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Saturation in Principal Channel for Nonlinear Media with Stokes Shifting Emission Bands

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Abstract: The phase response of nonlinear media is very usefull to study nonlinear processes such as : four-wave mixing, holography and amplification of laser's irradiations. Using a typical three-level configuration (S_0-S_1-S_2) for nonlinear media, the transitions of molecules in principal channel (S_0-S_1) can be realized by light fields of intensity I_{12} at frequency ω_0. At the same time, light fields with intensity I_{23} at frequency ω interact with excited molecules to realize their transitions in excited channel (S_1-S_2). The cubic nonlinearity of dye solutions is taken into consideration to study the saturation processes of nonlinear media with Stokes emission bands. The saturation of phase response of dye solution in principal channel (S_0-S_1) is realized at saturation intensity I_{12}^{sat}. The value of saturation intensity I_{12}^{sat} in principal channel decreases with increasing of radiation intensity in excited channel I_{23}. The saturation intensity I_{12}^{sat} has its optimum (minimum) values, when the frequency of light fields in principal channel is tuned with Stokes shift from the centre of principal absorption band. In addition, the saturation intensity I_{12}^{sat} has its optimum, when the radiations in excited channel have enough big intensity I_{23} and have a frequency tuning with anti-Stokes shift from centre of absorption excited band.

Key words: Saturation, principal channel, Stokes shift, dye solutions

INTRODUCTION

A cubic nonlinearity of nonlinear media represents a basic information to study nonlinear processes such as: four-wave mixing, amplification and holography[1-7]. A big attention was paid to the interaction of light beams with nonlinear media. This interaction appears in many nonlinear processes: interference, saturation of refractive index, bistability, phase-conjugation and others[8-11]. For this study, it is necessary to use three-level model for nonlinear media. The lifetimes of vibration energy levels of dye solutions are significantly lower than the lifetimes of electronic energy levels[12]. In this case, the electronic states can be taken as homogeneously broadened levels, which gives ability to use three-level model with averaged Einstein coefficients for many nonlinear medium[8]. This model for nonlinear media gives ability to control the nonlinear properties, which realized by independent light beams (optical pumping) acting in principal (excited) channel[8,9,13]. In three-level configuration the dye solution can be excited by light fields with two different frequencies: one group of light fields (with intensity I_{12} at frequency ω_0) acts in principal channel (S_0-S_1), and other group (with intensity I_{23} at frequency ω) acts in excited channel (S_1-S_2). Light fields in one channel can involve nonlinear processes, and other light field (optical pumping) acts in second channel[13]. The refractive index, extinction coeficient, absorption and emission of nonlinear media depend on the intensity and frequency of light fields acting in each of principal and excited channels.

The phase response of nonlinear media has a saturation character of intensity of light beams in principal and in excited channels[8,14]. The saturation processes is studied for dye solutions with coincident absorption and emission bands[14].

The aim of this theoretical study is to make an optimization of saturation processes in nonlinear media with Stokes shifting emission bands in both principal and excited channels, and to get theoretical analysis for optimal conditions of intensity and frequency tuning of radiations in both principal and excited channels.

THEORY

The saturation intensity I_{12}^{sat} in principal channel is defined as the value of radiation intensity, acting in principal channel, for which the absorption is decreasing in half of its initial value ($K_{12}(I_{12}^{sat}) = (1/2)\, K_{12}\, (I_{12}=0)$). The extinction coefficient in principal channel at frequency ω_0 can be found by the following expression:

$$\chi_{12}(\omega_0) = \frac{c}{2v} K_{12}(\omega_0) \quad (1)$$

Where, is the $K_{12}(\omega_0) = \frac{\bullet\omega_0}{v}(N_1 B_{12}(\omega_0) - N_2 B_{21}(\omega_0))$ - is the absorption coefficient in principal channel, N_i - is the population of i^{th} - energy level; P_{ij} - is the total probability of spontaneous and radiationless transitions in the i-j channel; v=c/n - is the light velocity in the nonlinear medium. The Einstein coefficients $B_{12}(\omega_0)$; $B_{21}(\omega_0)$ - are determined at the frequency of radiations ω_0 in principal (S_0-S_1) channel. Using the balance equations under a double frequencies excitation of dye solution modeled by three-level configuration and Eq. 1 the extinction coefficient will be:

$$\chi_{12}(\omega_0) = \chi_0(1 + aI_{23})/K \qquad (2)$$

Where, $K=1+JI_{12}+aI_{23}+bI_{12}I_{23}$; $J=(B_{12}+B_{21})/vp_{21}$; $a=B_{32}/vp_{32}$; $b=B_{12}B_{23}/v^2p_{21}p_{32}+aJ$; $\chi_0=N\bullet cB_{12}(\omega_0)/2v$ - is the linear extinction coefficient; $B_{23}(\omega)$; $B_{32}(\omega)$ - are determined at frequency of radiations ω in excited channel.

The extinction coefficient, included in Eq. 2, has a monotonic proportionality with intensity of radiations in each channel (I_{12} and I_{23}) and has its maximum value ($\chi_{12}=\chi_0$) at $I_{12}=I_{23}=0$. The extinction coefficient has the half of its maximum value at saturation intensity in principal channel (I_{12}^{sat}) with value:

$$I_{12}^{sat} = \frac{1 + aI_{23}}{J + bI_{23}} \qquad (3)$$

From Eq. 3 the saturation intensity (I_{12}^{sat}) in principal channel has a monotonic dependence on radiation intensity in excited channel (I_{23}).

To study the saturation processes in principal channel, let us take into consideration a nonlinear medium with a gaussian form of mirror-symmetric absorption and emission bands on Stokes shift by δ_{ij} of the profile

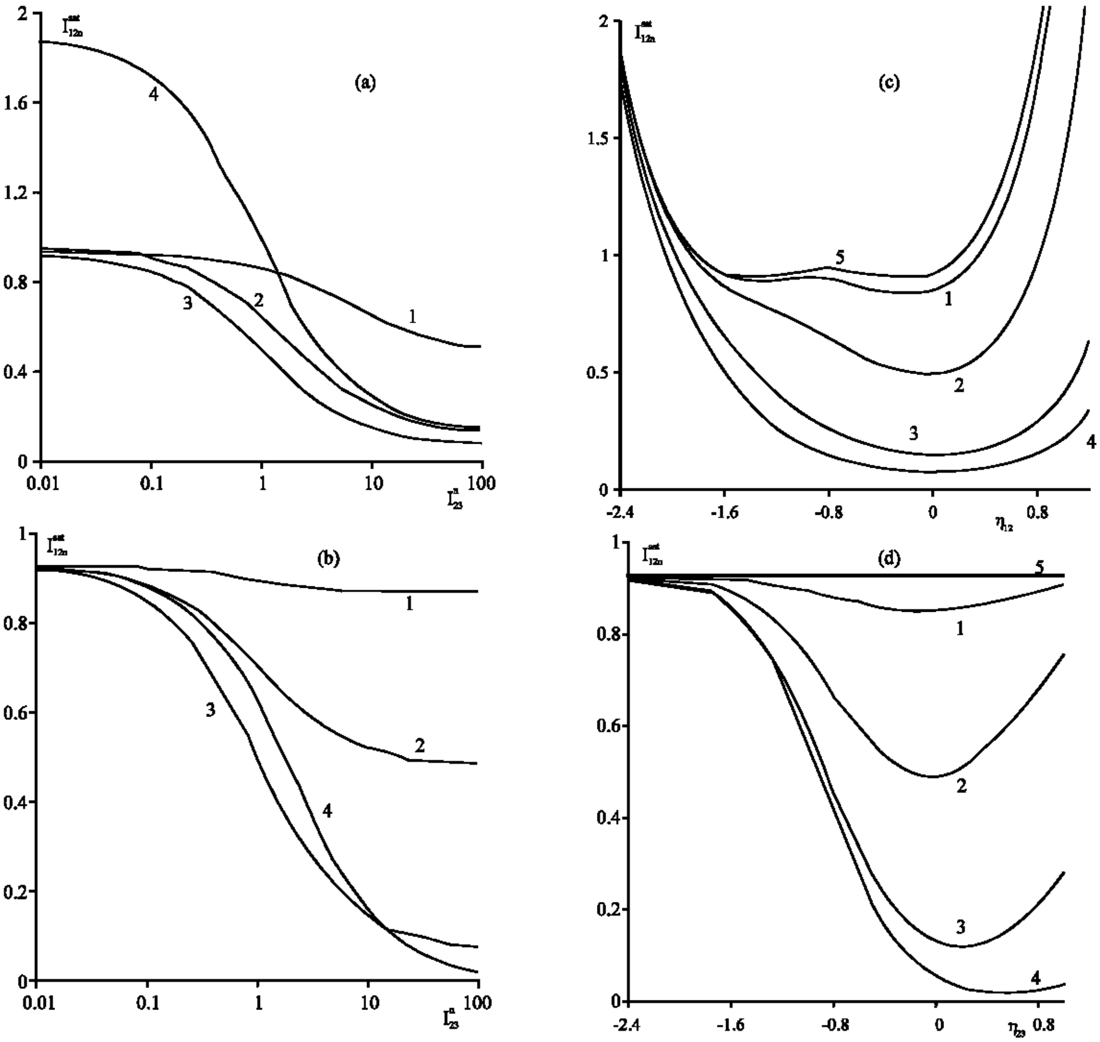

Fig. 1: Dependence of saturation intensity I_{12n}^{sat} on: Radiation intensity I_{23}^{n} (a) and (b); frequency tuning of radiation in principal η_{12} (c); in excited η_{23} (d) channels. Curves: 1, 2, 3 and 4 at: (a) η_{12}: - 1.6, - 0.8, 0 and 0.8; (b) η_{23}: - 1.6, - 0.8, 0 and 0.8; (c) and (d) I_{23}^{n} : 0.1, 1, 10 and 100, respectively. Where, for: (b), (d) η_{12}= 0; (a), (c) η_{23}= 0. Curve 5 is taken for two-level model.

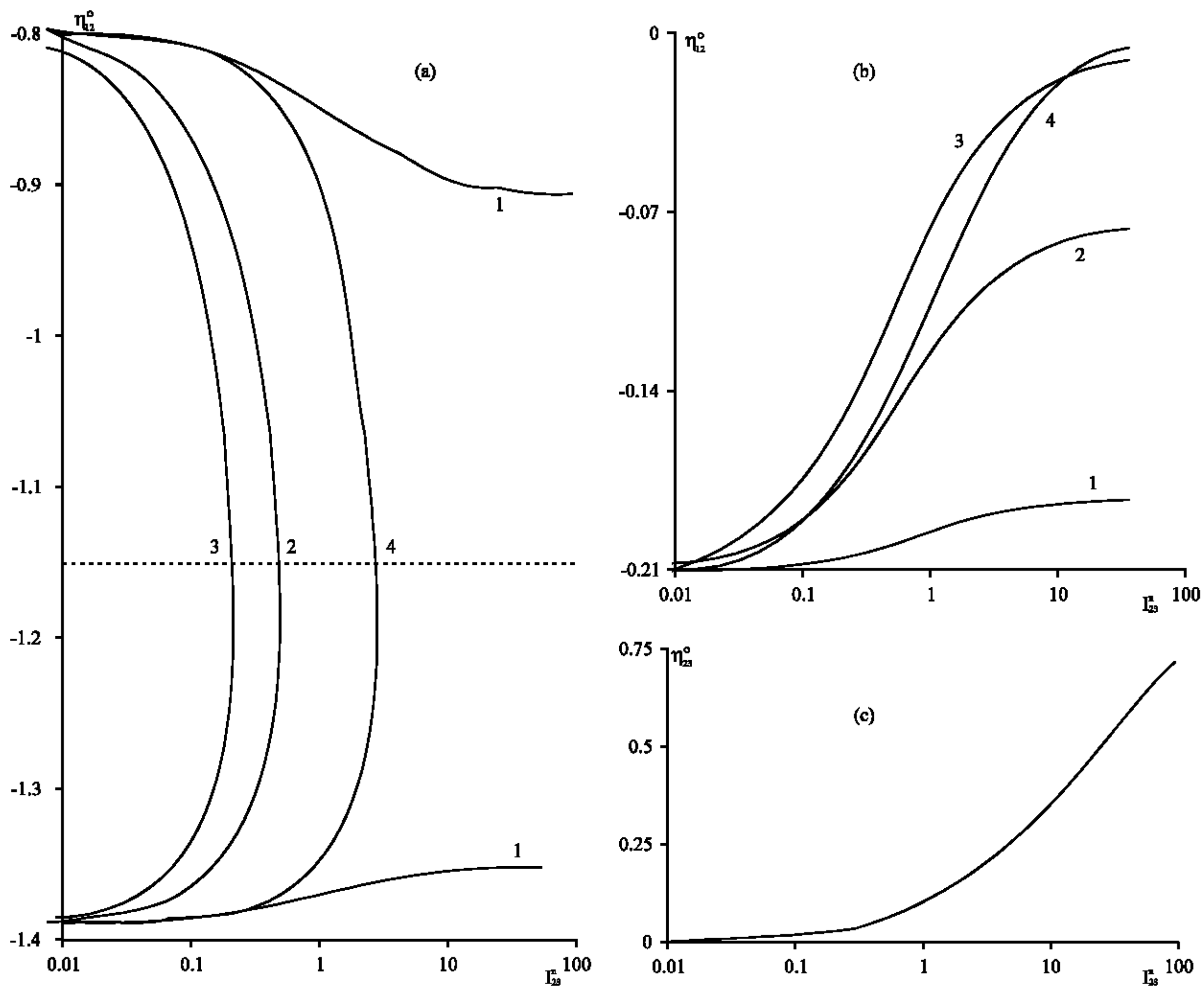

Fig. 2: Dependence of frequency tuning: (a) and (b) η_{12}^{0} in principal; (c) η_{23}^{0} in excited channels on radiation intensity in excited channel I_{23}^{n}. Where curves: 1, 2, 3 and 4 are taken for frequency tuning of radiations in excited channel η_{23} : - 1.6, - 0.8, 0 and 0.8, respectively

halfwidth Δ_{ij}. Where, $\delta_{ij}=(\omega_{ij}-\omega_{ji})/\Delta_{ij}$, ω_{ij} - is the centre of i-j band. For this matter the frequency tuning of radiations in principal ($\eta_{12}=(\omega_0-\omega_{12})/\Delta_{12}$) and excited ($\eta_{23}=(\omega-\omega_{23})/\Delta_{23}$) channels are used to find Einstein coefficients B_{ij}. Saturation intensity (I_{12}^{sat}) for this matter has extremum values at frequency tuning of radiations:

$$\eta_{12}^{0} = -\delta_{12}(B_{21}/vp_{21})\frac{1+aI_{23}}{J+bI_{23}} \quad (4)$$

in principal channel and

$$\eta_{23}^{0} = \delta_{23}(aI_{23}) \quad (5)$$

in excited channel.

In Fig. 1 and 2 the following statements have been considered: the absorption and emission bands are on Stokes shift with values $(\omega_{ij}-\omega_{ji})=\delta_{ij}\Delta_{ij}=1.6\Delta_{ij}$, the maximum values of Einstein coefficients are the same for all bands $B_{12}^{max}=B_{21}^{max}=B_{23}^{max}=B_{32}^{max}$. The radiations intensities are normalized to the value vp_{21}/B_{12}^{max} and vp_{32}/B_{32}^{max} in principal and excited channels, respectively ($I_{12n}^{(sat)}=I_{12}^{(sat)}B_{12}^{max}/vp_{21}$; $I_{23}^{n}=I_{23}B_{32}^{max}/vp_{32}$). Eq. 3 is demonstrated in Fig. 1 for three-level model by curves 1, 2, 3 and 4 and for two-level model ($I_{12}^{sat}=\frac{1}{J}$, I_{23}) by curve 5.

Figure 1 shows the monotonic dependence of saturation intensity I_{12}^{sat} on radiation intensity in excited channel I_{23}. Equation 3, 4 and Fig. 1 show that the optimum conditions for saturation intensity in principal channel (I_{12}^{sat}), small saturation intensity, are realized for: frequency tuning of radiations $\eta_{12}\approx 0$ in principal channel, and for: frequency tuning of radiations in excited channel more than zero ($\eta_{23}>0$).

The solution for Eq. 4 has a physical meaning, when the frequency tuning η_{12}^{0} in principal channel agrees with the following inequalities:

$$\frac{-\delta_{12}B_{21}}{B_{12}+B_{21}} < \eta_{12}^0 < \frac{-\delta_{12}B_{21}}{B_{12}(1+B_{23}/B_{32})+B_{21}} \qquad (6)$$

for minimum and

$$\frac{-\delta_{12}B_{21}}{B_{12}+B_{21}} > \eta_{12}^0 > \frac{-\delta_{12}B_{21}}{B_{12}(1+B_{23}/B_{32})+B_{21}} \qquad (7)$$

for maximum.

At the same time the solution for Eq. 5 is realized for positive frequency tuning in excited channel ($\eta_{23}^0 > 0$). The saturation intensity in principl channel I_{12}^{sat}, as a function of tuning frequency η_{12}, has three extremums (two minimums and one maximum). These extremums in general deprend on intensity in excited channel I_{23}. For small intensity in excited channel ($I_{23} \approx 0$, two-level model), these extremums are realized at frequency tuning η_{12}^0 -0.2 (minimum), η_{12}^0 -1.4 (minimum) and η_{12}^0 -0.8 (maximum), which agrees with Eq. 4 for two-level model $\eta_{12}^0 = -\delta_{12}(B_{21}/vp_{21})/J$ and with Fig. 1c curves 1, 5, Fig. 2a and b.

Figure 2 represents the frequency tuning of radiations, for which are realized the extremum of saturation intensity, in principal channel η_{12}^0 (a), (b) and in excited channel η_{23}^0 (c) as function of radiation intensity in excited channel I_{23}. Figure 2 illustrates the dependence of frequency tuning η_{12}^0 on radiation intensity in excited channel I_{23} for: (a) maximum in frequency diapason $-1.15 < \eta_{12}^0 < -0.8$ and local minimum in diapason $-1.40 < \eta_{12}^0 < -1.15$; (b) optimum (minimum) in frequency diapason $-0.2 < \eta_{12}^0 < 0$. Figure 2b shows that, the increasing of radiation intensity in excited channel I_{23} gives a little anti-Stokes shift of optimum tuning frequency η_{12}^0 . The same anti-Stokes shift happened for local minimum ($-1.40 < \eta_{12}^0 < -1.15$), but a Stokes shift happened for maximum with increasing of radiation intensity I_{23}.

Figure 2a shows that, for some frequency tuning η_{23} in excited channel curves 2, 3, 4, the local minimum and the maximum move closer one to other at frequency tuning $\eta_{12}^0 = -1.15$ (dashed line), with increasing of radiation intensity in excited channel I_{23}. In this case, both of the maximum and the local minimum are disappeared (Fig. 1c curves 2, 3, 4 and Fig. 2a curves 2, 3, 4 for big radiation intensity in excited channel I_{23}). The upper part of Fig. 2a, above dashed line, corresponds to maximum and the lower part, under dashed line, corresponds to a local minimum. In addition the dependence of optimum values of frequency tuning η_{12}^0 of radiations in principal channel on intensity I_{23} is illustrated in Fig. 2b. The optimum values of frequency tuning η_{23}^0 of radiations in excited channel, for which are realized the optimum values of saturation intensity in principal channel I_{12}^{sat} , are illustrated in Fig. 2c as a function of radiation intensity I_{23}. Figure 2c shows the monotonic dependence of frequency tuning η_{23}^0 on the radiation intensity I_{23}, where the anti-Stokes shift happens for optimum with increasing of radiation intensity in excited channel.

CONCLUSIONS

For double frequencies excitation of nonlinear medium, the saturation intensity in principal channel I_{12}^{sat} is decreasing with increasing of radiation intensity in excited channel I_{23}. The optimization of saturation intensity in principal channel I_{12}^{sat} is realized for enough big intensity of radiations in excited channel ($I_{23} > vp_{32}/B_{32}$). And the frequency of radiations in excited channel must be tuned with a little anti-Stokes shift from the centre of absorption band, not more than halfwidth of absorption band. The frequency tuning of radiations in the principl channel η_{12} must be tuned with a little Stokes shift from the centre of absorption band, not more than (1/5) of halfwidth of absorption band.

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