. Note, that at any reflection coefficient of the mirrors the increase of diffraction efficiency of intracavity dynamic grating can be one order of magnitude higher relatively to off-cavity four-wave mixing.

The results of Fig.1 is obtained for low values of optical density k_0L. Increasing optical density leads to increasing efficiency of distributed feedback due to scattering of the light fields on dynamic gratings of absorption coefficient and (or) reflection index. This may lead to the new regimes of intracavity four-wave mixing, namely, optical bistability mode. We obtained the mode of optical bistability both for the intensity of pump beams, signal and conjugated beams (Fig.2, curve 4).

Detuning of optical frequency from the center of absorption band of resonant medium leads to effective modulation of refractive index. This may be used for obtaining high values of diffraction efficiency of dynamic grating (Fig.3,

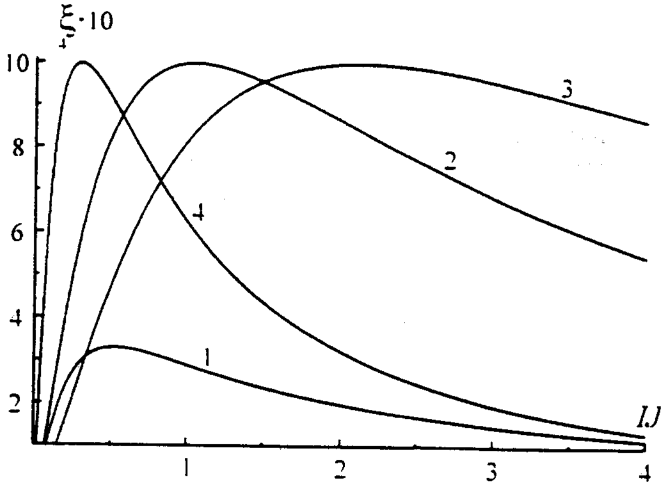
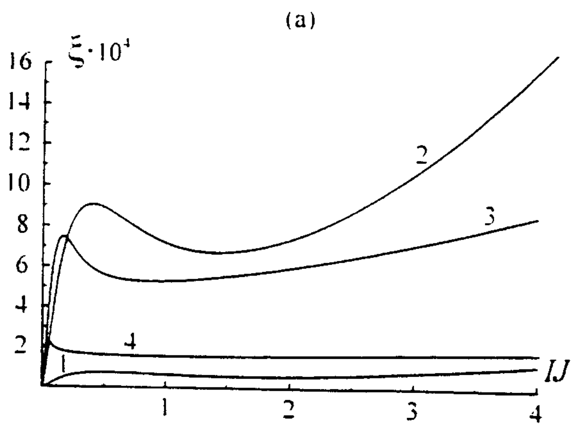


Fig. 3. Diffraction efficiency ξ versus the intensity of pump wave for intracavity four-wave mixing. $\eta = 1$, $k_0L = 0.05$, $R = 0(1), 0.75(2-4)$, $\Phi_0 = 0(2), 0.5(3), -0.5(4)$.

curve 1). The use of optical cavity in this situation provides an additional possibility for controlling of diffraction efficiency of intracavity dynamic gratings of refractive index (Fig.3, curves 2 – 4). As it is seen, maximum diffraction efficiency may be obtained for different intensities of input light beams depending on the relation between initial detuning from the transmission peak of interferometer, linear and nonlinear changes of refractive index of intracavity medium. In particular, the optimal intensity can be decreased due to initial detuning out of resonance transmission of interferometer (Fig.3, curve 4).

Let us consider further a more realistic model of optical nonlinearity for organic dye solutions that takes into account a linear absorption from the excited energy level S_1 and thermalization of energy due to nonradiative transitions. The modeling of an intracavity four-wave mixing process is based on numerical solution of the set of coupled-mode equations (3) - (4) with the use of Fourier expansion of nonlinear susceptibility (2) in the row of dynamic gratings formed in the medium volume.

The results of numerical solution of the set of coupled-mode equations are presented in Fig.4, where the diffraction efficiency of intracavity dynamic gratings is calculated in dependence of the recording beams intensity for different values of reflection coefficients of cavity mirrors. The calculation is performed with the use of spectral and thermo-optical characteristics of ethanol solution of Rodamine 6G supposing laser excitation in the centre of absorption band $S_0 - S_1$ and for pulse duration $\tau = 10\ ns$. As it is seen from Fig.4, a, it is possible to increase the diffraction efficiency of dynamic gratings in approximately one order of magnitude using the cavity configuration instead of usual scheme of four-wave mixing. Further optimization of the diffraction efficiency is related to proper choice of initial cavity detuning out of resonance transmission peak, see Fig.4, b. It should be noted also, that high values of diffraction efficiency obtained at high input intensity, when we compare with the case of two-level resonant medium (Fig.3), are due to effective recording of thermal gratings of refractive gratings under absorption from the excited energy level S_1.



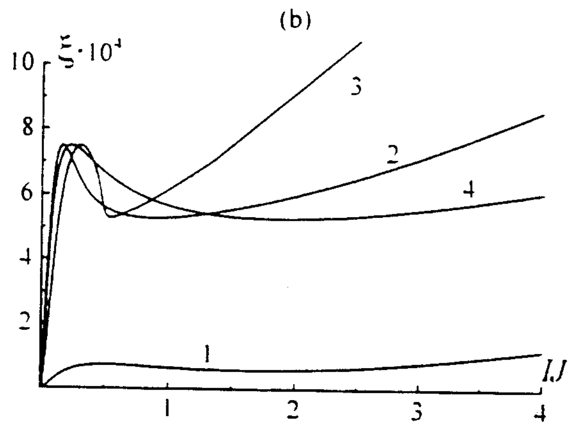


Fig. 4. Diffraction efficiency ξ versus the intensity of pump wave for intracavity four-wave mixing (three-level model). $\eta = 0$, $k_0L = 0.05$, (a): $\Phi_0 = 0$, $R = 0(1), 0.1(2), 0.5(3), 0.75(4)$; (b): $R = 0(1), 0.5(2-4)$, $\Phi_0 = 0(2), 0.5(3), -0.5(4)$

4. EXPERIMENTAL REALIZATION OF DYNAMIC GRATINGS RECORDING IN THIN FABRY-PEROT INTERFEROMETER

In Fig.5 the experimental setup for realization of four-wave mixing is shown. Dynamic gratings have been recorded by second-harmonic radiation of YAG laser *1* ($\lambda = 532\,nm$) with pulse duration $\tau = 15\,ns$. Interacting waves have been formed by mirrors *3, 4, 7, 8*. The typical angle between the signal and reference waves was 90 *mrad* and intensities of all interacting waves are related as $0.5I_2 = I_1 = 2I_S = I$. Ethanol solution of Rhodamine 6G dye in cell *5* has been used as a nonlinear medium. In case of counter-propagating reference E_1 and reading E_2 waves the diffracted wave E_D was exactly meeting signal wave E_S and the wave front conjugation was realized with degenerate four-wave mixing (phase synchronism for wave vectors $k_D = k_1 - k_S + k_2$, nonlinear polarization $P \sim \chi^{(3)} E_1 E_2 E_S^*$). The diffraction efficiency was measured by a recoding system *6*.

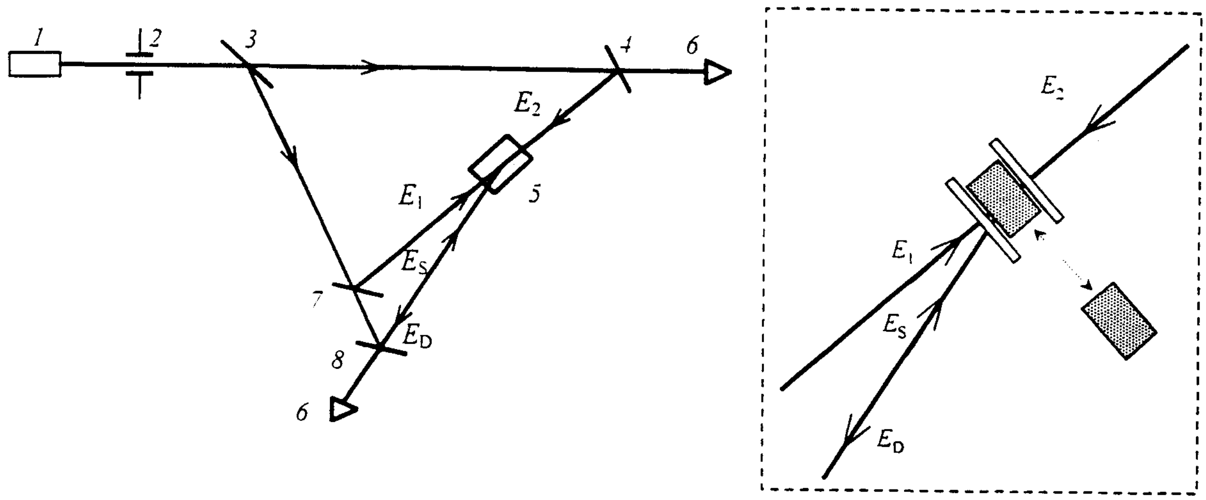


Fig. 5. Experimental setup for realization of four-wave mixing.

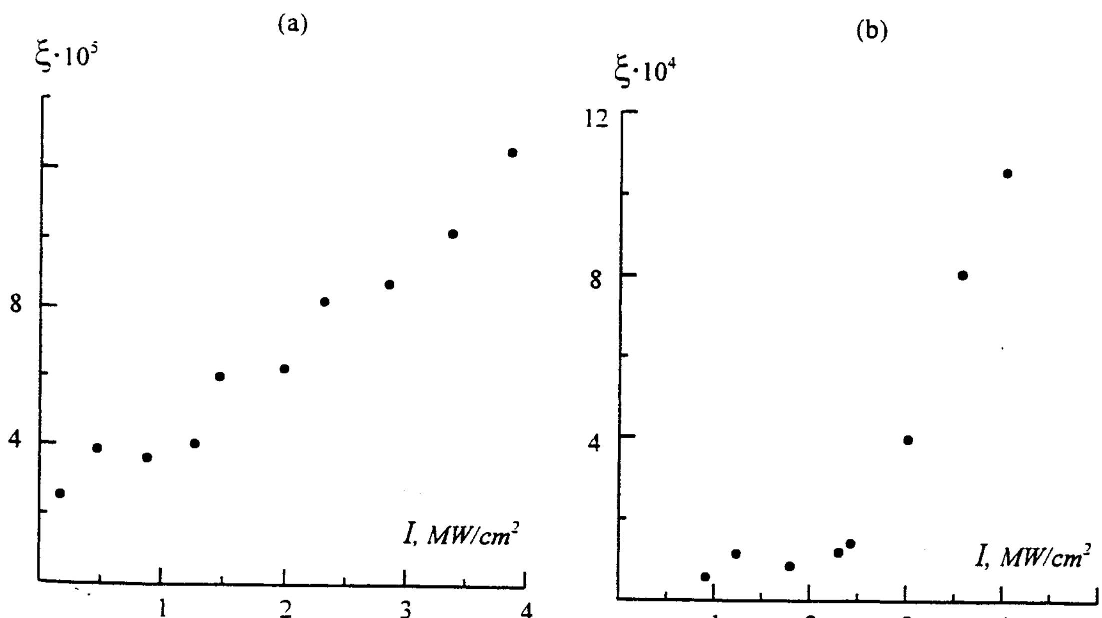


Fig.6. Diffraction efficiency ξ versus the intensity of pump wave for (a) – off-cavity FWM and (b) - intracavity FWM.

We performed two series of experiments to compare the usual scheme of FWM with intracavity configuration. Thickness of the cell with ethanol solution of Rodamine 6G was about $L = 100\,\mu m$ and concentration of dye was $C = 7 \cdot 10^{-3}\, g/l$. The cavity of Fabry-Perot type with the same thickness was performed using the mirrors with reflection coefficients $R_1 = 98\%$ and $R_2 = 73\%$. The results of measured diffraction intensity are presented in Fig.6 both for off-cavity four-wave mixing (a) and for the case of intracavity interaction (b). As it is seen, the use of cavity configuration increases significantly (in one order of magnitude) the diffraction efficiency dynamic gratings in a good agreement with theoretical predictions.

5. CONCLUSION

In this paper the conditions for amplification of dynamic gratings due to the interference effects taking place on four-wave mixing in microresonatotrs is established. Owing to the resonator feedback, the diffraction efficiency of dynamic holograms recorded in thin-layers of dye solutions is improved by an order of magnitude. The role of resonance and thermal dynamic gratings in the process of intracavity four-wave mixing is analyzed. It is demonstrated that over the nanosecond range of laser pulse lengths at the intensities above the absorption intensity of the principal resonance transition the main contribution to the effective energy exchange between the waves is made by thermal dynamic gratings. Enhancement of the diffraction efficiency of dynamic gratings studied in this paper may be used for the development of highly effective radiation transformation methods such as visualization of IR images by nondegenerate multiwave mixing, polarization holography, etc.

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