# TIME DEPENDENCE OF THE GENERATION POWER OF ORGANIC DYES

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## Abstract

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The theoretical calculation of the power of nonstationary generation was carried out in (1). In the calculation it was assumed that the gain coefficient is equal to the loss coefficient. This assumption is not always valid, and the limits of its applicability are not clear in advance. In the present work a more rigorous calculation of the temporal characteristics of generation is performed, with consistent allowance for the accumulation of particles at a metastable level. Absorption of radiation by excited particles was not taken into account.

Consider an elementary volume of a generating dye solution. The balance equations for determining the populations $n_1, n_2, n_3$ (see Fig. 6 in (1)) have the form (1)

$$dn_3/dt = B_{13}(\nu_\mathrm{p})u_\mathrm{p}(n_1 - n_3e^{-b}) - B_{31}(\nu_\mathrm{g})u_\mathrm{g}(n_3 - n_1e^{-a}) - (p_{31} + p_{32})n_3; \quad (1)$$

$$dn_2/dt = p_{32}n_3 - p_{21}n_2; \quad (2)$$

$$n_1 + n_2 + n_3 = n, \quad (3)$$

where $B_{ij}(\nu)$ is the Einstein coefficient; $p_{ij}$ are the probabilities of spontaneous transitions; $u_\mathrm{p}$ and $u_\mathrm{g}$ are the radiation densities of the pump and of the generated emission; $\nu_\mathrm{p}$ and $\nu_\mathrm{g}$ are the corresponding frequencies; $a = (\nu_\mathrm{el} - \nu_\mathrm{g})/kT$, $b = (\nu_\mathrm{p} - \nu_\mathrm{el})/kT$. The value of $u_\mathrm{p}$ is specified, while the density $u_\mathrm{g}$ is unknown. To determine it one uses the usual equation (2)

$$du_\mathrm{g}/dt = v\mu(\chi y - k_\mathrm{loss})u_\mathrm{g}, \quad (4)$$

where $v$ is the speed of light, $\mu$ is the ratio of the optical length of the active layer to the total optical length of the resonator; $\chi y$ is the gain coefficient, $\chi = B_{31}(\nu_\mathrm{g})h\nu_\mathrm{g}n/v$ is the limiting value of the gain coefficient,

$$y = \frac{n_3}{n} - \frac{n_1}{n} e^{-a} \tag{5}$$

is the degree of inversion; $k_{\text{loss}}$ is the loss coefficient calculated per unit volume. From (1)–(3) it follows that

$$dy/dt = G - Dy - B'_{31}(\nu_{\text{g}}) y u_{\text{g}} - (G - p_{21} e^{-a}) n_2/n; \tag{6}$$

$$d(n_2/n)/dt = p'_{32}(e^{-a} + y) - \alpha n_2/n, \tag{7}$$

where

$$B'_{31}(\nu_{\text{g}}) = B_{31}(\nu_{\text{g}})(1 + e^{-a}), \quad p'_{32} = p_{32}/(1 + e^{-a}), \quad \alpha = p'_{32} e^{-a} + p_{21},$$

$$G = B_{13}(\nu_{\text{p}}) u_{\text{p}} [1 - e^{-(a+b)}] - (p_{31} + p_{32}) e^{-a}; \tag{8}$$

$$D = B_{13}(\nu_{\text{p}}) u_{\text{p}} (1 + e^{-b}) + (p_{31} + p'_{32}). \tag{9}$$

The system of equations (4), (6), (7) with respect to $u_{\text{g}}, y$, and $n_2$ can be integrated numerically* if the form of the function $u_{\text{p}}(t)$ is specified. More general information can be obtained by using for the solution the approximate method of A. M. Samson (2).

* In work (3) such a calculation was performed for certain values of the parameters without taking into account deactivation of the second level.

Let us set, in the first approximation,

$$y = y^* = k_{\text{loss}}/\chi; \tag{10}$$

$$n_2(t) = n p'_{32}(e^{-a} + y^*)(1 - e^{-at}) + n_2(0) e^{-at}; \tag{11}$$

$$u^*_{\text{g}}(t) = \frac{[1 - e^{-(a+b)}](1 - n_2/n) - y^*(1 + e^{-b})}{y^*} [u_{\text{p}}(t) - u'_{\text{p}}(t)], \tag{12}$$

where

$$u'_{\text{p}}(t) = \frac{1}{B_{13}(\nu_{\text{p}})} \cdot \frac{(p_{31} + p'_{32})(e^{-a} + y^*) - (p_{31} + p'_{32} - p_{21}) e^{-a} n_2/n}{[1 - e^{-(a+b)}](1 - n_2/n) - y^*(1 + e^{-b})}, \tag{13}$$

and we shall seek the solution of (6), (7) in the form

$$u_{\mathrm{g}}(t) = u_{\mathrm{g}}^{*}(t) + \Delta u_{\mathrm{g}}, \qquad y = y^{*} + \Delta y. \tag{14}$$

Expression (11) follows from (7) when (10) is satisfied; expression (12), from (6) when (10) and (11) are satisfied.

If the values $\Delta y$ and $\Delta u_{\mathrm{g}}$ were equal to zero, this would mean that at each instant $t$ a quasi-stationary regime is established, determined by the value $u_{\mathrm{p}}(t)$ and $n_2(t)$. For $u'_{\mathrm{p}}(t) > u_{\mathrm{p}}(t)$, quasi-stationary generation is impossible. In view of this, the quantity $u'_{\mathrm{p}}(t)$ may be called the threshold value of the pump radiation for quasi-stationary generation. As particles accumulate at the metastable level, the value $u'_{\mathrm{p}}(t)$ increases. Equating $u_{\mathrm{p}}(t)$ and $u'_{\mathrm{p}}(t)$ and solving the resulting equation with respect to $t$, one can find the moment of breakdown of quasi-stationary generation.

Substituting (13) and (10)–(12) into (6) and (4), we obtain

$$d\Delta y/dt = -[B'_{31}(\nu_{\mathrm{g}})u_{\mathrm{g}}^{*} + D]\Delta y - B'_{31}(\nu_{\mathrm{g}})y^{*}\Delta u_{\mathrm{g}}; \tag{15}$$

$$d\Delta u_{\mathrm{g}}/dt = \nu\mu\chi u_{\mathrm{g}}^{*}\Delta y - du_{\mathrm{g}}^{*}/dt. \tag{16}$$

In the calculation the terms $\Delta y \Delta u_{\mathrm{g}}$ have been omitted; as will be seen below, in most cases they are indeed small.

For generation one chooses dyes with the smallest possible values of $p_{32}$ and, consequently, $a$. Therefore the accumulation of particles at level 2 is comparatively slow. When dye generation is excited by pulsed lamps, the change in $u_{\mathrm{p}}(t)$, and consequently in $u_{\mathrm{g}}^{*}(t)$, is also small. The system (15)–(16) can be solved over a certain time interval by assuming that the quantities $n_2$, $u_{\mathrm{g}}^{*}$, and $du_{\mathrm{g}}^{*}/dt$ within the interval are practically unchanged. In some cases a similar procedure is also valid for excitation of dye generation by laser sources.

The solution of (15)–(16) under the assumption made has the form

$$\Delta y = \frac{1}{\nu\mu\chi u_{\mathrm{g}}^{*}}\left(c_1\lambda_1 e^{\lambda_1 t} + c_2\lambda_2 e^{\lambda_2 t}\right) + \frac{1}{\nu\mu\chi u_{\mathrm{g}}^{*}}\frac{du_{\mathrm{g}}^{*}}{dt}; \tag{17}$$

$$\Delta u_{\mathrm{g}} = c_1 e^{\lambda_1 t} + c_2 e^{\lambda_2 t} + \frac{B'_{31}(\nu_{\mathrm{g}})u_{\mathrm{g}}^{*} + D}{\nu\mu k_{\mathrm{loss}} B'_{31}(\nu_{\mathrm{g}})u_{\mathrm{g}}^{*}}\frac{du_{\mathrm{g}}^{*}}{dt}, \tag{18}$$

where $c_1, c_2$ are constants of integration, and

$$\lambda_{1,2} = -\frac{1}{2}[B'_{31}(\nu_{\mathrm{g}})u_{\mathrm{g}}^{*} + D] \pm \frac{1}{2}\{[B'_{31}(\nu_{\mathrm{g}})u_{\mathrm{g}}^{*} + D]^2 - 4\nu\mu k_{\mathrm{loss}} B'_{31}(\nu_{\mathrm{g}})u_{\mathrm{g}}^{*}\}^{1/2}. \tag{19}$$

In the most typical cases the inequality

$$[B'_{31}(\nu_\text{g})u^*_\text{g} + D]^2 \gg 4\nu\mu k_\text{loss} B'_{31}(\nu_\text{g})u^*_\text{g} \tag{20}$$

is valid.

It is sometimes violated only at very small pump powers. If $2\nu\mu k_\text{loss} < p_{31} + p'_{32}$, then relation (20) is satisfied for any pump–

exceeding the threshold value. From (20) and (19) it follows that

$$\lambda_1 = -[B'_{31}(\nu_\text{g})u^*_\text{g} + D], \tag{21}$$

$$\lambda_2 = -\nu\mu k_\text{loss} \frac{B_{31}(\nu_\text{g})u^*_\text{g}}{B'_{31}(\nu_\text{g})u^*_\text{g} + D}. \tag{22}$$

Even for very small excesses above threshold ($u_\text{p} > 1.05u'_\text{p}$), the quantity $\lambda_2$ becomes close to $\nu\mu k_\text{loss}$, and the modulus of $\lambda_1$ is always greater than the modulus of $\lambda_2$. When lasing is excited by pulsed lamps, resonators with small loss coefficients of the order of 0.01 $\text{cm}^{-1}$ are usually used. In these cases $\lambda_2 \sim -10^8\ \text{s}^{-1}$. The first two terms of (17) and (18) reach zero in a time $\sim 10^{-8}$ s, and therefore, when calculating the properties of lasing excited by pulsed lamps ($\Delta t \sim 10^{-5}$–$10^{-4}$ s), they may be neglected. If, however, lasing is excited by laser sources of short duration ($\Delta t \sim 10^{-8}$ s), then the second terms of (17) and (18) are significant, especially at the initial moments of time. The formulas (10) and (12) may be used for laser excitation only when $\mu k_\text{loss} > 0.1\ \text{cm}^{-1}$.

The arguments given are valid practically for any $n_2/n$. Accumulation of particles on the metastable level can cause a sharp decrease in $|\lambda_2|$ (down to zero) only in the case when the quantity $n_2/n$ approaches unity and the lasing threshold approaches infinity.

It is also not difficult to show that the third terms of (17)–(18) are, as a rule, very small, and their inclusion may make sense only near the lasing threshold. Already for $u_\text{p} > 1.1u'_\text{p}$

$$\frac{\Delta y}{y^*} = \frac{\Delta u_\text{g}}{u^*_\text{g}} = \frac{1}{\nu\mu k_\text{loss}} \frac{1}{u^*_\text{g}} \frac{du^*_\text{g}}{dt}. \tag{23}$$

The quantity

$$\frac{1}{u^*_\text{g}} \frac{du^*_\text{g}}{dt}$$

is somewhat smaller than $1/\Delta t$, where $\Delta t$ is the duration of the lasing pulse. With lamp pumping, $\Delta u_{\mathrm{g}}$ is several orders of magnitude smaller than $u_{\mathrm{g}}^*$. When lasing is excited by laser sources of short duration, the ratio (23) is small only for $\mu k_{\mathrm{loss}} > 0.1\ \mathrm{cm}^{-1}$.

Thus, the formulas of the quasi-stationary regime (10)–(12) correctly describe the properties of dye lasing excited by pulsed lamps. When excitation is by laser sources, they may be applied for $\nu\mu\Delta t k_{\mathrm{loss}} > 1$.

In some cases, pulsations of the density of the generated radiation about its quasi-stationary value (12) may occur. Such a situation will be realized for complex $\lambda$, i.e., according to (20), in the interval of pump-radiation densities from $u_{\mathrm{p}}^{(1)}$ to $u_{\mathrm{p}}^{(2)}$, determined by the relation (for $p_{21} \ll p_{31} + p_{32}$)

$$B_{13}(\nu_{\mathrm{p}})u_{\mathrm{p}} = (p_{31}+p'_{32})e^{-a} + \frac{2k_{\mathrm{loss}}}{\chi(1-n_2/n)^2}\left\{\nu\mu k_{\mathrm{loss}}\left[1-\frac{n_2}{n}-\frac{k_{\mathrm{loss}}}{\chi}(1+e^{-b})\right] \pm \sqrt{M}\right\}, \tag{24}$$

where

$$M = \nu\mu k_{\mathrm{loss}}\left\{\nu\mu k_{\mathrm{loss}}\left[1-\frac{n_2}{n}\frac{k_{\mathrm{loss}}}{\chi}(1+e^{-b})\right]^2 - (p_{31}+p'_{32})\left(1-\frac{n_2}{n}\right)^2(1+e^{-a})\right\}. \tag{25}$$

The value of $M$ must be greater than zero. This condition is realized only for large loss coefficients. For not too large $n_2$ it is equivalent to the inequality

$$\nu\mu k_{\mathrm{loss}} > p_{31} + p'_{32}. \tag{26}$$

The decrement of damping of the pulsations is very large. In view of this, observation of pulsations is possible only at very small $u_{\mathrm{n}}$ (near $u'_{\mathrm{n}}$) in the first instants of time.

A numerical solution of the system of equations (6), (7) and equation (4) with an additional term describing spontaneous emission was carried out by P. A. Karamaliev. The calculations show that the quasistationary solution (10)–(12) correctly describes the process. Deviations are observed only in the very first instants of formation of the generation. The experimental data obtained in our laboratory agree with (10)–(12). In some cases pulsations of the radiation are observed.

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## CITED LITERATURE


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*Note: Figure translations are in progress. See original paper for figures.*