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INFLUENCE OF TRIPLET LOSSES ON THE RELAXATION OSCILLATIONS IN DYE LASERS

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A qualitative discussion of the influence of triplet parameters on spiking generation in dye lasers is presented. The investigations are based on numerical solutions of the rate equations of a dye laser.

1. Introduction

Pulse excited lasers often reveal relaxation oscillations (spiking). The phenomenon is due to the interaction between the excess population inversion in the active medium and the energy of the electromagnetic field in the cavity [1–3]. In contrast to solid state lasers where the spiking phenomenon is very common, in dye lasers the relaxation oscillations are seldom observed and can be obtained only under special experimental conditions. It follows from previous investigations on spiking phenomenon (theoretical as well as experimental) [5–7] that pulses much shorter than the exciting pulse can be obtained from the dye laser by a specific choice of laser parameters such as photon cavity decay time, pumping level and others. It was also pointed out [4–7] that controlled relaxation oscillations offer a convenient technique for the generation of subnanosecond, high repetition-rate, and tunable, pulses from a N_2-laser pumped dye laser. The theoretical analyses given in [4–7] were based on the assumption that the active medium of the dye laser can be represented by a four-level system, which means that the triplet states populations are neglected. This assumption is valid in the case of dye solutions for which the intersystem crossing rate is sufficiently small. The aim of this paper is to show how the dynamical triplet losses influence the relaxation oscillations in dye lasers. In particular, it is shown that some of the molecular parameters describing triplet losses have a considerable effect on the spiking generation in dye lasers.

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2. Numerical results

Our investigations are based on the numerical solutions of the following rate equations [8]:

$$\dot{n} = b_1 p_S n - (t_c^{-1} + b_2 p_T)n + F p_S,$$

$$\dot{p}_S = P(t) - (a_{SS_0} + a_{ST})p_S - b_1 p_S n,$$

$$\dot{p}_T = a_{ST} p_S - a_{TS_0} p_T - \frac{b_2 p_T n a_{T'S_0}}{a_{T'S_0} + a_{T'T}}. \quad (1)$$

Here n is the photon number in the cavity per one molecule, p_S, p_T — normalized populations of excited singlet and triplet states, b_1, b_2 — Einstein coefficients for induced transitions between singlets and triplets respectively, t_c — photon cavity decay time, $P(t)$ — pum-

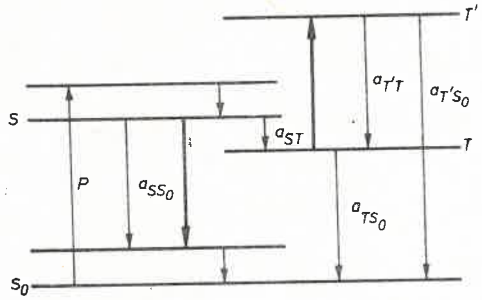

Fig. 1. Energy level diagram of a dye molecule. Heavy arrows indicate induced transitions

ping function, a_{SS_0}, a_{ST}, a_{TS_0}, $a_{T'T}$ and $a_{T'S_0}$ — appropriate spontaneous transitions rates (see Fig. 1). The term Fp_S in the first equation is added to account for the spontaneous emission in the mode considered.

The time dependent term $b_2 p_T$ is due to triplet-triplet absorption and is called triplet losses. The total losses of the cavity are:

$$k = t_c^{-1} + b_2 p_T. \quad (2)$$

Equations (1) were solved numerically with the aid of the Runge–Kutta method. The calculations were performed for fixed values of parameters b_1, a_{SS_0}, (namely, $b_1 = 8.79 \times 10^{11}\ s^{-1}$, $a_{SS_0} = 1.6 \times 10^8\ s^{-1}$), and for different values of b_2, a_{ST}, a_{TS_0}, $a_{T'S_0}$ and $a_{T'T}$. Besides, two pumping functions are considered:

1) $$P(t) = \begin{cases} 0, & t \leqslant 0 \\ P = \text{const}, & t > 0 \end{cases}$$

2) $$P(t) = P_{max} \times \begin{cases} \exp\left[-\left(\frac{t}{T_1}\right)^2 \ln 2\right], & t \leqslant 0 \\ \exp\left[-\left(\frac{t}{T_2}\right)^2 \ln 2\right], & t > 0. \end{cases}$$

The second form of the pumping function is adapted to the N_2-laser pulse. All the laser and molecular parameters used in our calculations are accessible experimentally.

As was recently shown [9], the set (1) of rate equations is valid as long as the increase of the photon number per round-trip of the cavity is small i.e. when the approximation

$$\frac{n(t+T)}{n(t)} = 1 + \frac{T}{n(t)} \frac{dn(t)}{dt} \tag{3}$$

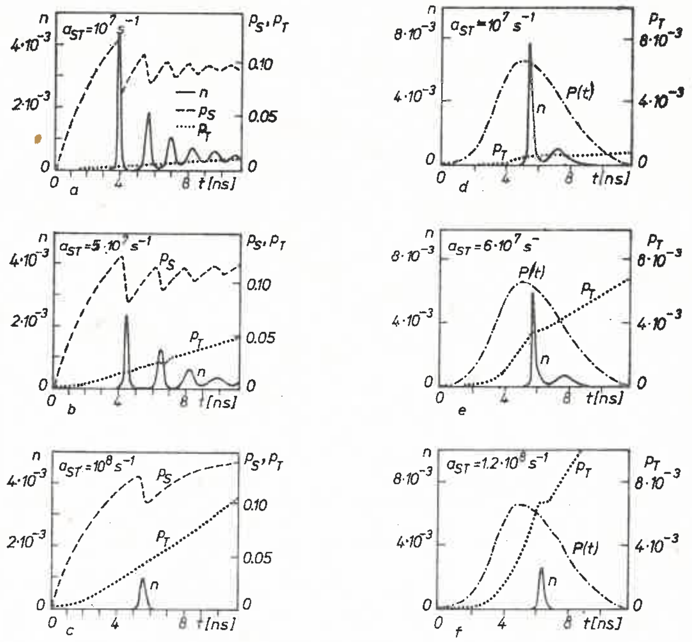

Fig. 2. Influence of the intersystem crossing rate on spiking generation in dye lasers. (a, b, c) — time dependences of n, p_S and p_T; photon cavity decay time $t_c = 10^{-11}$s; the pumping function $P(t)$ is assumed to be 0 for $t \leqslant 0$ and 5×10^7 s^{-1} for $t > 0$; $P/P_{th} = 3$ (threshold pump P_{th} is calculated for $p_T = 0$). (d, e f) — time dependences of n and p_T with $t_c = 1.7\times10^{-10}$ s and pumping function as given in the figures ($P_{max} = 8 \times 10^6$ s^{-1}, $P_{max}/P_{th} = 4.5$)

is valid. Here $T = 2L/c$ is the cavity round-trip time. In particular, the above condition is fulfilled in systems with small cavity length L. In our calculations (3) is valid when $L \leqslant 1$ mm. The results of the calculations are given in Figs 2a–f, where the time evolutions of $n(t)$, $p_S(t)$ and $p_T(t)$ are plotted for different values of the intersystem crossing rate a_{ST}.

3. *Discussion*

A general conclusion that may be deduced from the numerical results is that the stronger the effects due to the triplet parameters are the larger is the ratio of the triplet losses to the total losses in the laser system. In particular, it can be noticed that when a_{ST}

increases the intensity of spikes becomes smaller and they are more distant one from another. Besides, the increase of the intersystem crossing rate a_{ST} causes the retardation of the initial spike with respect to the pumping pulse. From Figs 2c, f it is seen that for some values of a_{ST} the laser generates a single spike whose time duration is much shorter than that of the exciting pulse. A simple explanation of the appearance of a single spike is as follows: for suffciently large a_{ST} the triplet losses rise so quickly that, after the first spike has been generated, the threshold condition can no more be met.

The above results are ones for varying values of a_{ST}, all the other parameters being fixed. Analogical calculations were performed for the parameter b_2 increasing and, as was expected, the effects were very similiar. The remaining triplet parameters i.e. a_{TS_0}, $a_{T'S_0}$ and $a_{T'T}$ do not influence the spiking phenomenon vitally for typical dye laser solutions.

While the above considerations are based on the numerical solutions of the rate equations (1), it is interesting to notice that similar conclusions may be deduced from the small signal analysis of these equations. In case the triplet losses are negligible and the pumping is constant in time, the small signal solutions of (1) are [4, 6]:

$$n = n_0 + A \exp\left[-\sigma t\right] \cdot \cos(\omega t + \varphi_1),$$

$$p_S = p_{S_0} + B \exp\left[-\sigma t\right] \cos(\omega t + \varphi_2). \tag{4}$$

Here $\sigma = \frac{1}{2} P b_1 t_c$ is the damping constant, $\omega = [P b_1 - (a_{SS_0} + a_{ST}) t_c^{-1} - (\frac{1}{2} P b_1 t_c)^2]^{1/2}$ the oscillation frequency. As was shown [4, 6], the relaxation oscillations occur if the necessary condition $t_c^{-1} > a_{SS_0} + a_{ST}$ is fulfilled.

The effect of triplet losses onto the above solutions may be accounted, roughly, by replacing, in (4), t_c^{-1} by k. Since for the numerical values of parameters used in the above calculations $\frac{d\omega^2}{dk} = -(a_{SS_0} + a_{ST}) + \frac{P b_1}{2k^3} < 0$, ω decreases with k, i.e. the spikes become more distant one from another. On the other hand, it is clear from (4) that the damping constant σ decreases when k increases. Thus, although the small signal analysis does not offer rigorous solutions of the rate equations (1), qualitative conclusions obtained from it are correct and valuable.

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REFERENCES

[1] G. Birnbaum, *Optical Masers*, New York, Academic Press 1964, pp. 51 and 191.
[2] D. Ross, *Laser Lichtverstärker und Oszillatoren*, Frankfurt am Main 1966, p. 291.
[3] R. Stemme, G. Herziger, W. Weber, *Opt. Commun.* **10**, 221 (1971).
[4] Y. Miyazoe, M. Maeda, *IEEE J. Quantum Electron.* QE-7, 36 (1971).
[5] C. Lin, C. V. Shank, *Appl. Phys. Lett.* **26**, 389 (1975).
[6] C. Lin, *IEEE J. Quantum Electron.* **QE-11**, 602 (1975).
[7] S. Mory, R. König, *Exp. Tech. Phys.* **25**, 581 (1977).
[8] A. Bączyński, T. Marszałek, *Bull. Acad. Pol. Sci. Ser. Sci. Math. Phys. Astron.* **26**, 91 (1978).
[9] P. Flamant, *Opt. Commun.* **25**, 247 (1977).