

# Nonlinear dependence of the delayed luminescence yield on the intensity of irradiation in the framework of a correlated soliton model

Larissa Brizhik\*

*Bogolyubov Institute for Theoretical Physics, 03143 Kyiv, Ukraine*Francesco Musumeci, Agata Scordino,<sup>†</sup> Maurizio Tedesco, and Antonio Triglia*Dipartimento di Metodologie Fisiche e Chimiche per l'Ingegneria, Unità INFN, Università di Catania, viale A. Doria 6, I-95125 Catania, Italy*

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We generalize the correlated soliton model in order to describe the delayed luminescence arising from biological systems after their exposition to the irradiation by relatively high dose (high intensity and/or long duration of irradiation). The quantum yield of the delayed luminescence is calculated as a function of the irradiation and is shown to depend nonlinearly on the intensity and dose of the irradiation. At relatively low intensity, the yield of luminescence increases with increasing dose, and monotonously reaches saturation. At high intensity of the irradiation, the yield of the photosystem under study is restricted from above by the concentration of photosystem units. As a result, the total yield of the delayed luminescence first increases with the dose till the maximum value that, in the general case, is less than the maximum number of available photosystem units. With further increase of the dose, the yield gradually decreases, reaching the saturation value at large dose of illumination. These results are obtained within the steady state approximation in the description of the luminescence kinetics. To check the applicability of this approximation at high levels and large time of illumination, the corrections to the steady state solution have been calculated, and shown to decrease exponentially with increase in time till the small finite constant value. The results of the theoretical model are shown to describe well the experimental data on the dose dependence of the quantum yield of the luminescence of algae *Acetabularia acetabulum*, for which the correlated soliton model describes well the kinetics of the delayed luminescence at low levels of irradiation.

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## I. INTRODUCTION

The phenomenon of delayed luminescence (DL), which consists of photoinduced light emission long time (seconds and more) after illumination, takes place not only in conventional solid state systems, but in biological systems as well [1]. It has been actively studied during the last period and a lot of experimental data has been accumulated. The studies of biosystems reveal a number of characteristic properties of the phenomenon, which are sensitive to the chemical, physical, and physiological state of the system and can give the global information about the state of the organism [2–12]. This is one of the main reasons of interest to studying the mechanisms of the DL and its importance for medical investigations, food and water quality control, etc.

In our previous papers [13,14] we have developed the soliton model, which (as has been shown there) describes both qualitatively and quantitatively the main features of the DL from biosystems. Among these features the most important are the linear in a log-log scale kinetics of the DL in a wide time interval, the same time trends for the various components of the emission spectrum, the nonlinear dependence of the initial intensity of DL,  $I(t=0)$ , on the intensity of the stimulating light, and others.

The solitons, which give rise to the DL, are formed in the one-dimensional chains due to the interaction of electrons with the local deformation of a chain at intermediate values of the electron-phonon coupling constant. The concept of molecular solitons or one-dimensional large polaron-type states, which participate in charge and energy transport along macromolecules during the metabolic processes, was first suggested by Davydov and Kislukha [15,16]. The polypeptide chains in these macromolecules are formed by the periodically placed peptide groups and are characterized by the strong exchange and/or resonance interaction between the neighboring molecules, while the interaction between the chains is much weaker. These characteristic properties of biological macromolecules favor the existence of coherent collective electron and exciton states, in general, and solitons in particular. Moreover, it has been shown in Ref. [14] that the account of the correlation between electrons, which are self-trapped in macromolecules in the soliton states, gives better fit of the experimental data for the algae *Acetabularia acetabulum* (*A.a.*) than the noncorrelated model. These unicellular algae possess a large variety of macroscopically long (up to few millimeters) quasi-one-dimensional macromolecular structures, such as  $\alpha$ -helical polypeptide proteins, actin filaments, microtubules, etc., due to which the correlation between solitons is essential. Indeed, the correlation between the DL and chloroplast organization has been experimentally proved [17].

Both correlated and noncorrelated soliton models describe the kinetics of the DL starting from the time moment when

\*Email address: brizhik@bitp.kiev.ua

<sup>†</sup>Email address: demone@dmfci.unict.it

the illumination is switched off. As a result, theoretical models fit the experimental data well on the dependence of the quantum yield of the DL on the intensity and dose of irradiation at their not very large values. In this case the total number of photons emitted in the process of DL (quantum yield of the DL) is proportional to the dose of illumination. To study the dependence of quantum yield of the DL on the intensity and dose of illumination in the wide intervals of these values, the models have to be changed in such a way that they are valid starting from the initial time of the whole process, i.e., to describe also the process of illumination with account of the luminescence processes during the time of illumination. Such model is developed in the present paper.

It is shown that in the general case the quantum yield of the DL is a nonlinear function of the intensity and dose of illumination. At relatively small intensities of the illuminating light, the quantum yield of the DL increases with dose increasing, and reaches monotonously some saturation at long enough time of illumination. The situation is different at large intensities of illumination: there is some optimum value of the dose when the quantum yield of the DL takes the maximum value, and with further increase of the dose, the quantum yield decreases gradually reaching some saturation, which is less than the maximum value at the optimum dose. The corresponding results of this model are shown to explain the experimental data for alga *A.a.* in a large interval of values of the intensity and dose of illumination.

## II. KINETICS OF THE DELAYED LUMINESCENCE

The general scheme of the process of delayed luminescence is the following [13,14]. Due to the illumination of the system, electrons released in the ionized centers of luminescence (the charge separation complexes) are with a certain probability self-trapped in the macromolecules, which from the point of view of electronic structure are quasi-one-dimensional systems with filled valence band and empty conduction band. The luminescence arises from the decay of these localized states into the conductive electron band with the following fast transition into the recombination centers, i.e., ionized centers of luminescence. The experimentally measured fluorescence and DL emission spectra [17] reveal no energy shift, therefore, the probability of the direct transitions of electrons from the self-trapped states into reaction centers can be neglected.

According to the above, the electron-hole recombination processes determine the intensity of DL,

$$I = -\frac{dn}{dt}. \quad (1)$$

In the presence of the illumination, the kinetics of the DL is described by the following system of equations:

$$\frac{dn}{dt} = -p_{rec}Nn + Y, \quad (2)$$

$$\frac{dN}{dt} = p_{diss}\nu - p_{rec}Nn - p_{loc}N(\nu_0 - \nu) + Y, \quad (3)$$

$$\frac{d\nu}{dt} = -p_{diss}\nu + p_{loc}N(\nu_0 - \nu), \quad (4)$$

where  $n$ , the number of the ionized reaction centers, is connected with the number of free electrons in the conduction band,  $N$ , and the number of electrons in localized (bi)soliton states,  $\nu$ , by the relation

$$n = N + \nu. \quad (5)$$

In the system above,  $Y = Y(n)$  is the quantum yield of the ionized reaction centers due to the illumination,  $\nu_0$  is the number of the available localized states;  $p_{diss}$ ,  $p_{rec}$ , and  $p_{loc}$  are the rates of (bi)soliton dissociation, of electron-hole recombination at the reaction center, and of electron localization, respectively (all rates are calculated per unit time). Furthermore,  $p_{loc}$  is determined by the energy of electron localization in the corresponding state, and therefore, in the general case, it depends on  $\nu$ :  $p_{loc}(\nu) = E_{loc}(\nu)/\hbar$ .

In the case where solitons are noncorrelated (at very small concentrations, in not long enough macromolecules, etc.), electrons localize in the independent (bi)soliton states, and the probability of electron localization does not depend on soliton concentration (this corresponds to the model suggested in Ref. [13]). In the opposite case of strongly correlated coherent electrons, the dependence of the energy of localized level on the concentration of electrons is essential. We consider this latter case in the present paper, and in this case the relation is valid (see Ref. [18]):

$$p_{loc} = p_0 \left(1 - \frac{\nu}{\nu_0}\right), \quad (6)$$

where

$$p_0 = \frac{Jg_0^2}{3\hbar}, \quad \nu = 2\delta, \quad (7)$$

$\nu_0 = 2\delta_0$  is determined by the maximum available concentration of bisolitons in a macromolecule,  $\delta_0 = 2g/\pi^2$ , which is related to the maximum capacity of electron transport reactions. Here  $g$  is the dimensionless electron-phonon coupling constant, and  $J$  is the exchange interaction constant of a polypeptide chain.

To estimate the yield of the reaction centers due to the illumination, it should be taken into account that the total number of electrons that participate in the process of luminescence,  $n$ , is restricted from above by the maximum value equal to the available number of the reaction centers in the system,  $n_{max}$ . For instance, in the biological systems that possess photosystem, this maximum value is related to the concentration of photosystem units. Therefore, the quantum yield of the reaction centers,  $Y$ , in the general case is nonlinear: it depends on the intensity of illumination,  $I_{ill}$ , and is a function of the ratio  $n/n_{max}$ . It is known experimentally that at high intensities of illumination, the inhibition of photosynthesis occurs [19–21]. Taking this into account, we can assume that the quantum yield of the reaction centers can be approximated by a power series

$$Y(n) \approx Y_0 - Y_1 \frac{n}{n_{max}} + Y_2 \frac{n^2}{n_{max}^2} + O\left(\frac{n^3}{n_{max}^3}\right), \quad (8)$$

where  $Y_0, Y_1, Y_2$  are constants proportional to the intensity of the illuminating light,  $I_{ill}$ .

For further study, it is convenient to introduce the dimensionless time

$$\tau = p_{diss} t, \quad (9)$$

dimensionless variables

$$x = \frac{\nu}{\nu_0}, \quad y = \frac{N}{\nu_0}, \quad z = \frac{n}{\nu_0}, \quad (10)$$

and dimensionless parameter of the kinetics rate,

$$\gamma = \frac{p_0}{p_{rec}}, \quad (11)$$

which turn out to be essential for the kinetics of the DL (see also Refs. [13,14]). Obviously, due to Eq. (5), the relation  $z = x + y$  is valid.

In these units the DL intensity is determined from the equation

$$I(\tau) = -\frac{dz}{d\tau}, \quad (12)$$

where

$$I(\tau) = \frac{I(t)}{p_{diss} \nu_0}, \quad (13)$$

and the system of equations (2)–(4) takes the form

$$\frac{dz}{d\tau} = -ayz + P(z), \quad (14)$$

$$\frac{dy}{d\tau} = x - ayz - a\gamma y(1-x)^2 + P(z), \quad (15)$$

$$\frac{dx}{d\tau} = -x + a\gamma y(1-x)^2. \quad (16)$$

Here, according to Eq. (8),

$$P(z) \approx P_0 - P_1 z + P_2 z^2 \quad (17)$$

and

$$a = \frac{p_{rec} \nu_0}{p_{diss}}. \quad (18)$$

### III. STEADY STATE REGIME

It is known that the rate of (bi)soliton dissociation,  $p_{diss}$ , is much smaller than the decay rate of band electrons [22]. Hence, the relation  $y \ll x$  is fulfilled, which corresponds to the quasistationary (steady state) regime,

$$\frac{dy}{d\tau} \ll \frac{dx}{d\tau}. \quad (19)$$

In this case we get from Eqs. (15) and (16) with account of the relation  $z = x + y$ , the following equality:

$$y = \frac{z + P(z)}{a[z + \gamma(1-z)^2]}, \quad (20)$$

and therefore,

$$\frac{dz}{d\tau} = \frac{1 - 2z + \left[1 - \frac{1}{\gamma P(z)}\right] z^2}{1 + \left(\frac{1}{\gamma} - 2\right) z + z^2} P(z). \quad (21)$$

Thus, the system of equations (1)–(4), which determines the whole process of luminescence (including illumination of the system and the following delayed luminescence) is reduced to the systems (12) and (21). The first stage of the illumination is described by Eqs. (12) and (21) at the time interval  $0 < \tau < \tau_{ill}$ . Calculation of the value  $z(\tau_{ill})$  by integrating Eq. (21) from  $\tau = 0$  till  $\tau = \tau_{ill}$  gives the value  $z_{tot} = z(\tau_{ill})$ , which gives us the initial number of the ionized reaction centers and determines the level of excitation for the process of the delayed luminescence. This process takes place starting from the time moment  $\tau_0 = \tau_{ill}$  and is described by the system of equations (1)–(4), or equivalently, by systems (12) and (21), in which one has to set  $P = 0$ . It is easy to see that this system coincides with the system of equations obtained in Ref. [14], which is written there for the values that are renormalized with respect to  $n_0$ .

Therefore, the quantum yield of the DL, arising from illuminating the system during finite time interval  $0 < \tau < \tau_{ill}$ , i.e., the total number of photons emitted by the system after switching off the illumination, is given by the value  $z(\tau_{ill})$  calculated in the result of integrating Eq. (21) from  $\tau = 0$  till  $\tau = \tau_{ill}$ . This integration cannot be carried out in the general form even for the case of approximation (17). Below we consider separately the cases of low and high intensities of illuminating light, or, in other words, of low and high pumping.

### IV. LINEAR PUMPING

In this section we consider the linear pumping, i.e., the situation where the intensity of the illuminating light is not very high, so that in the end of the process of illumination the total number of ionized reaction centers is expected to be less than the value  $n_{max}$ . In this case the nonlinearity of pumping is negligibly small, and the yield of the ionized reaction centers is constant:

$$P(z) = P_0. \quad (22)$$

It is easy to obtain the explicit relation  $z(\tau)$  by integrating Eq. (21) after separating variables  $z$  and  $\tau$ :

$$\tau = \frac{\gamma}{P_0\gamma - 1} z + A \log[R(z)] - B \log[S(z)]. \quad (23)$$

Here  $a$  is the constant given by Eq. (18),  $A$  and  $B$  are constants that depend on the parameters of kinetic rate and linear pumping:

$$A(P_0, \gamma) = \frac{\gamma(2 + P_0) - 1}{2(P_0\gamma - 1)^2}, \quad (24)$$

$$B(P_0, \gamma) = \sqrt{\frac{\gamma}{P_0}} \frac{P_0\gamma(1 + P_0) - P_0 + 1}{2(P_0\gamma - 1)^2},$$

and finally,  $R(z)$  and  $S(z)$  are the polynomials of  $z$  determined below:

$$R(z) = 1 - 2z + \left(1 - \frac{1}{P_0\gamma}\right) z^2, \quad (25)$$

$$S(z) = \frac{1 + \sqrt{P_0\gamma} \sqrt{P_0\gamma - P_0\gamma + (P_0\gamma - 1)z}}{1 - \sqrt{P_0\gamma} \sqrt{P_0\gamma + P_0\gamma - (P_0\gamma - 1)z}}. \quad (26)$$

It is worth mentioning that relation (23) in the limit  $P_0 \rightarrow 0$  coincides with the corresponding relation of the correlated soliton model of the DL [14].

Thus, the kinetics of the luminescence in the presence of linear pumping (22), according to Eqs. (12) and (20), is given by the equation

$$I = \frac{z(z + P_0)}{\gamma + (1 - 2\gamma)z + \gamma z^2} - P_0, \quad (27)$$

where the time dependence of  $z = z(\tau)$  is determined from expression (23). The latter can be numerically inverted by the parametrizing procedure.

## V. NUMERICAL RESULTS AND COMPARISON WITH THE EXPERIMENTAL DATA

The numerical analysis of Eq. (21) shows that at the fixed value of  $\gamma$ , the value  $z$  is a monotonously increasing function of  $P_0$  and of the dose  $D = P_0\tau_{ill}$ , and reaches some saturation value at large  $P_0$  and  $D$ . These results are represented in Fig. 1. The fact of the saturation of the quantum yield of DL is connected with the luminescence channel during the process of illumination. In the absence of this channel the total number of ionized reaction centers, as it follows from Eqs. (14) and (22), would increase infinitely with time and pumping increasing.

Figure 2 shows the dependence of the quantum yield of DL on the dose at three fixed levels of pumping  $P_0 = 0.05, 0.36, 6.75$  at the value of the kinetic rate  $\gamma = 10$  together with the experimental data, shown in different markers for different intensities of the illuminating light (see figure captions). Note, this value of  $\gamma$  coincides with the value at which our model gives the best fit of the experimental data on the kinetics of the DL for alga *A.a.* [14] (more general estimate shows that this value for biological systems can

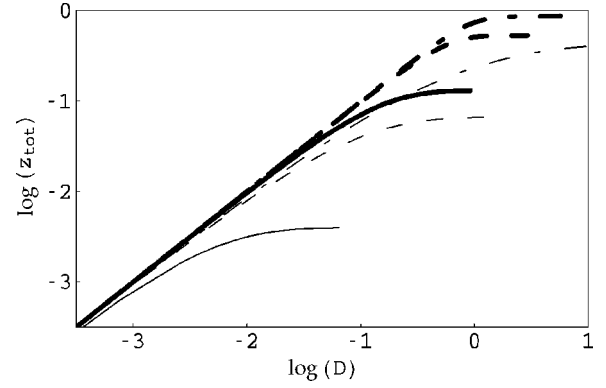


FIG. 1. The dependence of the quantum yield of DL  $z_{tot} = z(\tau_{ill})$  on dose at various intensities of illuminating light,  $P_0$ : 0.01 (solid lines); 0.2 (dashed lines), 5.0 (dot-dashed lines). Thick lines correspond to  $\gamma = 50$ , normal lines correspond to  $\gamma = 1$ .

vary in the interval  $\gamma = 1 - 100$ ).

The experiments consisted in illuminating the sample, consisting of an *A.a.* cell placed in a plastic Petri dish filled with artificial sea water, with a pulse of selected wavelength and measuring the time dependence of the number of photons emitted. In the results reported below, we used stimulating light of 565 nm and 585 nm produced by light-emitting diodes (Oshino OLUY153, OLUG153) that were fed current from a stabilized power supply (Hewlett Packard 6216A). The following incident fluxes were used:  $0.8 \times 10^{12}$ ,  $5.8 \times 10^{12}$ , and  $10.9 \times 10^{13}$  photons/(s cm<sup>2</sup>). The illumination durations varied from 1 to 1000 s [23]. The experimental setup, materials, and method are described elsewhere [10,17,23]. The comparison of the experimental data and the predictions of the model allows us to conclude that our correlated soliton model of the DL, in the presence of pumping, explains both qualitatively and quantitatively the experimental data on alga *A.a.* not only on the time trends of the DL, but also on the nonlinear dependence of the total quantum yield of DL on the intensity of the illuminating light.

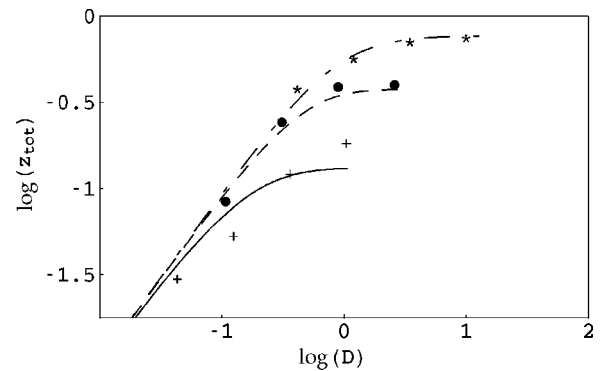


FIG. 2. Quantum yield of the DL  $z_{tot} = z(\tau_{ill})$  as a function of dose in a log-log scale. Curves correspond to the theoretical model for different values of  $P_0$ : 0.05 (solid line); 0.36 (dashed line); 6.75 (dot-dashed line) at  $\gamma = 10$ . Markers correspond to the experimental data (in arbitrary units) at various intensities of the illuminating light  $I_{ill}$ : 0.08 (+), 0.58 (full circle),  $10.87 \times 10^{13}$  photons/(cm<sup>2</sup> s) (\*).

**VI. NONLINEAR PUMPING**

When the saturation value of the quantum yield of the DL is large and close to the value of the maximum available number of states  $z_{max}$ , one cannot neglect the inhibition of the yield of the ionized reaction centers, and it is necessary to take into account the nonlinearity of the pumping, which we model by the function (17). We study this situation below.

First, we consider the first-order approximation of expression (17):

$$P(z) = P_0 - P_1 z. \tag{28}$$

Substituting this into Eq. (20), we get

$$y = \frac{(1 - P_1)z + P_0}{a[\gamma z^2 + (1 - 2\gamma)z + \gamma]}, \tag{29}$$

which, together with Eq. (14), gives us the following equation:

$$\frac{dz}{d\tau} = \frac{\mathcal{P}_3(z)}{\mathcal{P}_2(z)}, \tag{30}$$

where  $\mathcal{P}_i(z)$  are the polynomial functions of  $z$  of the  $i$ th order:

$$\mathcal{P}_2(z) = z^2 - 2\left(1 - \frac{1}{2\gamma}\right)z + 1, \tag{31}$$

$$\mathcal{P}_3(z) = -P_1 z^3 + \left(P_0 + 2P_1 - \frac{1}{\gamma}\right)z^2 - (2P_0 + P_1)z + P_0. \tag{32}$$

The numerical analysis of the polynomial  $\mathcal{P}_3(z)$  shows that it has one zero, which corresponds to the maximum of  $z$  attained at  $\tau \rightarrow \infty$ , the saturation of the DL yield at a large dose. In general, the behavior  $z(\tau)$  is very close to the one for the constant pumping, although it gives somewhat less absolute values than before (these results are not represented here). This is, obviously, related to the inhibition of the saturation due to the second term in Eq. (28).

Next, consider the yield of the reaction centers with the accuracy of the second order:  $P(z) = P_0 - P_1 z + P_2 z^2$ . Substituting Eq. (17) into Eq. (20), we get

$$y = \frac{P_2 z^2 + (1 - P_1)z + P_0}{a[\gamma z^2 + (1 - 2\gamma)z + \gamma]}, \tag{33}$$

which, together with Eq. (14), gives us the following equation:

$$\frac{dz}{d\tau} = \frac{\mathcal{P}_4(z)}{\mathcal{P}_2(z)}, \tag{34}$$

where  $\mathcal{P}_2(z)$  is given by expression (31), and

$$\begin{aligned} \mathcal{P}_4(z) = & P_2 z^4 - (P_1 + 2P_2)z^3 + \left(P_0 + 2P_1 + P_2 - \frac{1}{\gamma}\right)z^2 \\ & - (2P_0 + P_1)z + P_0. \end{aligned} \tag{35}$$

The numerical analysis shows that the polynomial  $\mathcal{P}_4$  at large values of pumping  $P_0 \gg 1$  has two zeros at  $z = z_1$  and  $z = z_2$ , the numerical values of which, obviously, depend on the value of all parameters  $P_i (i=0,1,2)$  and  $\gamma$ , incoming into Eq. (35). As a result, Eq. (34) cannot be integrated straightforwardly by separating variables  $z$  and  $\tau$ . The two zeros correspond to the two extrema, which are the two maxima, of  $z$  as a function of  $\tau$ . The presence of the second zero of the polynomial changes qualitatively the dependence of the quantum yield of the DL on the dose of irradiation. This change occurs due to the photoinhibition. One of the two zeros corresponds to the finite value of  $\tau = \tau_0$ , at which  $z$  reaches the absolute maximum  $z_0$ . The other one corresponds, as above, to  $\tau \rightarrow \infty$  where  $z$  reaches the saturation  $z_{sat}$  such that  $z_{sat} < z_0$ . Obviously, both are less than the maximum available number of reaction centers in the system (this, in the units used above, is set to be equal to 1). Therefore, we conclude that at high level of pumping (high intensity of the illuminating light), the total quantum yield first increases with the dose increasing, reaches the maximum value, and then decreases till the saturation level. Thus, there exists an optimum dose of the illumination, at which the total quantum yield of the DL has the maximum value. These results qualitatively agree with the experimental data on *A.a.* for very high intensity of the illuminating light.

**VII. ANALYSIS OF THE STABILITY**

The model, developed above, describes the process of the DL by a system of three nonlinear equations (14)–(16), which include pumping. The results, obtained in the above sections, both for the linear and nonlinear pumping, have been obtained in the steady state approximation. In the meantime it is known that not every solution of nonlinear systems is stable (see, e.g., Refs. [24,25]), especially in the presence of the external source, like the case considered above for the system with pumping. At certain conditions the auto-oscillations can arise in such a system, and even transition to chaos can occur. In the general case this problem for nonlinear systems is quite complex, and usually it is studied in the linear limit for the perturbations/corrections [26–28]. Just for this reason we study in this section the properties of the above derived solutions with respect to their stability.

With account of the small corrections, which can arise with time due to the instability, when the system deviates from the steady state solution, we assume

$$z(\tau) = z_{st}(\tau) + u(\tau), \quad y(\tau) = y_{st}(\tau) + v(\tau), \tag{36}$$

$$x(\tau) = x_{st}(\tau) + w(\tau),$$

where  $z_{st}, y_{st}, x_{st}$  are steady state solutions obtained in the previous sections, and  $u, v, w$  are the corresponding correc

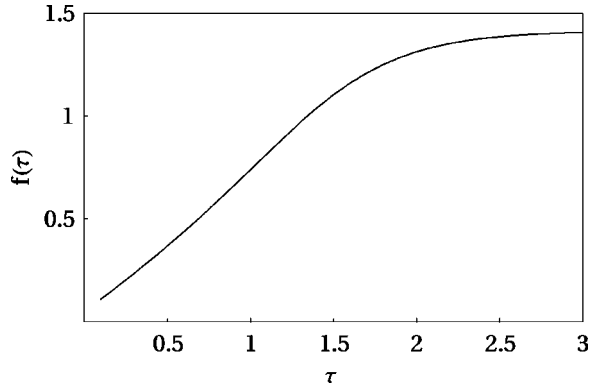


FIG. 3. Time dependence of function  $f(\tau)$  in Eq. (37) at  $P_0 = 0.5$ ,  $\gamma = 10$ .

tions. The self-trapped soliton states possess the property of the inner stability as essentially nonlinear objects, their lifetime is much higher than the lifetime of the delocalized electron states. Moreover, in our model it is assumed that in the result of the illumination electrons, released during the charge separation of the reaction centers, transit first into the conduction band of a macromolecule straightforward or via fast transition on a light molecule on one end of the chain (the so-called prothetic group). Therefore, in the same order of the approximation, we can put  $w=0$ ; and then, due to relation (5), we get  $v=u$ .

First, we study the case of linear pumping. Substituting ansatz (36) into Eqs. (14)–(16) and linearizing them with respect to  $u$ , we get the following equation:

$$\frac{du}{d\tau} = -f(\tau)u. \quad (37)$$

Here

$$f(\tau) = a \left\{ z_{st} + \frac{z_{st} + P_0}{a[\gamma z_{st}^2 - (2\gamma - 1)z_{st} + \gamma]} \right\}, \quad (38)$$

and  $a$  is determined in Eq. (18). The numerical calculations of Eq. (38) with the data for  $z_{st}$  taken from relation (23), for the parameter values that were used above for the fit of the experimental data, are shown in Fig 3. From this we see that at small time the function  $f(\tau)$  linearly increases with time, being positive, so that it can be approximated as  $f(\tau) \approx \Gamma_1 \tau$ . At large time,  $\tau \gg \tau_{sat}$  (where  $\tau_{sat}$  is the characteristic time when the steady state solution reaches the saturation), the function  $f(\tau)$  reaches some maximum value and remains constant,  $f(\tau) \approx \Gamma_2 = \text{const}$ . Therefore, the solution of Eq. (37) can be represented as

$$\begin{aligned} u(\tau) &= u_{0,1} e^{-\Gamma_1 \tau^2/2}, & 0 < \tau < \tau_{sat}, \\ u(\tau) &= u_{0,2} e^{-\Gamma_2 \tau}, & \tau \gg \tau_{sat}, \end{aligned} \quad (39)$$

where  $u_{0,1}, u_{0,2}$  are the constants of integration.

Similar situation takes place for that values of  $\gamma$  and  $P_0$  close to that used above, and we can conclude that the linear corrections to the steady state solution for the linear pumping are exponentially small in the whole interval of time.

In the case of nonlinear pumping, the equation for the corrections in the linear limit reads as

$$\frac{du}{d\tau} = -F(\tau)u - P_1 z_{st}, \quad (40)$$

where

$$F(\tau) = a \left( z_{st} + y_{st} + \frac{P_1}{a} \right), \quad (41)$$

and, similar to the above, the solution of Eq. (40) reads as

$$u(\tau) = u_0 e^{-\Gamma \tau + \frac{P_0 z_{st}(2 - z_{st})}{\Gamma}}, \quad \tau \gg \tau_{sat}, \quad (42)$$

with  $u_0$  being the constant of integration and  $\Gamma \approx a z_{st}$ . Therefore, in the case of the nonlinear pumping the corrections tend with time to some small constant value. Mind that, these results are valid only for the parameter values close to those used for the fit of the experimental data. In the general case this problem requires much more elaborate study and is in progress.

## VIII. CONCLUSION

The above analysis shows that the nonlinear coherent model of the DL in the presence of pumping provides qualitative and quantitative explanation of the main characteristic features of the DL from *A.a.* These include the kinetics of time decay, the nonlinear dependence on the intensity of the stimulating light, and the same time trend for all the emission spectral components.

The account of pumping in the model due to the process of irradiation, and the nonlinear character of the quantum yield of the photosystem, results in the nonlinear dependence of the quantum yield of the DL on the dose of irradiation. At relatively low intensities of the illuminating light, the quantum yield of the DL monotonously increases with dose, and gradually reaches the saturation. At high intensities of the illuminating light, the photoinhibition of the photosystem comes into play. As a result, the quantum yield of the DL attains the maximum value at the optimum dose, but with further increase of the dose the quantum yield decreases from this optimum value till some limiting (asymptotical) value. The theoretical results fit well the experimental data on alga *A.a.* in the wide interval of dose and intensity of the illuminating light.

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