IMPROVED MODEL OF NITROGEN LASER-PUMPED DYE LASERS

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The time behaviour of nitrogen laser-pumped dye lasers was investigated theoretically. The time-space dependent rate equation model of dye lasers was improved. A numerical solution of these equations proved very time consuming. Therefore a simplification of equations was performed. The simplified equations were solved numerically, the delay of dye laser pulses, the oscillation having cavity round-trip time modulation, and the modulation depth were investigated. The production of subnanosecond pulses by long cavity dye lasers, and by two wavelength dye lasers, were demonstrated by the model.

Introduction

The temporal behaviour of laser-pumped dye lasers has been investigated for a long time. One of the first papers of P. P. SOROKIN et al. [1] gave a simple rate equation model of dye lasers, describing the fundamental properties of the dye laser produced by them. J. B. ATKINSON and F. P. PACE investigated this model more precisely [2]. The small signal approximation, and computer solution of rate equations [3, 4] predicted relaxation oscillations, and this phenomenon was found experimentally as well. Moreover, this model proved successful in the investigation of spectral properties of pulsed dye lasers [5].

The most important disadvantage of these models is that, the real resonator configuration cannot be taken into account directly, the cavity decay time alone depends on the cavity length, but this parameter is not affected by the place of the dye cell. A rigorous analysis shows a further problem of these equations, when the change in the photon number, and in the populations is considerable during a round trip time. U. GANIEL et al. eliminated these problems by using time and space dependent rate equations for ring lasers and injection. The validity of their solution is limited by the simplifications used [6]. A more general situation was treated by R. WYATT by using real dye, resonator, and pumping parameters. The solutions showed the possibility of production of subnanosecond dye laser pulses [7].

The aim of this paper is to show a calculation method which has the simplicity of time dependent rate equations but also allows the real resonator construction to be taken into account. By using this method, the most important properties of long cavity dye lasers can be explained.

The dye laser model

For the sake of simplicity let us suppose that:

— the effect of triplet states is negligible

— the excited state absorption is negligible

— the time duration of the thermalization of higher excited state is very short compared to other processes

— photochemical processes are not taken into account

— a two-level system of states is taken into consideration

— the active volume has a square cross section

— the excitation light intensity is constant along the optical axis

— the photon flux and populations perpendicular to the optical axis are constant

— the reflections in the boundary of active volume are not considered

— the divergence of the laser photon flux is equal to the experimental value

— the effect of scattering is negligible

— the reflectivity of the output mirror does not depend on the wavelength

— the divergence of amplified spontaneous emission (ASE) is equal to the angle between the diagonals of the excited volume.

The geometry of the dye laser can be seen in Fig. 1. Therefore, the time and space dependent rate equations are:

$$\frac{\partial N_1(x,t)}{\partial t} = I_p(t)\sigma_a(\lambda_p)N_0(x,t) - N_1(x,t)\int_0^\infty \sigma_e(\lambda)[I_L^+(x,t,\lambda)+I_A^+(x,t,\lambda)+ + I_L^-(x,t,\lambda)+I_A^-(x,t,\lambda)]\,d\lambda$$

$$\pm\frac{dI_L^\pm(x,t,\lambda)}{dx} = N_1(x,t)\sigma_e(\lambda)I_L^\pm(x,t,\lambda)+\tau^{-1}N_1(x,t)E(\lambda)g_L^\pm(x)$$

$$\pm\frac{dI_A^\pm(x,t,\lambda)}{dx} = N_1(x,t)\sigma_e(\lambda)I_A^\pm(x,t,\lambda)+\tau^{-1}N_1(x,t)E(\lambda)g_A^\pm(x)$$

$$N_0(x,t)+N_1(x,t) = N \doteq \text{const.}$$

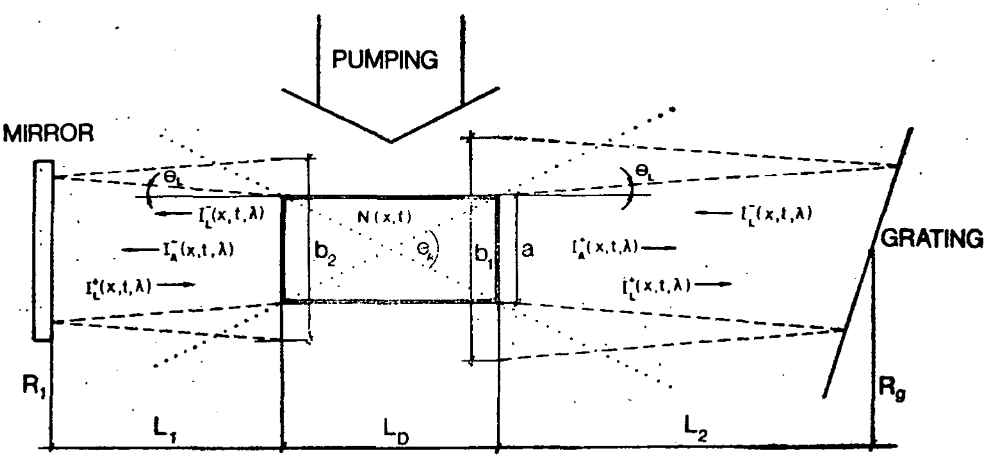

Fig. 1. The dye laser model

where:

$$\frac{d(I^{\pm})}{dx} = \frac{\partial(I^{\pm})}{\partial x} \pm \frac{n}{c} \cdot \frac{\partial(I^{\pm})}{\partial t}$$

supposing:

$$\frac{d\lambda}{dx} = 0.$$

The symbols are the following:
N the density of active molecules,
$N_0(x,t)$, $N_1(x,t)$ the population of ground and excited states,
$I_p(t)$ the pumping power density,
$\sigma_a(\lambda)$, $\sigma_e(\lambda)$ absorption and emission cross sections of the dye,
τ fluorescence lifetime,
$I_L^{\pm}(x, t, \lambda)$ laser photon flux per unit wavelength propagating in the $\pm x$ direction,
$I_A^{\pm}(x, t, \lambda)$ ASE photon flux per unit wavelength propagating in the $\pm x$ direction,
$E(\lambda)$ fluorescence emission spectrum normalized to quantum yield,
$g_L^{\pm}(x) \cdot g_A^{\pm}(x)$ geometrical factor for spontaneous emission,
n the refractive index of solution,
c the velocity of light.
Boundary and initial conditions are the following:

$$I_L^{\pm}(x, 0, \lambda) = 0, \quad I_A^{\pm}(x, 0, \lambda) = 0, \quad N_1(x, 0) = 0,$$

$$I_L^{-}\left(0, t+\frac{L_1}{c}\right) = \frac{a^2}{(a+L_1\theta_L)^2} I_L^{-}(L_1, t),$$

$$I_L^{+}(0, t) = R_1 I_L^{-}(0, t),$$

$$I_L^{+}\left(L_1, t+\frac{2L_1}{c}\right) = \frac{a^2}{b_2^2} R_1 I_L^{-}(L_1, t),$$

$$I_L^{+}\left(L_1+L_0+L_2, t+\frac{L_2}{c}\right) = \frac{a^2}{(a+\theta_L L_2)^2} I_L^{+}(L_1+L_D, t),$$

$$I_L^{-}(L_1+L_D+L_2, t) = R_g I_L^{+}(L_1+L_D+L_2, t), \quad I_A^{+}(L_1', t) = I_A^{-}(L_1+L_D, t) = 0,$$

$$I_L^{-}\left(L_1+L_D, t+\frac{2L_2}{c}\right) = \frac{a^2}{b_1^2} R_g I_L^{+}(L_1+L_D, t), \quad I_A^{-}(L_1, t) = I_A^{+}(L_1+L_D, t).$$

The output of the laser can be calculated as:

$$I_L(0, t) = I_L^{-}(0, t)(1-R_1).$$

The coupled rate equations were solved numerically by a PET2001 computer using finit difference method, supposing:

$$N_1(x, t) \int_0^{\infty} \sigma_e(\lambda)[I_L^{+}(x, t, \lambda)+I_A^{+}(x, t, \lambda)+I_L^{-}(x, t, \lambda)+I_A^{-}(x, t, \lambda)]\, d\lambda =$$

$$= N_1(x, t)\bar{\sigma}_{eL}[I_L^{+}(x, t)+I_L^{-}(x, t)]\, \Delta\lambda_L + N_1(x, t)\bar{\sigma}_{eA}[I_A^{+}(x, t)+I^{-}(x, t)]\, \Delta\lambda_A$$

meaning that spectral changes were not taken into consideration. The parameters were the following: $I_p(t)=5.07\cdot10^{23}t^4e^{-0.75t}\,\frac{\text{photon}}{\text{cm}^2\,\text{s}}$ $\sigma_a(\lambda_p)=2.4\cdot10^{-17}\,\text{cm}^2$, $R_1=0.04$, $R_g=0.6$, $a=0.2$ mm, $\theta_L=3$ mrad, $\theta_A=20$ mrad, $L_1=6$ cm, $L_D=1$ cm, $L_2=15.5$ cm, $\tau=5.5$ ns, $\Delta\lambda_L=1$ nm, $\Delta\lambda_A=10$ nm, $N=3\cdot10^{18}\,\frac{1}{\text{cm}^3}$.

The numerically stable convergent solution can be seen in Fig. 2. The most important informations about this figure:

— the dye laser output has oscillations,

— the amplitude and modulation of pulses change,

— the modulation with $2L/c$ period is due to a group of subpulses going forth and back between the mirror and the grating. Unfortunately, the calculation of this pulse was very time consuming, and for this reason, further simplifications were made. It is obvious that this model has two important parts, the amplifier cell, and a storage and delay system of the photon flux. The amplifier can be simplified without modifying the storage and delay system.

Let us assume that:

$$\int_0^{L_D} N_1(x,t)\,dx = N_1(t)L_D$$

and the spontaneous emission is negligible compared to the laser photon flux *i.e.*:

$$\pm\frac{dI_L^{\pm}(x,t)}{dx} = N_1(t)\bar{\sigma}_{eL}I_L^{\pm}(x,t).$$

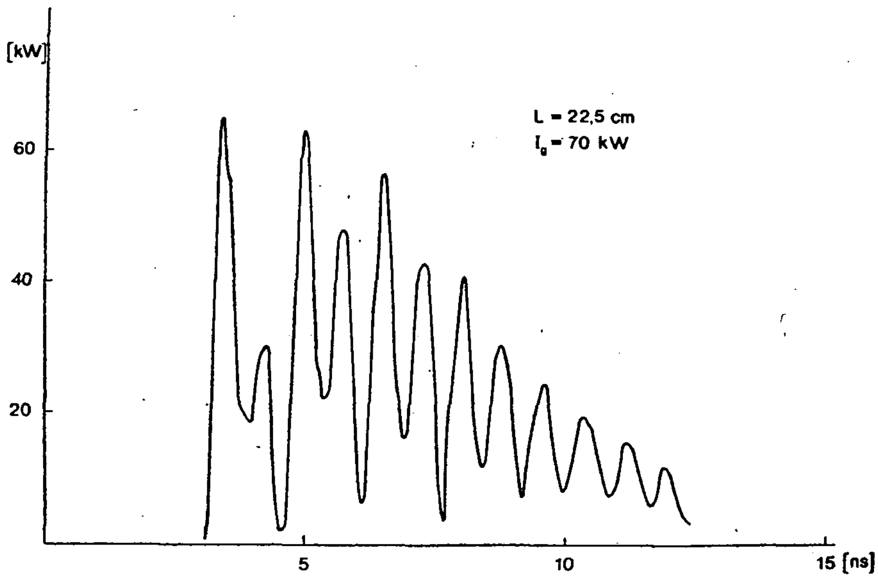

Fig. 2. The numerical solution of time and space dependent rate equations

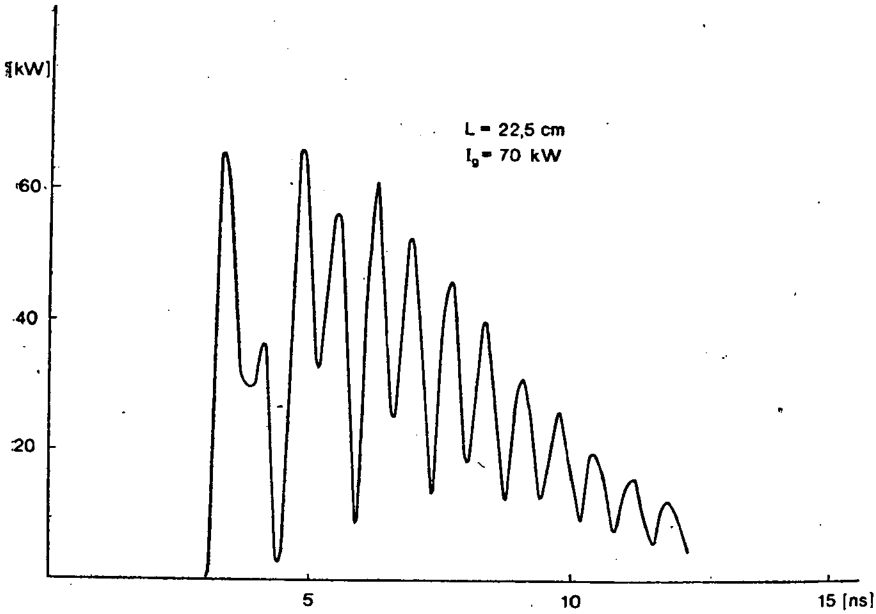

Fig. 3. The numerical solution of simplified equations

The solution of this equation is the following:

$$I_L^+(L_1+L_D,t) = I_L^+(L_1,t)e^{N_1(t)\sigma_{eL}L_D}.$$

The ASE photon flux is:

$$I_A^+(t) = \int_0^{L_D} E(\lambda)\frac{N_1(t)}{\tau}g_A^+ e^{N_1(t)\bar{\sigma}_{eA}(L_D-x)}dx = \frac{E(\lambda)}{\bar{\sigma}_{eA}\tau}[e^{N_1(t)\bar{\sigma}_{eA}L_D}-1].$$

Therefore, the equations to be solved are:

$$\frac{\partial N_1(t)}{\partial t} = I_p(t)\sigma_a(\lambda_p)N_0(t) - \frac{N_1(t)}{\tau} - N_1(t)\bar{\sigma}_{eL}[I_L^+(t)+I_L^-(t)]\Delta\lambda_L - N_1(t)\bar{\sigma}_{eA}[2I_A^+]\Delta\lambda_A,$$

$$I_L^+(L_1+L_D,t) = I_L^+(L_1,t)e^{N_1(t)\bar{\sigma}_{eL}L_D} + sI_A^+(L_1+L_D,t),$$

$$I_L^-(L_1,t) = I_L^-(L_1+L_D,t)e^{N_1(t)\bar{\sigma}_{eL}L_D} + sI_A^-(L_1,t),$$

$$I_A^-(L_1,t) = \frac{E(\lambda)}{\bar{\sigma}_{eA}\tau}[e^{N_1(t)\bar{\sigma}_{eA}L_D}-1],$$

$$I_A^+(L_1+L_D,t) = \frac{E(\lambda)}{\bar{\sigma}_{eA}\tau}[e^{N_1(t)\bar{\sigma}_{eA}L_D}-1],$$

where s is the portion of ASE propagating along the laser photon flux, the initial and boundary conditions are the same as before. When the same parameters are used as for the original equations, the solution can be seen in Fig. 3. The curves of Fig. 2 and Fig. 3 are equal better than 5%. The simplified equations were solved by using a set of different parameters.

Results

The time position of the rise of dye lasers was investigated experimentally in [9]. These measurements showed that: when the cavity length was increased, the delay between exciting and the dye laser pulse also increased. In contrast to this fact, the numerical solution of time dependent rate equations showed the decrease of the delay when the cavity length was increased. This discrepancy disappears in the solution of time and space dependent equations. Fig. 4 shows the calculated time delay as a function of the resonator length. The agreement between theoretical and experimental results is satisfactory, the slope of the curves is equal.

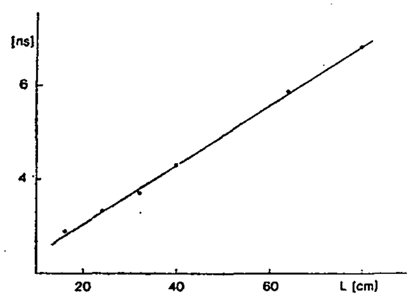

Fig. 4. The calculated delay of dye laser pulses as a function of resonator length

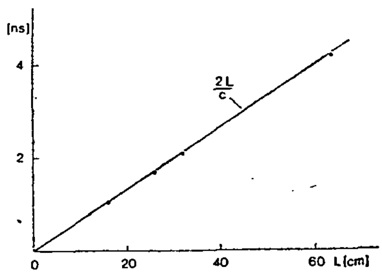

Fig. 5. The calculated time between pulse groups as a function of cavity length

The oscillations of long cavity dye lasers with cavity round trip modulation were discovered and investigated experimentally [8, 9]. The solutions of the time and space dependent equations can describe these important properties of nitrogen laser pumped dye lasers well. The pulsations of Fig. 2, 3 seem to be irregular just as experimental time behaviours do. The detailed study of these pulses showed the cavity round trip time modulation in every case. The number and amplitude of subpulses in Fig. 2, 3 depend on the resonator parameters, *i.e.* front mirror-dye cell distance, and the reflectivity of front mirror and the grating. Fig. 5 shows the time between pulse groups as a function of resonator length. The agreement with experimental results is apparent.

The modulation depth of oscillations of this type increases with the decrease in feedback efficiency. Fig. 6 (a), (b), (c) show the time behaviour of a 16 cm long cavity dye laser as increasing the reflectivity of the grating. The decrease of modulation depth can also be seen very well. Similar results obtained for 32 cm long cavities.

The investigated properties of dye lasers allowed us to give the condition for generation of single subnanosecond pulses. The condition is the following: cavity round trip time must be high enough in comparison with the duration of the exciting pulse. According to experimental conditions, the resonator length was increased to 1 m and the calculated solution can be seen in Fig. 7.

The application of this model to investigating the optimum conditions for subnanosecond pulses, and two-wavelength dye lasers is in progress.

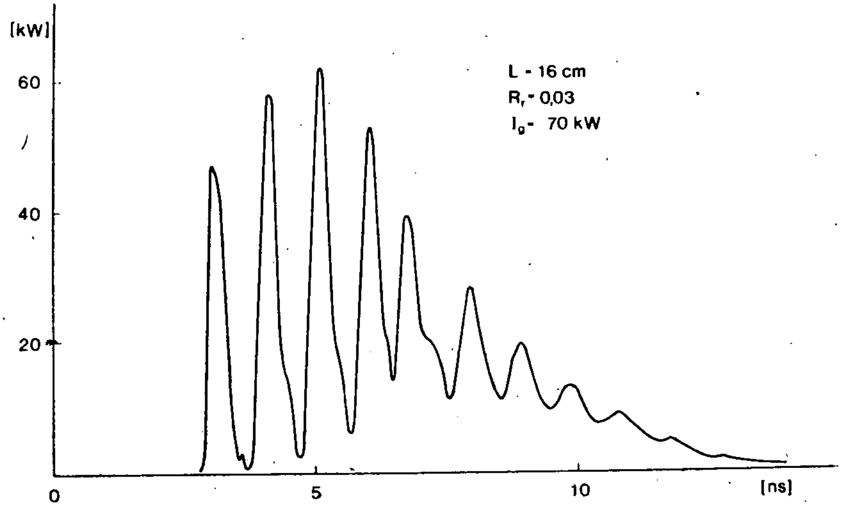

Fig. 6. (a)

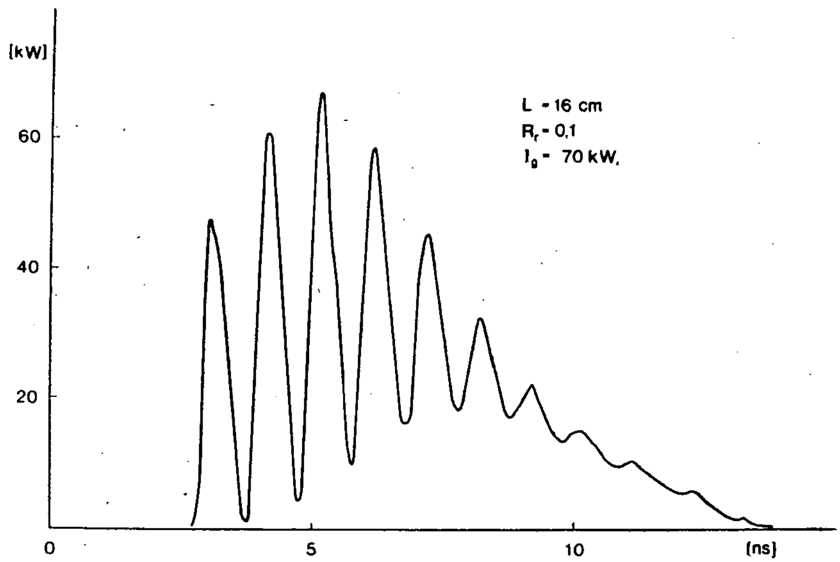

Fig. 6. (b)

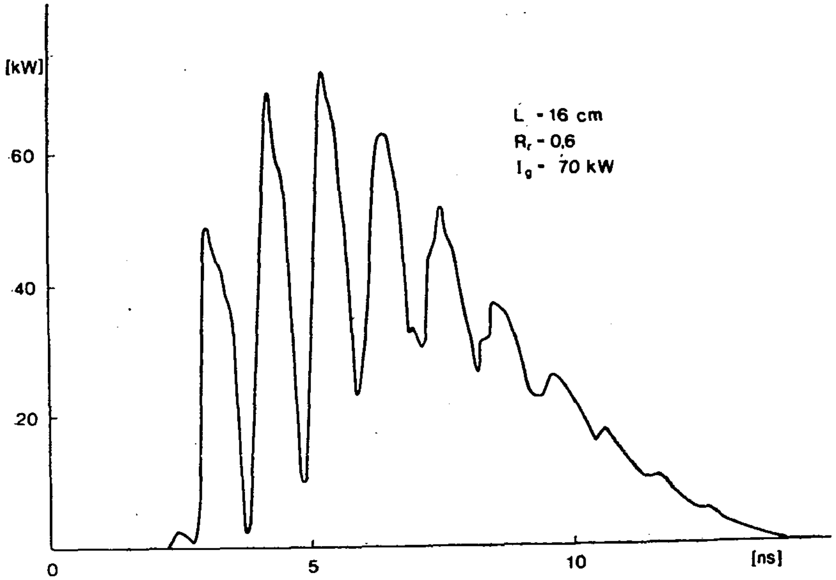

Fig. 6. (c). The modulation depth of subpulses as a function of feedback efficiency ($R_r = R_g$)

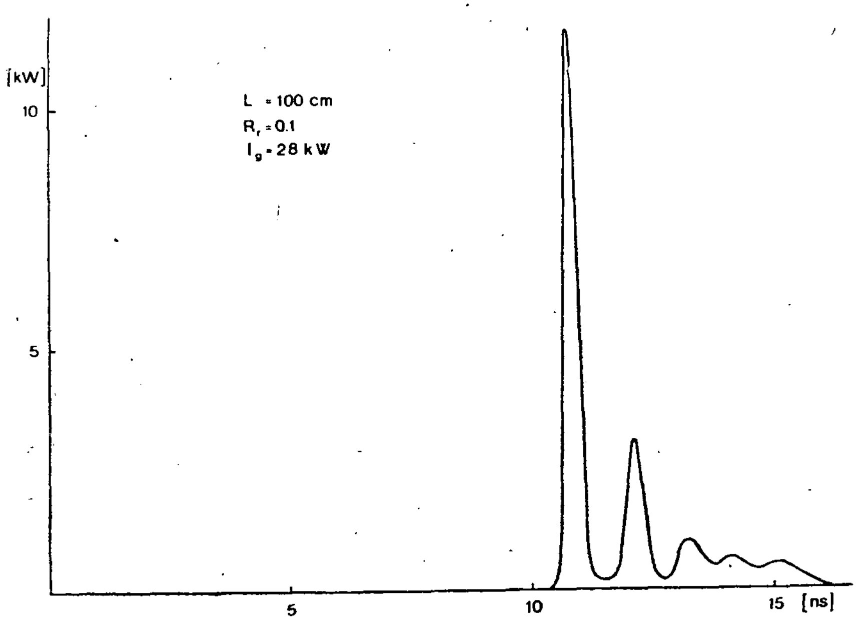

Fig. 7. Single subnanosecond pulse

References

[1] *Sorokin, P. P., J. R. Lankard, E. C. Hammond, V. L. Moruzzi:* IBM J. Res. Develop., **II.** 130 (1967).
[2] *Atkinson, J. B., F. P. Pace:* IEEE J. Quantum Electron. **QE—9,** 569 (1973).
[3] *Lin, Ch.:* IEEE J. Quantum Electron. **QE—11,** 602 (1975).
[4] *Rácz, B., Zs. Bor, G. Szabó, Cs. Zoltán:* Acta Phys. et Chem. Szeged **23,** 367 (1977).
[5] *Juramy, P., P. Flamant, Y. H. Meyer:* IEEE J. Quantum Electron. **QE—13,** 855 (1977).
[6] *Ganiel, U., A. Hardy, D. Treves:* IEEE J. Quantum Electron. **QE—12,** 704 (1976).
[7] *Wyatt, R.:* Applied Physics **21,** 353 (1980).
[8] *Wyatt, R.:* Opt. Comm. **25,** 375 (1978).
[9] *Rácz, B., Zs. Bor, G. Szabó, S. Szatmári:* this volume.

УЛУЧШЕНИЕ МОДЕЛИ ЛАЗЕРОВ НА КРАСИТЕЛЕ, ВОЗБУЖДЕННЫХ АЗОТНЫМ ЛАЗЕРОМ

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Теоретически исследована временная характеристика лазеров на красителях, возбужденных азотным лазером. Развиты кинетические уравнения зависящие от пространства и времени. Числовое решение этих уравнений требует много времени, поэтому было осуществлено упрощение уравнений. Было проведено числовое решение упрощеных уравнений. Проведено исследование задержки импульсов лазера на красителе, осцилляции, имеющей модуляцию возвратно-поступательного времени резонатора, и глубина модулации. Были показаны на модели субнаносекундные импульсы из длинного резонатора и лазера на красителе с двумя длиннами волн.