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Saturation Processes in Principal Channel of Dye Solutions with Coincident Absorption and Emission Bands-Part II

[1]Jihad S.M. Addasi, [1]Mohanad Jadan and [2]Saleh A. Abushendi
[1]Department of Applied Physics,
[2]Department of Mathematics and Computer Science,
Faculty of Science, Tafila Technical University, Jordan

Abstract: A phase response of dye solution and saturation processes can be taken into consideration to describe a nonlinear medium. Nonlinear medium can be modeled by a typical three-level configuration (S_0-S_1-S_2), for which the transition of molecules in principal channel (S_0-S_1) are occurred by the 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 form their transitions in excited channel (S_1-S_2). The phase response of dye solution in principal channel (S_0-S_1) reaches saturation at intensity I_{12}^{sat}. At this point, the saturation intensity decreases with increasing radiation intensity in the 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 into the centre of principal absorption band. In addition, the saturation intensity I_{12}^{sat} has its optimum value when the radiations in excited channel have enough high intensity I_{23} and a frequency tuning into the centre of absorption excited band.

Key words: Saturation, principal channel, coincident bands, 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 (Popov *et al.*, 2000; Poliakov *et al.*, 1998; Bolotskikh *et al.*, 1986; Shalaev, 2002). 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 (Agishev *et al.*, 1998; Pashinin *et al.*, 1997; Popov and Shalaev, 1980). The lifetimes of vibration energy levels of dye solutions are significantly lower than the lifetimes of electronic energy levels (Tichonov and Shpak, 1979). In this case, the electronic states can be taken as homogeneously broadened levels, which gives ability to use two and three-level models with averaged Einstein coefficients for many nonlinear medium (Agishev *et al.*, 1998). Two-level model can be used to study nonlinear processes involved by radiations of intensity I_{12} at frequency ω_0, but in this model its difficult to control these nonlinear processes, (Fig. 1a). Three-level model for nonlinear media gives ability to control the nonlinear properties, which occurred by independent light beams (optical pumping) acting in principal or excited channel (Agishev *et al.*, 1998; Pashinin *et al.*, 1997; Rubanov *et al.*, 2000). 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 the second channel (Rubanov *et al.*, 2000) (Fig. 1b). The refractive index, extinction coefficient, absorption and emission of nonlinear media depend on the intensity and frequency of light fields acting in each of principal and excited channels.

The aim of this theoretical study is to find an optimal conditions of saturation processes in principal channel for nonlinear media with coincident absorption and emission bands.

The balance equations under a double frequencies excitation of dye solution modeled by three-level configuration can be written as follows:

$$\begin{aligned} N_1 B_{12}(\omega_0) I_{12} &= N_2 (B_{21}(\omega_0) I_{12} + v p_{21}); \\ N_2 B_{23}(\omega) I_{23} &= N_3 (B_{32}(\omega) I_{23} + v p_{32}); \\ N &= N_1 + N_2 + N_3 \end{aligned} \qquad (1)$$

where, N_i is the population of i is energy level, N is the number of molecules in the unit volume of nonlinear

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Corresponding Author: Jihad S.M. Addasi, Department of Applied Physics, Faculty of Science, Tafila Technical University, Jordan

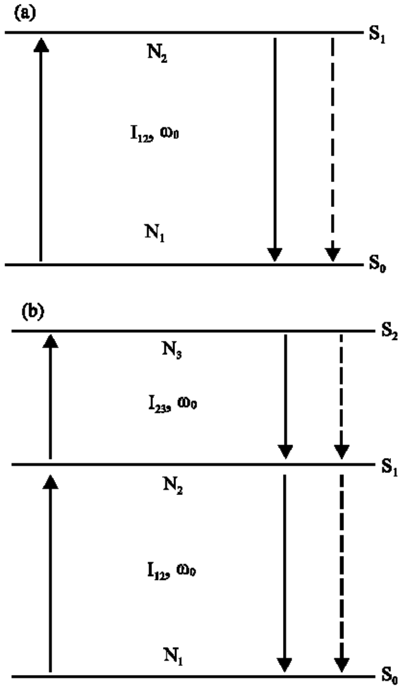

Fig. 1: Diagram of (a) two-level and (b) three-level models. The solid lines denote the radiation-induced transitions of molecules and the dashed lines denote spontaneous and radiationless transitions. Where, S_0 is the ground state; S_1 (S_2) are the first (second) excited states; N_i is the population of i- energy level; I_{12}, ω_0 (I_{23}, ω) are the intensity, frequency of radiations in principal (excited) channel

medium, 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} (ω_0), B_{21} (ω_0) are determined at the frequency of radiations ω_0 in principal (S_0-S_1) channel. At the same time B_{23} (ω), B_{32} (ω) are determined at frequency of radiations ω in excited channel. The refractive index determined by balance equations can be used to study many nonlinear processes such as bleaching processes.

THEORY

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

$$\chi_{12}(\omega_0) = \frac{c}{2v}K_{12}(\omega_0) = \frac{c\hbar\omega_0}{2v}(N_1B_{12}(\omega_0) - N_2B_{21}(\omega_0)) \quad (2)$$

where, K_{12} (ω_0) is the absorption coefficient in principal channel.

From Eq. 1 and 2 the extinction coefficient χ_{12} (ω_0) will be:

$$\chi_{12}(\omega_0) = \chi_0(1 + aI_{23})/K \quad (3)$$

where:

$K = 1 + JI_{12} + aI_{23} + bI_{12}I_{23}$;
$a = B_{32}/vp_{32}$;
$J = (B_{12}+B_{21})/vp_{21}$;
$b = B_{12}B_{23} + aJ$;
$\chi_0 = N\hbar cB_{12}(\omega_0)/2v$;

χ_0 (ω_0) is the linear extinction coefficient. The extinction coefficient, Eq. 3, has a monotonic proportionality with intensity of radiations in each channel (I_{12}, I_{23}). It has maximum value ($\chi_{12} = \chi_0$) at intensities $I_{12} = I_{23} = 0$. The extinction coefficient has the half of its maximum value at saturation intensity in the principal channel with value:

$$I_{12}^{sat} = \frac{1 + aI_{23}}{J + bI_{23}} \quad (4)$$

From Eq. 4 the saturation intensity (I_{12}^{sat}) has a monotonic dependence on radiation intensity in excited channel (I_{23}). To study the dependence of saturation intensity I_{12}^{sat} on frequency tuning of radiations in both principal and excited channels, some parameters of nonlinear medium must be determined. Moreover, this dependence has an optimum values as a function of frequency tuning.

RESULTS AND DISCUSSION

Taking into consideration a nonlinear medium with a Gaussian form of coincident mirror-symmetric absorption and emission bands ($\omega_{ij} = \omega_{ji} \Rightarrow \delta_{ij} = (\omega_{ij} - \omega_{ji})/\Delta_{ij} = 0$, where, Δ_{ij}; ω_{ij} are the profile halfwidth and the centre of i-j channel. 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}. Eq. 4 and Fig. 2a and b show the monotonic dependence of saturation intensity I_{12}^{sat} on radiation intensity in the excited channel I_{23} for different frequency tuning of radiations in principal channel η_{12} (Fig. 2a) and in excited channel η_{23} (Fig. 2b). From Eq. 4 the saturation intensity (I_{12}^{sat}) in principal channel, for this form of absorption and emission bands, has optimum values (minimum) at radiations frequency tuned into the centre of principal absorption band $\eta_{12}^0 = 0$ (Fig. 2c). At the same time, the optimum of I_{12}^{sat} is occurred at radiations frequency in excited channel tuned into the centre of excited absorption band ($\eta_{23}^0 = 0$) (Fig. 2d).

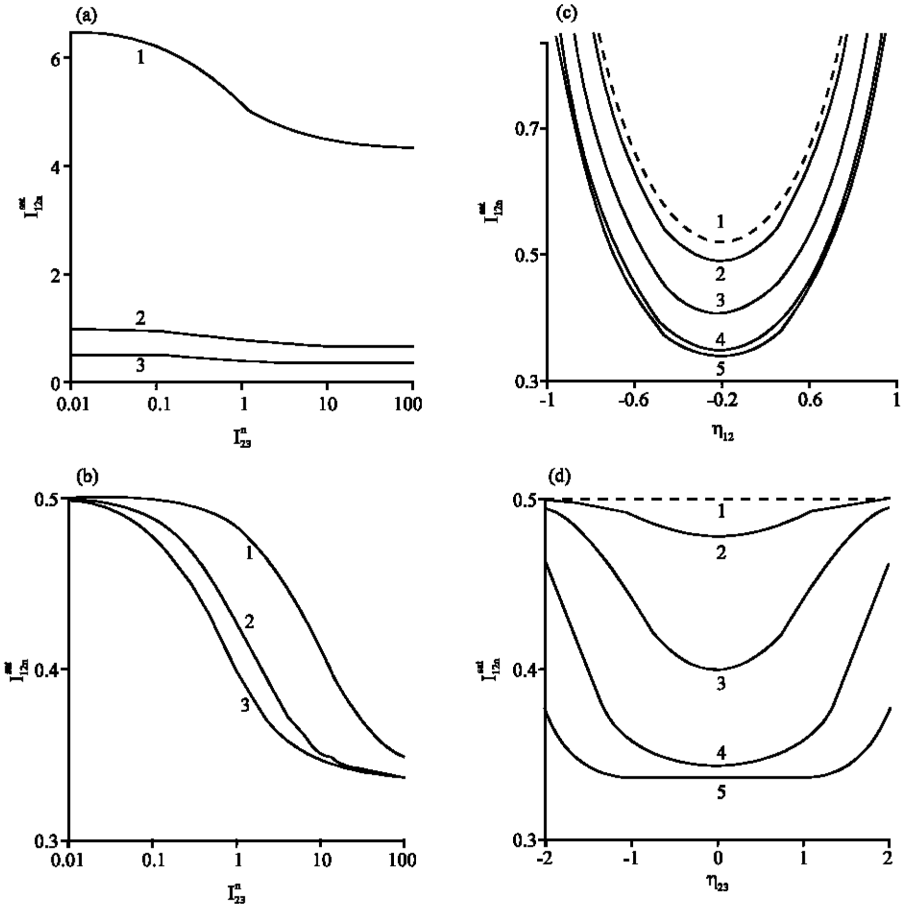

Fig. 2: Dependence of saturation intensity I_{12n}^{sat} on: radiation intensity I_{23}^{n} (a) (b) and on frequency tuning of radiations in principal η_{12} (c) and in excited η_{23} (d) channels. Curves: 1, 2 and 3 at: (a) η_{12}: ±1.6, ±0.8 and 0; (b) η_{23}: ±1.6, ±0.8 and 0. Curves: 1, 2, 3, 4 and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

All Eq. 1-4 will be true for two-level model, when there are no radiations in excited channel ($I_{23} = 0 \Rightarrow N_3 = 0$). In this case, let us rewrite Eq. 4 for two-level model:

$$I_{12}^{sat} = (1/J) \qquad (4^*)$$

The saturation intensity I_{12}^{sat} for two-level model equation (4*), has its optimum values at radiations frequency tuned into the centre of absorption band ($\eta_{12}^{0} = 0$) for nonlinear medium with coincident absorption and emission bands, (Fig. 2c) curve 1. In Fig. 2 the following statements are considered: the maximum values of Einstein coefficients are the same for all bands. The radiation intensities are normalized to the value vp_{21}/B_{12}^{max} and vp_{32}/B_{32}^{max} in the principal and in the excited channels, respectively.

The saturation in principal channel is achieved with small enough intensity when an effective excitation of molecules occurs in excited channel. Radiations in excited channel must have a high enough intensity and the frequency tuned into the centre of excited absorption band (I_{23}^{n}>10;$\eta_{23} \approx$ 0).

Equation (4*), for two-level model, is shown (Fig. 2c and d) by dashed line (curve 1), where the optimum value of saturation intensity (I_{12n}^{sat}=0.5) is achieved at frequency tuning η_{12}^{0}. Figure 2c and d show the saturation intensity I_{12}^{sat} in principal channel for the three-level model (curves 2, 3, 4 and 5) which are always lower than that of two-level model. Figure 2 shows that a little frequency shift of radiations from centre of principal absorption band ($\eta_{12} \neq 0$) increases the saturation intensity (I_{12}^{sat}) significantly, but the frequency shift in excited channel (η_{23}) makes a little change in saturation intensity of radiations in principal channel I_{12}^{sat}.

CONCLUSION

For medium with coincident mirror-symmetric absorption and emission bands the saturation intensity in the principal channel has optimum values (minimum) at radiations frequency tuned into the centre of principal absorption band ($-0.2<\eta_{12}<0.2$). The optimum values (minimum) of saturation intensity are occurred at radiations with high intensity ($I_{23}^{n}>10$), tuned into the centre of excited absorption band ($-0.8<\eta_{23}<0.8$).

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