SUBNANOSECOND RELAXATION OSCILLATIONS IN NITROGEN LASER PUMPED DYE-LASERS

By

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A four-level kinetic dye-laser model was investigated using N_2 laser pumping. The damped oscillation of dye-lasers and the generation of subnanosecond pulses are investigated theoretically and experimentally. The numerical solutions of coupled rate equations are in good qualitative agreement with experimental data.

An ever increasing number of laser applications demand light sources emitting subnanosecond pulses. The best known method to achieve subnanosecond pulses is mode-locking. In this paper a theoretical calculation is presented for studying the kinetic properties of nitrogen laser-pumped dye-lasers and conditions of damped subnanosecond oscillation are given. The rate equations of organic dye-lasers and the solutions for different cases are well known [1—3]. We are applying the equations given by ATKINSON and PACE [3]. The energy levels (involved in lasing) of an organic dye are schematically depicted in Fig. 1. (1 is the singlet ground, 2 is the first excited singlet, T and 3 are triplet states). Each electronic state consists of a set of vibrational (heavy lines) and rotational (light lines) sublevels. The radiative transitions are denoted by solid, the nonradiative transitions by wavy, and the forbidden transitions by dashed lines. The rate equations can be written as follows,

$$\frac{dn_m}{dt} = n_m \left[\left(n_2 \sigma(\lambda) - n_T \sigma_{TT}(\lambda) - n_1 \sigma_{ss}(\lambda) \right) \frac{Fc}{\eta} - \frac{1}{\tau_m} \right] + \frac{Fcn_2 \sigma(\lambda)}{\eta V}, \tag{1}$$

$$\frac{dn_2}{dt} = W_2 n_1 - \frac{n_2}{\tau} - \sum_m n_2 n_m \sigma(\lambda) \frac{c}{\eta} + \sum_m n_1 n_m \sigma_{ss}(\lambda) \frac{c}{\eta}, \tag{2}$$

$$\frac{dn_T}{dt} = kn_2 - \frac{n_T}{\tau_T} - \sum_m n_T n_m \sigma_{TT}(\lambda) \frac{c}{\eta} + \frac{n_3}{\tau_3}, \tag{3}$$

$$\frac{dn_3}{dt} = \sum_m n_T n_m \sigma(\lambda) \frac{c}{\eta} - \frac{n_3}{\tau_3}, \tag{4}$$

$$n = n_1 + n_2 + n_T + n_3. \tag{5}$$

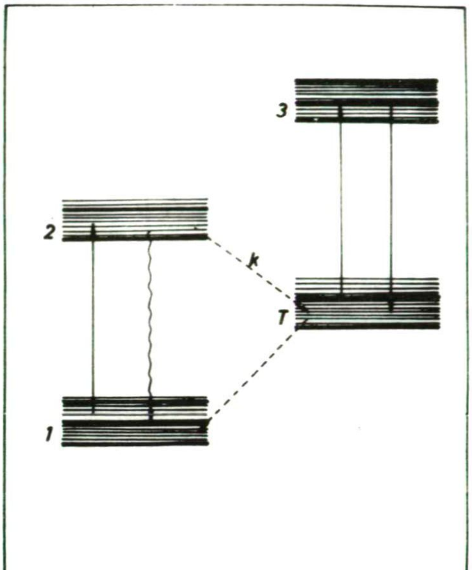


Fig. 1. Energy levels of a typical dye

Here we used the following notation: n_m is the photon number in mode m, n_1, n_2, n_T, and n_3 are the population densities of the 1, 2, T, and 3 levels, V is the volume of cavity, c is the velocity of light, η is the refractive index of the solution, $\sigma(\lambda), \sigma_{ss}(\lambda)$, $\sigma_{TT}(\lambda)$ are the cross sections of stimulated emission, singlet-singlet absorbtion, and triplet-triplet absorption, τ_m is the cavity lifetime, $W_2 n_1$ is the term of optical excitation, τ, τ_T, τ_3 are lifetimes of states 2, T and 3, k is the intersystem crossing rate, n is the concentration of dye molecules.

Let us assume that densities are uniform throughout the medium, and $F \cdot V$ denotes the volume of that part of the cavity which is filled by the lasing solution. We shall rewrite Eqs. (1)—(5) because the magnitude of k is the order of 10^7 [4]. In case of N_2 laser pumping, where the pulsewidth is less than 10 nsec, the first triplet state T cannot have considerable population, consequently the triplet-triplet absorption is negligible, so Eqs. (3) and (4) and the second term $\left(n_m n_T \sigma(\lambda)_{TT} \frac{Fc}{\eta}\right)$ in Eq. (1) can be omitted. Let us further assume that the singlet-excited singlet -state absorbtion is negligible (it is a reasonable assumption in the case of many dyes), and there is no coupling between modes (in mathematical terms the differential equations are separable), and the photon number is uniformly distributed among modes.

This way we get

$$\frac{dn_m}{dt} = n_m \left(n_2 \frac{\sigma(\lambda) Fc}{\eta} - \frac{1}{\tau_m} \right) + \frac{Fc\sigma(\lambda)}{\eta V}, \tag{6}$$

$$\frac{dn_2}{dt} = W_2 n_1 - \frac{n_2}{\tau} + n_2 n_m \frac{m\sigma(\lambda) c}{\eta}. \tag{7}$$

Using the following notation

$$n_0 = \frac{1/\tau_m}{(N_m + 1)\frac{\sigma(\lambda) Fc}{\eta}}, \quad y = \frac{n_2}{n_0}, \quad q = N_m,$$

$$b = \frac{m}{VN_0}, \quad W = \frac{W_2 n_1 \tau_m}{n_0}, \quad a = \frac{\tau_m}{\tau}, \quad x = \frac{t}{\tau_m},$$

our equations can be simply written as follows:

$$\frac{dq}{dx} = q(y-1)+y, \tag{8}$$

$$\frac{dy}{dx} = W-y(a+bq). \tag{9}$$

According to ATKINSON and PACE [3] the cavity lifetime is equal to the following expression

$$\tau_m = \frac{2L}{c\ln(R_1 R_2)},$$

where R_1 and R_2 are the reflectivity of mirrors, and L is the length of the cavity. Conditions of damped-relaxation oscillation can be determined by applying small-signal analysis. If the pumping rate is constant, the laser should reach steady state, *i. e.*

$$\frac{dy}{dx} = \frac{dq}{dx} = 0 \quad \text{and} \quad q = q_0 = \frac{W_0 - a}{b}, \quad y = y_0 \approx 1.$$

If the laser departs from the steady state by y^* and q^* $y^* \ll 1$, $q^* \ll q_0$ we obtain from Eqs. (8) and (9) by substituing $y = y_0 + y^*$ and $q = q_0 + q^*$,

$$\frac{d^2 y^*}{dx^2} + W\frac{dy^*}{dx} + (W-a)y^* = 0, \tag{10}$$

$$\frac{d^2 q^*}{dx^2} + W\frac{dq^*}{dx} + (W-a)q^* = 0. \tag{11}$$

These differential equations describe the damped-relaxation oscillation. Note that these differential equations have periodical solutions providing that $\omega = \sqrt{W - a\frac{W^2}{4}}$ is real. Therefore, we may write

$$y^* = A_y \exp\left(-\frac{W}{2}x\right)\cos\left[\left(\sqrt{W-a-\frac{W^2}{4}}\right)x\right]$$

A_y is determined by the initial conditions. (A similar equation holds for q^*.) For periodical solution it is necessary that $q \leqq 1$, which means: $\tau_m \leqq \tau$. The reason why relaxation oscillation did not occur was that $\frac{\tau_m}{\tau} \ll 1$ was not satisfied [1, 4]. Eqs. (8), (9) were solved by an R—40 computer with the Runge—Kutta method.

The pumping pulse was approximated as follows:

$$W = p \cdot 3.46 \cdot 10^{-4} x^{2.7} \exp(-5.18 \cdot 10^{-2} x)$$

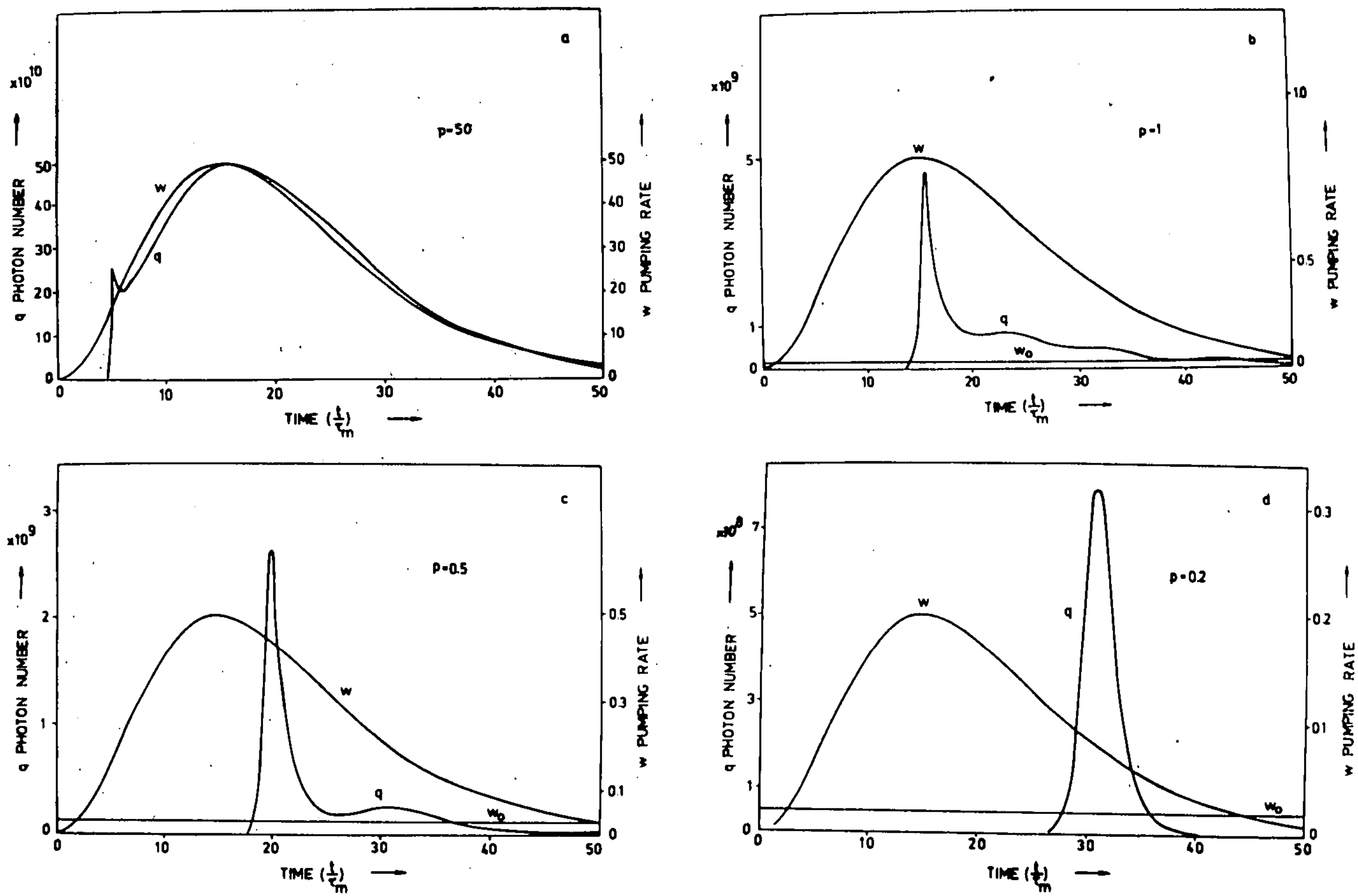


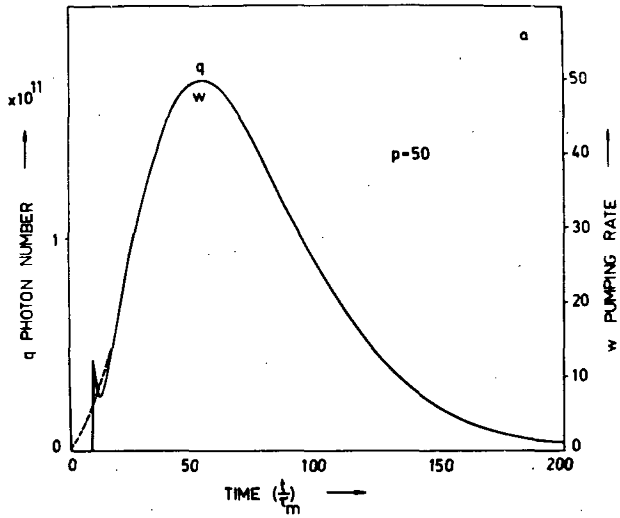
Fig. 2. Numerical solution of coupled-rate equations at the following parameters: $\tau_m = 150$ psec, $\tau = 5.5$ nsec, $b = 2.882 \cdot 10^{-10}$ and (*a*) $W_{max} = 50$, (*b*) $W_{max} = 1$, (*c*) $W_{max} = 0.5$, (*d*) $W_{max} = 0.2$

LIN carried out similar calculations approximating the pumping pulses by trapesoidal function [5]. For calculations the following parameters were used:

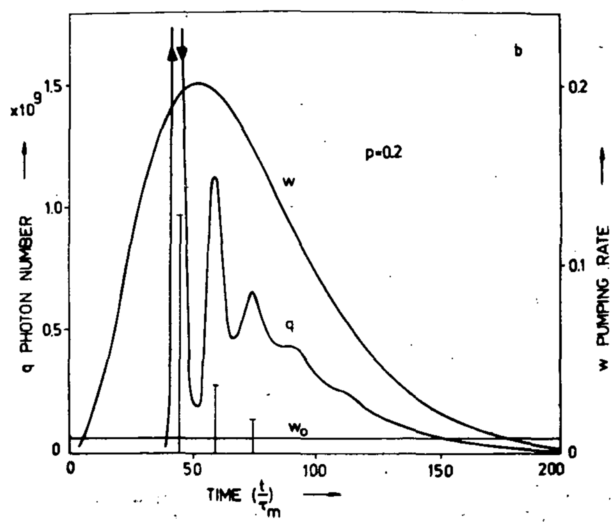
$$\tau_m = 150p\,\text{sec} \quad p = 0.2, 0.5, 1, 50$$

$$\tau_m = 42.3p\,\text{sec} \quad p = 0.05, 0.1, 0.2, 50$$

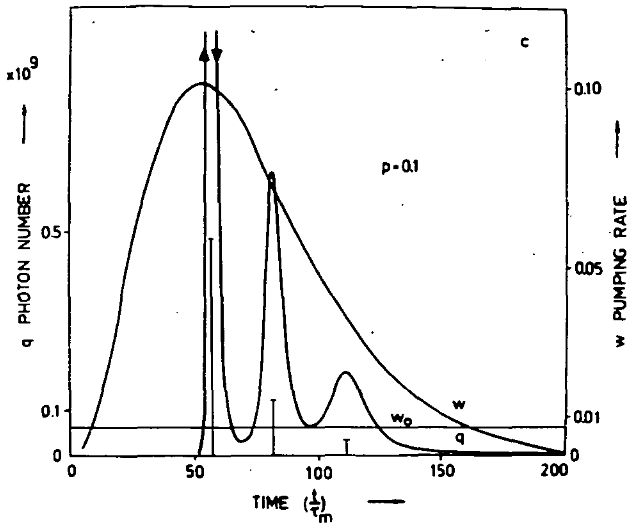
(42.3 psec was the smallest that we could realize experimentally). Fig. 2 *a* shows that, at high pumping levels, the numerical solution is in agreement with the experi-



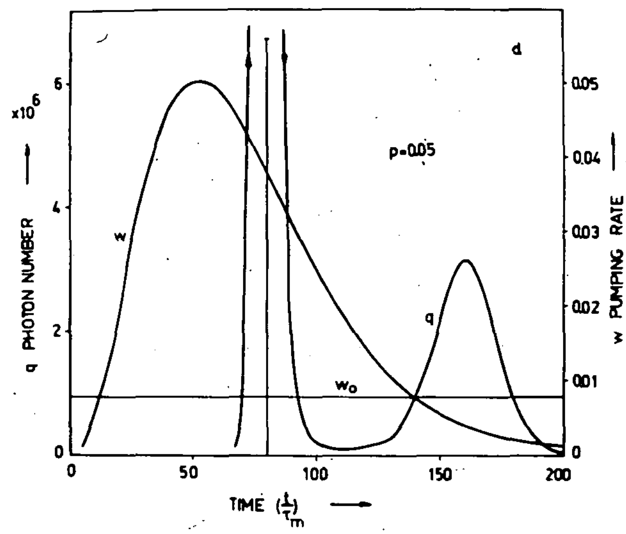
a)



b)



c)



d)

Fig. 3. Numerical solutions of coupled-rate equations at the following parameters: $\tau_m = 43.2$ psec, $\tau = 5.5$ nsec, $b = 8.479 \cdot 10^{-9}$ and (*a*) $W_{max} = 50$, (*b*) $W_{max} = 0.2$, (*c*) $W_{max} = 0.1$, (*d*) $W_{max} = 0.05$

mental results [1, 2, 4]. Fig. 2*b* shows the output pulse in the case of strongly damped relaxation oscillation. As shown above damping is proportional to $W/2$. In Figs. 2*c* and 2*d* the case of small damping is given, and there is no oscillation, because the pumping intensity has been dropped before the evolution of the second pulse.

In Fig. 2*d* there is a single pulse of very low intensity. By decreasing the cavity lifetime the width of pumping pulse is relatively increased, because it is measured in τ_m units. In Figs. 3*a*–3*d* the solutions for 43.2 psec cavity lifetime are shown. Fig 3*a*, Figs. 3*b*, 3*c* how definite relaxation oscillation. It can be seen that on decreasing τ_m the pulsewidth was also decreased *e.g.* the initial pulsewidth in Fig. 2*d* is about 1.2 nsec, while in Fig. 3*b* it is about 0.4 nsec. By changing the pumping intensity and the cavity lifetime the required subnanosecond pulses could be produced [6].

Our experimental arrangement is shown in Fig. 4. The pumping source was a N_2 laser [7] operating at 3371 Å with a typical output power of 0.2 MW, pulse duration of 6 nsec, and repetition rate of 50 pps. The pumping beam was focused by a cylindrical lens onto the front window of the dye cell, containing a $5 \cdot 10^{-3}$ mole/l ethanol solution of Rhodamine 6G. The cavity was formed by the cell wall and mirror M_3. The detector was a FEK—15 KM-type biplanar photodiode, with 500 psec risetime. The output was monitored by a I 2—7-type travelling-wave oscilloscope, or a S7—8-type sampling oscilloscope. The experimental results are summarized in Figs. 5—6. Fig. 5 shows the signal on a travelling-wave oscilloscope; here the dye laser signal was delayed by M_1 and M_2 mirrors. Peaks in the dye laser pulse can be very well seen. Fig. 6*a* shows a typical N_2 laser pulse, Figs. 6*b* and *c* shows typical dye laser pulses at high and low pumping level, respectively. These results are in good qualitative agreement with our theoretical calculations.

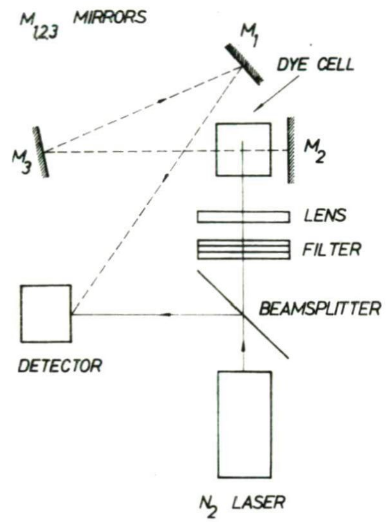


Fig. 4. Experimental arrangement

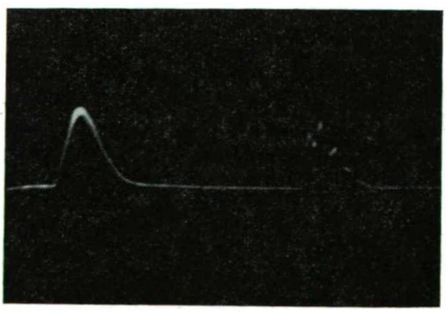


Fig. 5. Time behaviour of N_2 laser (*a*) and dye-laser oscillation (*b*)

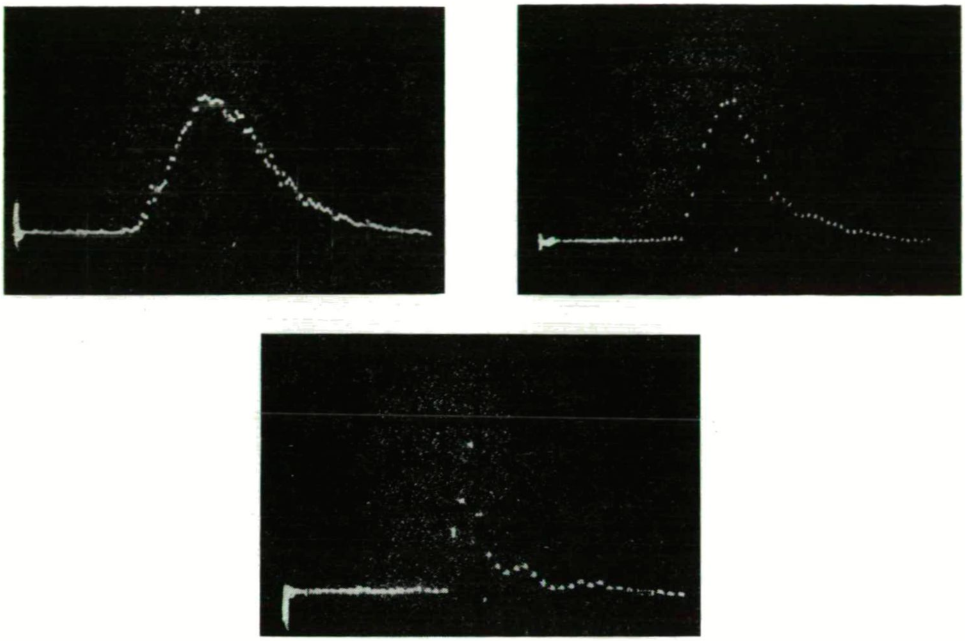


Fig. 6. Oscillograms of N_2 laser (*a*), dye laser at high pumping level (*b*) and dye laser at low pumping level (*c*).
Sweep speed 2 nsec/div, risetime of photodiode 500 psec

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СУБНАНОСЕКУНДНЫЕ РЕЛАКСАЦИОННЫЕ ОСЦИЛЛЯЦИИ В ЛАЗЕРАХ НА КРАСИТЕЛЯХ ПРИ НАКАЧКЕ АЗОТНЫМ ЛАЗЕРОМ

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Рассматривается кинетическая четырехуровневая модель лазера на красителе, возбуждаемого азотным лазером. Теоретически и экспериментально исследованы затухающие осцилляции и генерация субнаносекундных импульсов в лазере на красителях. Численные результаты решений уравнений скоростей и экспериментальные данные находятся в хорошем согласии.