ANALYSIS OF NONSTATIONARY SOLUTIONS OF RATE EQUATIONS OF DYE LASER. I*,**

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(*Received April 24, 1978*)

Numerical solutions of rate equations of a six-level model of dye laser pumped with a linearly increasing excitation pulse were obtained. It was found that photon number in the cavity and populations of molecular energy levels under high excitations conditions tend towards the appropriate values characteristic for stationary case. Behaviour of dye laser predicted on the basis of previously used rate equations and on the basis of a six-level model are compared.

1. *Introduction*

It has been shown [1–3] that for the description of a dye laser seven differential equations have to be used. The whole set of rate equations of a dye laser with active medium represented by the energy level diagram presented in Fig. 1 is of the form

$$\begin{aligned}
\dot{n} &= -2\kappa n + b_1(p_3-p_2)n - b_2(p_5-p_6)n + b_1' p_3,\\
\dot{p}_1 &= a_{12}p_2 + a_{15}p_5 + a_{16}p_6 - Wp_1,\\
\dot{p}_2 &= a_{23}p_3 - a_{12}p_2 + b_1(p_3-p_2)n,\\
\dot{p}_3 &= a_{34}p_4 - (a_{53}+a_{23})p_3 - b_1(p_3-p_2)n,\\
\dot{p}_4 &= Wp_1 - a_{34}p_4,\\
\dot{p}_5 &= a_{53}p_3 + a_{56}p_6 - a_{15}p_5 - b_2(p_5-p_6)n,\\
\dot{p}_6 &= -(a_{56}+a_{16})p_6 + b_2(p_5-p_6)n,\\
& p_1+p_2+p_3+p_4+p_5+p_6 = 1,
\end{aligned} \tag{1}$$

where $p_1, p_2, p_3, p_4, p_5, p_6$ are the probabilities of populations of respective energy levels of dye molecule, n — photon number per one molecule, 2κ — cavity losses, b_1 and b_2 — Einstein coefficients of $2 \leftrightarrow 3$ and $5 \leftrightarrow 6$ transitions respectively, a_{ij} — spontaneous

* Dedicated to Professor Aleksander Jabłoński on the occasion of his 80th birthday.

** This work was supported within the project MR. I. 5.

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radiation and radiationless transition rates, and W is the pumping parameter. The term $b'_1 p_3$ in the first equation is added to account for the spontaneous emission in the mode considered.

The stationary solutions of Eqs (1) were discussed in [1–3]. The aim of this paper is to investigate the nonstationary solutions of these equations and in particular to examine

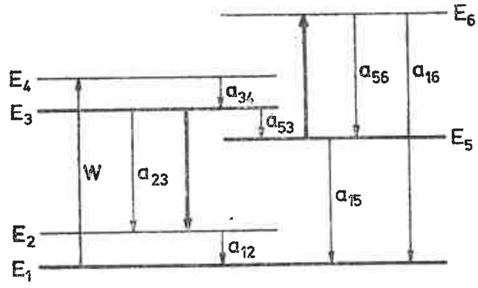

Fig. 1. Energy level diagram of dye molecule

the influence of rise-time of pumping pulse on the time evolution of $n(t)$ and $p_i(t)$. The performed analysis is based on numerical solutions with typical for dye laser molecular and cavity parameters. These calculations were performed assuming that the pumping function $W(t)$ is a linear function of time. Such pumping function is the simplest one next to the stationary case. In practice pumping pulses are much more complicated and for such pulses the analysis of the influence of the rise-time on laser action would be difficult. It is obvious that the performed analysis for linear pumping pulses illustrates well the development of laser action and can be easily adopted for pumping sources used in practice.

2. *Results of numerical calculations*

In the numerical calculations the following molecular and cavity parameters were used:

$$a_{12} = a_{34} = a_{56} = 10^{11}\,\mathrm{s}^{-1}, \qquad a_{23} = 2\times 10^{8}\,\mathrm{s}^{-1},$$
$$a_{53} = 0.82\times 10^{7}\,\mathrm{s}^{-1}, \qquad a_{16} = 10^{8}\,\mathrm{s}^{-1},$$
$$b_1 = 1.56\times 10^{10}\,\mathrm{s}^{-1}, \qquad b_2 = 8.59\times 10^{9}\,\mathrm{s}^{-1},$$
$$2\kappa = 10^{8}\,\mathrm{s}^{-1}. \tag{2}$$

These values are typical for flash-pumped dye lasers. The parameter a_{16}, which accounts for desactivation rate of the higher triplet state in the path other than $6 \to 5 \to 1$, is not known yet, but the chosen value seems to be reasonable.

The pumping function is chosen in the form

$$W(t) = Ft, \tag{3}$$

where the parameter F is the measure of the rise-time of the pumping pulse. Its values are taken in the range of $6\times 10^{13}\,\mathrm{s}^{-2} - 8\times 10^{15}\,\mathrm{s}^{-2}$. Some of the results of numerical calculations are illustrated in Fig. 2.

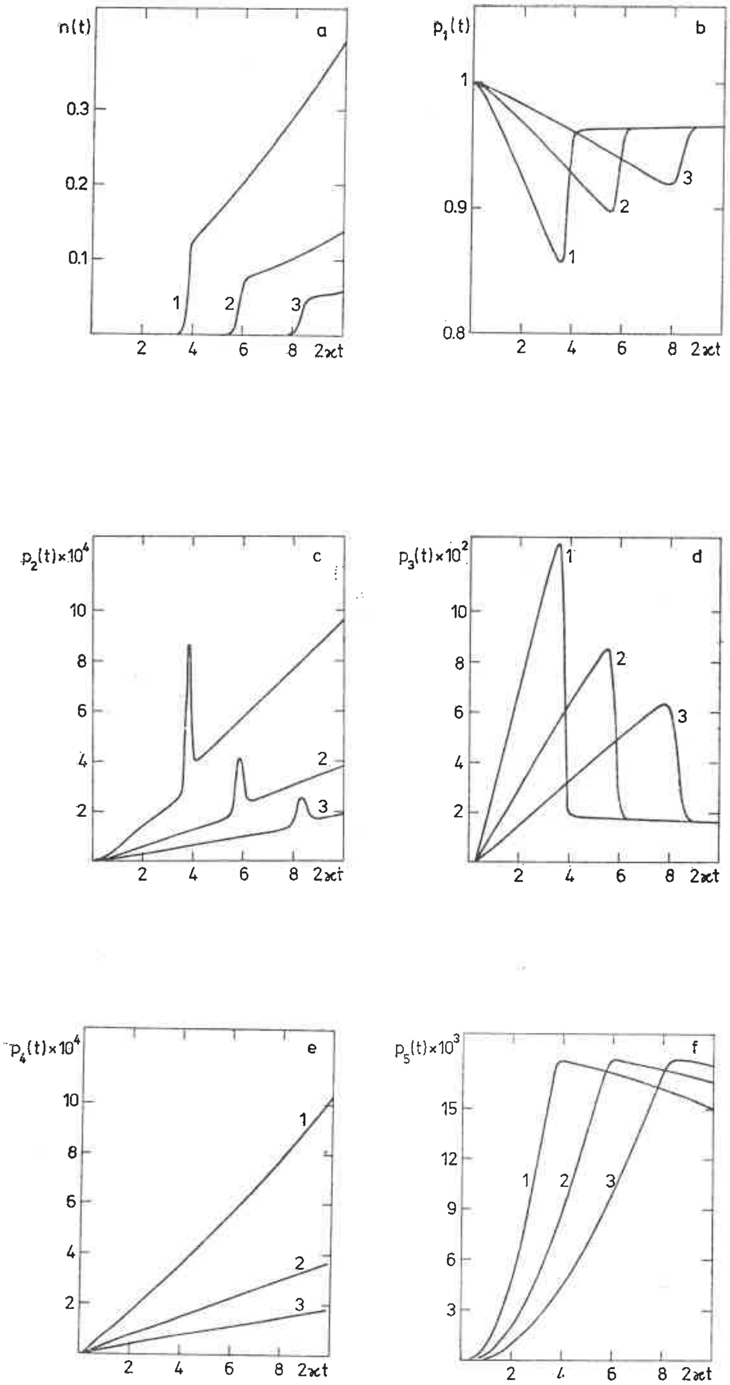

Fig. 2. Time dependences of photon number and respective population probabilities. *1* —$F = 10^{15}s^{-2}$, *2* — $F = 4\times10^{14}\,s^{-2}$, *3* — $F = 2\times10^{14}\,s^{-2}$

3. *Discussion*

In general the time dependences of photon number and the population of the 1-st excited singlet state of a dye laser thought as a six-level system are similar to those for other type of lasers. The only difference between dye lasers and other types of lasers is due to the existence of dynamical triplet losses in the first one. According to the definition given in [2] these losses are given by the formula:

$$T(t) = b_2(p_5(t)-p_6(t)). \tag{4}$$

In a similar way the gain coefficient can be defined:

$$I(t) = b_1(p_3(t)-p_2(t)). \tag{5}$$

The first equation of the set [1] can be written in the form:

$$\dot{n}(t) = G(t)n+b_1'p_3(t), \tag{6}$$

where

$$G(t) = I(t)-T(t)-2\kappa \tag{7}$$

is a net gain. The net gain $G(t)$ calculated with parameters (2) is presented in Fig. 3. The time evolution of laser generation may be divided into 4 intervals. The first two intervals are separated in the point $G(t) = 0$, which is the threshold condition. In the 1-st interval the net gain is negative and photons are produced mainly by the spontaneous emission.

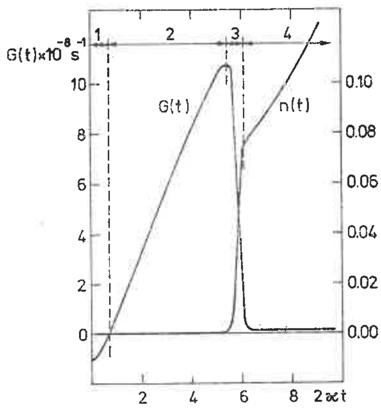

Fig. 3. Net gain (as specified by Eq. (7)) and photon number in the cavity, versus time for pumping function $W(t)$ with slope $F = 4\times10^{14}\ s^{-2}$

Crossing the threshold the number of photons increases not only in a spontaneous emission process but also by induced emission. The term $G(t)n(t)$ in a very close vicinity of the threshold becomes much larger than the term $b_1'p_3(t)$, what means that in a very good approximation the last term in equation (6) can be emitted in intervals 2, 3 and 4. It does not mean however that in the interval 2 the spontaneous desactivation process of E_3 level can be neglected. As a matter of fact because of the large number of modes of radiative spontaneous transitions and because of radiationless transitions, spontaneous desactivation of the first excited singlet state is dominating and only at the end of this interval the induced emission starts to compete with spontaneous process.

Neglecting the term $b_1' p_3$ in equation (6) the photon number may be given by the formula:

$$n(t) = n_0 \exp \left[\int_{t_0}^{t} G(t')dt' \right], \tag{8}$$

where n_0 is the photon number in the time point t_0 after which the condition $G(t)n(t) \gg b_1' p_3(t)$ is well fulfilled. For different slopes of $W(t)$, the values of t_0 are very close to the threshold point of t, and can be so chosen that original values n_0 are equal to each other. In these conditions the number of photons in every time point $t > t_0$ depends only on the integral $\int_{t_0}^{t} G(t')dt'$. This explains the regularity that the delay time for the development of laser generation is shorter for larger F values (see Fig. 2). It has to be noted that the net gain $G(t)$ is larger in interval 2 than its value in interval 4 where well

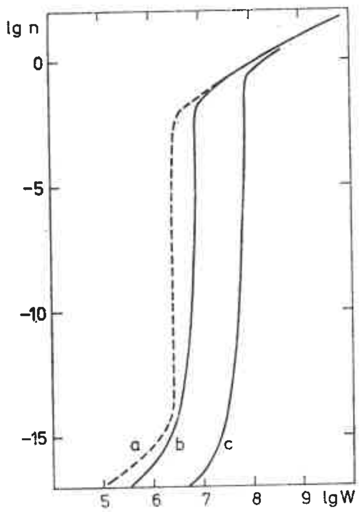

Fig. 4. Photon number n versus pumping parameter W for different slopes F of pumping pulse. a — stationary solution, b — $F = 6 \times 10^{13}\ s^{-2}$, c — $F = 4 \times 10^{15}\ s^{-2}$

developed laser action takes place. All the excess inversion produced in interval 2 is rapidly used in photon production process in interval 3. This excess inversion is larger when the slope of the pump becomes larger. In interval 3 desactivation of the 1-st excited singlet state by the induced emission process starts to be more important than spontaneous desactivation processes. In the last interval the induced emission processes dominate.

In the last stage of laser generation (interval 4) the photon number starts to increase with the increase of the pump like in the stationary behaviour. This is illustrated in Fig. 4 where the dependence of photon number versus intensity of pumping for two different slopes of pump pulses is compared with the stationary solution. As is to be expected the stationary solution will be reached faster for slower pumps. It is worth while to note that for used parameters in the case of nonstationary laser action always less photons are produced than in the stationary case for the same intensity of the pump.

Besides the above characteristic feature of nonstationary solutions of Eqs. (1) some other, typical for dye laser, can be listed:

1. Populations p_2, p_4 and p_6 are much smaller than the remaining ones. This is due to the fact that the desactivation probabilities of respective energy levels are much larger than the desactivation probabilities of remaining energy levels. The population p_4 is determined by the transition probability a_{34} and intensity of the pump. The population p_6 depends on the desactivation rates $a_{56}+a_{16}$ and on the number of photons in the cavity as well and its time behaviour is similar to that presented in Fig. 2a. The time dependence of p_2 is more complicated. This population depends not only on the depopulation rate of E_2 level but also on the rate of its population in two different processes, namely on the spontaneous radiative and radiationless transitions $3 \rightarrow 2$ and on the induced emission proces $3 \rightarrow 2$ which is proportional to the number of photons. Rapid increase of photon number causes that p_2 has a characteristic maximum (see Fig. 2c).

2. Time variation of p_5 seems to be very interesting. For molecular parameters used p_5 decreases with the increase of $W(t)$ and $n(t)$, when laser generation takes place. It means that in this case triplet losses due to triplet-triplet absorption are decreasing. This is a specific property of dye lasers. Such a property cannot be described by simplified rate equations in which a dye laser is thought as a 4-level system with triplet-triplet absorption losses added [5, 6].

3. The population p_3 in the generation interval (interval 4 in Fig. 3) changes slowly tending to the asymptotic solution which is equal to the stationary one.

We are greatful to Professor L. Wolniewicz for advice in numerical problems.

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