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Nonlinear response of biophoton emission to external perturbations

Q. Gu[a, b] and F.-A. Popp[a]

[a]*International Institute of Biophysics, Technology Center, Opelstr. 10, D-6750 Kaiserslautern 25 (Federal Republic of Germany) and* [b]*Department of Physics, Northwest University, Xi'an 710060 (People's Republic of China)*

Abstract. By considering an exciplex system consisting of collective molecules in interaction with both the 'pumping' fields and the biophoton fields, the two-level exciplex model and the three-level exciplex model are presented. They are useful for the investigation of the quasi-stationary behaviour of biophoton emission, and biophoton emission as a dynamic process in the presence of external perturbations. Our theoretical results predict a series of nonlinear effects, such as chaos, fractal behaviour, and non-equilibrium phase transition. These effects characterize the coherence nature of living systems. In our approaches, there are two important quantities f and x, which can be used to mark the working points of the two-level and three-level exciplex systems. All the influences of external perturbations on the exciplex systems, e.g. change of temperature, the addition of agents, exposure to light, etc., can be interpreted as shifts of the working points of the systems, leading to a diversity of nonlinear response of biophoton emission. In addition, the agreements of the theoretical results and the corresponding experimental observations on biophoton emission from biological systems in the presence of external perturbations are demonstrated.
Key words. Exciplex formation; two-level exciplex model; three-level exciplex model; chemical potential; pumping field; collective molecules; chaos; fractal behaviour; non-equilibrium phase transition; working point.

Introduction

'Biophoton emission' is now a topical field in contemporary science[33, 65, 66, 102]. It concerns weak light emission from biological systems, with an intensity of the order of a few up to some hundred photons per second and per square centimeter of surface area.

The origin of biophoton research can be traced back to A. G. Gurwitsch[27, 28, 48]. He performed various experiments on 'mitogenetic radiation' with the aid of biological detectors. Gurwitsch claimed that the most fundamental biological function, namely cell division, is triggered by a very weak photocurrent originating from the cells themselves. Since biological detector systems found little support in the scientific world, and because of the unavailability of sufficiently sensitive technical equipment at that time, no generally accepted conclusion on mitogenetic radiation was reached for quite a long period.

In 1955 Colli et al.[10] succeeded in proving the existence of photon emission from cereals by using a photomultiplier tube. The photons were regarded as visible radiation between 390 and 650 nm with intensities of some hundred photons/(s · cm^2).

In the 1960s most of the research work on biophoton emission was performed by Russian scientists[40, 64, 107, 114], who measured the biophoton emission from about 90 kinds of biological samples, including yeast, frog nerve and mouse liver, again using photomultipliers.

In the last fifteen years, essential progress in this field has been accomplished, involving the following topics: the source of the biophotons[2, 3, 42, 60, 67–69, 86], their correlations with biological, biophysical and biochemical processes[34, 39, 65, 66, 84, 88, 93, 94, 101, 113], the temperature dependence[70, 71, 96, 97], the spectral distribu-

tion[30,85,89,92,95,98], the optical transparency[4,49,72], the influence of external factors[13,52,73,74,104], the photocount statistics[37,70,74,75], the relaxation dynamics after excitation[5–8,21,29,41,43,50,55,56,61,70,74,76,77,91,105], and the coherence of swarming[16,17,58]. Currently, biophoton research has developed to a stage of modern analysis, including the detection technique[30–32,54,70,88], the search for mechanisms[9,44,57,78–80], theoretical descriptions[21,43,45–47,70,74,76,77], and possible applications[14,19,38,87,90,100,106,110,111]. Recently, the current status and prospects of biophoton emission have been extensively reported in a number of review articles[65,81,82,99] and books[33,66,102].

A variety of experimental observations give evidence that biophotons originate from a delocalized coherent electromagnetic field within living matter[18]. As a generator of this field, living matter displays an energy-level distribution characterized by the f_v = constant-rule, which means that in the ideal case (enough 'pumping' energy is always available) all the relevant excited states of living matter are occupied with about the same probability, independent of the excitation energy[70,74,75,82]. The f_v = constant-rule governs a non-equilibrium phase transition between a 'chaotic' and an 'ordered' regime[21,75]. Around the critical point of this phase transition coherent radiation of a multi-mode biophoton field induced by collective bioradiators can become stabilized. The coherence of biophoton emission can be understood in terms of the emission of the phase-locked and mode-locked biophotons of living systems in their quasi-stationary-state operations[21,29]. A sufficient condition for coherence is the hyperbolic relaxation after ergodic excitation[5–8,21,29,41,50,55,56,61,77,91,105] which follows from a coherent nonlinear coupling among the collection of molecules within living matter[1,24,25,109]. The coherence of biophotons from a swarm of Daphnia can be analysed in terms of an interference pattern of swarming[16,17,58], which can be described well by a destructive-interference model[83] as a consequence of Dicke's theory[11,12]. The low intensity of biophoton emission can be explained in terms of the fairly high degree of coherence of non-classical light[15,20,53,63,103] with very high signal/noise-ratio[22,23,112]. The essential source of biophoton emission may be displayed by the exciplex system[26,51,108], in particular by the exciplexes of DNA[2,3,42,60,67–69,86].

More and more, the experimental results indicate that biophoton emission displays a non-linear response to external perturbations, where a typical example that has already been considered is the non-linear temperature response of biological systems as demonstrated by low level luminescence of cucumber seedlings[71].

In the present paper we report the recent results of theoretical research on the non-linear response of biophoton emission to external perturbations, including chaos and phase transition in biophoton emission. Specifically, we study the fractal behaviour in biophoton emission starting with the two-level exciplex model and then consider the non-equilibrium phase transition with the aid of the three-level exciplex model. In addition, we show the agreement of the theoretical results and the corresponding experimental observations on biophoton emission from biological systems in the presence of external perturbations.

Exciplex models

Let us consider exciplex formation of a biomolecular system, which is a very common process in living matter[26,51,108]. Figure 1 (a) gives a representation of its potential energy profiles as a function of the separation of monomers. The potential curves $\sum^*$ and $\sum$ represent the bound excited state and the repulsive ground state, respectively. The radiative behaviour of such a system may

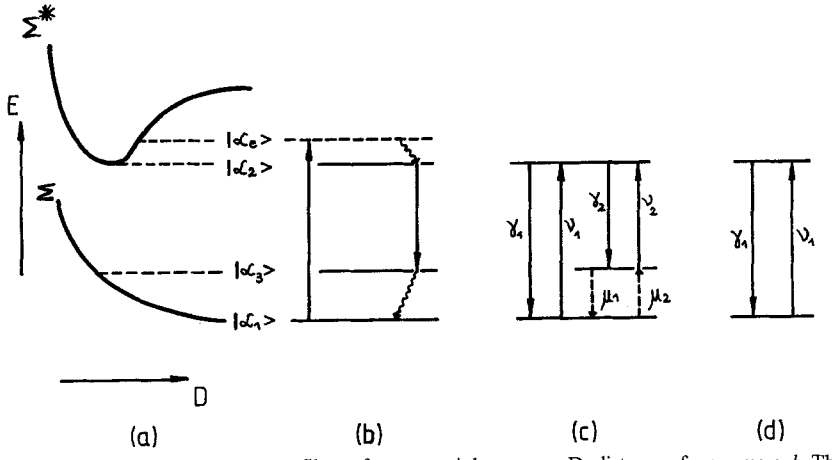

Figure 1. *a* Representation of the potential energy profiles of a biomolecular system with exciplex formation, where $\sum^*$ and $\sum$ represent the bound excited state and the repulsive ground state, respectively. E: exciplex energy, D: distance of monomers. *b* The diagram of the exciplex model. *c* Simplified three-level system. *d* Further simplification to a two-level system.

be simplified to that of a four-level laser (the exciplex model[45]) as shown in figure 1 (b). The exciplex model describes the biophoton emission process as follows. The molecules from the lowest level $|\alpha_1\rangle$ are pumped into state $|\alpha_e\rangle$, which is an arbitrary excited state confined within the potential curve Σ^*. The pumping energy supply is maintained by the metabolism (glycolysis, ATP, etc.), and even direct pumping with sunlight provides an inexhaustible source[81]. One may imagine that the energy supply originates from a 'pumping field'. From state $|\alpha_e\rangle$ the molecules then decay very rapidly through a non-radiative transition to state $|\alpha_2\rangle$ which is a metastable state having a long lifetime. Thus the pump effectively transfers molecules from $|\alpha_1\rangle$ to $|\alpha_2\rangle$ through $|\alpha_e\rangle$. From state $|\alpha_2\rangle$ the molecules decay under photon emission to state $|\alpha_3\rangle$. One can see from the exciplex model that the main biophoton emission follows from the transition from $|\alpha_2\rangle$ to $|\alpha_3\rangle$. However, in general, the radiative transition from $|\alpha_2\rangle$ to $|\alpha_1\rangle$ is also possible. After the molecules arrive in state $|\alpha_3\rangle$, most of them may relax down through a non-radiative transition into state $|\alpha_1\rangle$, ready to be pumped again to $|\alpha_e\rangle$, while the rest may be pumped immediately by another 'pumping field' to $|\alpha_e\rangle$. Note that $|\alpha_3\rangle$ should be considered as an arbitrary state on the potential curve Σ, which can take all possible values of vibrational energy of the lattice system of biomolecules[81].

The above-described understanding of biophoton emission seems to indicate a four-level system. However, the four-level system can usually be simplified to a three-level system. In fact, the transition $|\alpha_1\rangle \rightarrow |\alpha_e\rangle \rightarrow |\alpha_2\rangle$ under consideration can be effectively simplified to $|\alpha_1\rangle \rightarrow |\alpha_2\rangle$. So, in our approach, only three levels ($|\alpha_1\rangle$, $|\alpha_2\rangle$ and $|\alpha_3\rangle$) need to be taken into account, as shown in figure 1 (c), which may be regarded as a three-level exciplex model. Provided that the $|\alpha_3\rangle$ state is very close to the $|\alpha_1\rangle$ state, one can arrive at the two-level exciplex model, as shown in figure 1 (d). This model ignores some details, but it can be solved exactly and has significance in research on many problems[1].

Chaos and fractal behaviour of biophoton emission

In this section we study the chaos and fractal behaviour of biophoton emission using the two-level exciplex model as shown in figure 1 (d). If a system consisting of N_0 units of a biopolymer, for instance base-pairs in DNA, rests in a stationary state, this always leads to N_1 unexcited monomers and N_2 exciplexes such that

$$N_1 + 2N_2 = N_0 . \tag{1}$$

For simplicity we assume that the unexcited monomers are subject to the mean thermal energy kT, whereas the excited molecules have to be assigned to a chemical potential μ. In a stationary state a general energy balance equation holds

$$N_1 kT + N_2(\mu + C_1) = C_0 N_0 , \tag{2}$$

where C_1 is the energy dissipated over the system per exciplex formation. For $\mu = 0$ we have $C_0 = \frac{C_1}{2} = kT$, representing thermal equilibrium. The interaction between radiation and matter is described by Einstein's famous balance equation in the form

$$\dot{\rho} = h\nu[AN_2 + (N_2 - N_1)\rho B], \tag{3}$$

where ρ is the radiated energy density and A and B are the Einstein coefficients of spontaneous and induced emission, respectively. Thus we have

$$f = \frac{\rho B}{A} = \left[\exp\left(\frac{h\nu - \mu}{kT}\right) - 1\right]^{-1} . \tag{4}$$

By use of Eq. (4) and the abbreviations

$$\rho = \frac{A}{B}\left[\exp\left(\frac{h\nu - \mu}{kT}\right) - 1\right]^{-1} , \tag{5}$$

$$y \equiv \frac{N_1}{N_2} = \frac{C_1 + \mu - 2C_0}{C_0 - kT} , \tag{6}$$

we obtain the iteration equations:

$$f_{i+1} = f_i + \frac{\beta}{y_i + 2}[1 + f_i(1 - y_i)], \tag{7}$$

$$y_{i+1} = y_i + \frac{y_i + 2}{\frac{\mu + C_1}{kT} - 2}\,\frac{\frac{f_{i+1}}{f_i} - 1}{f_i + 1} , \tag{8}$$

where $\beta = AN_0 h\nu \Delta t$ and $i = 0, 1, 2, \ldots$ denotes a consecutive number of points in the time-evolution of the system.

For any set of f_0 and y_0, the dynamic behaviour of photon emission is determined, provided that $(\mu + C_1)$ is known.

Since Δt can be chosen freely, we can set $\beta = 1$ without losing general validity. Let us assume $\mu + C_1$ to be independent of time, then the photon intensity $\dot{\rho}_i \propto \frac{f_{i+1} - f_i}{\beta}$, displays a fractal behaviour[62], since the quantity $\dot{\rho}$ does not depend explicitly on Δt. Figures 2 and 3 display an example where the fractal behaviour becomes obvious. Every emission pattern can be truthfully described by using Eqs (7) and (8), as shown in figures 2 and 3.

An attractor of the system is $y = 1 + \frac{1}{f}$, since in that case $f_{i+1} = f_i$ (Eq. (7)) and consequently $y_{i+1} = y_i$ (Eq. (8)). As the system is an open one, it is subject to maximum entropy under the constraint of the stationarity of both $\dot{\rho} = 0$ and exciplex number $N_2 = \text{constant}$. So for $\left(\frac{\delta N_2}{\delta \rho}\right)_{\rho_0} = 0$ and taking into account

$$\frac{N_1}{N_2} = \frac{A}{\rho B} + 1 \tag{9}$$

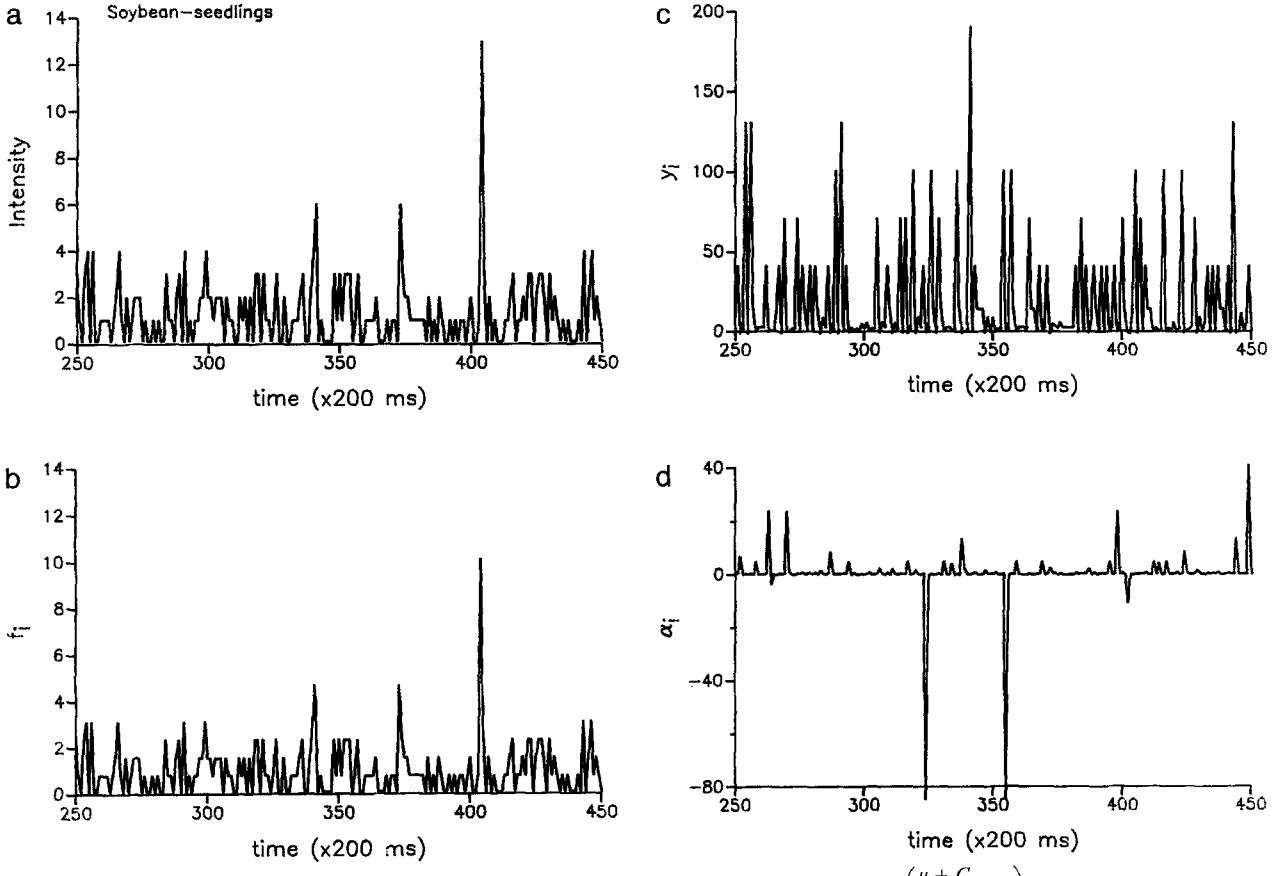

Figure 2. *a* Photocount rate of soybean seedlings during the course of time from 250 × 200 ms (= 50 s) to 90 s in a preset time interval Δt of 200 ms. *b* Calculated f_i-values from fig. 2a, where f_i has been taken as a measure of the count rate at time t_i. *c* Calculated y_i-values from fig. 2b, by use of formula (7). *d* $\alpha_i = \left(\frac{\mu + C_1}{kT} - 2\right)$-values according to formula (8) by use of the f_i- and y_i-values of figs 2b and 2c. It turns out that α_i stabilizes around the value 0, indicating highest sensitivity of the system around $f = 1$ and $y = 2$.

from Eq. (3), and then substituting μ from Eq. (4) into Eq. (2), we get

$$\frac{N_2}{N_0}\frac{kT}{C_0} = \frac{\frac{\rho B}{A}}{1 + \frac{\rho B}{A}\left(1 + \frac{h\nu}{kT} + \frac{C_1}{kT} + \ln\frac{\rho B}{A}\right)}. \tag{10}$$

Differentiating with respect to ρ $\left(\text{or } \frac{\rho B}{A}\right)$ yields

$$\frac{\partial\left(\frac{N_2}{N_0}\frac{kT}{C_0}\right)}{\partial\left(\frac{\rho B}{A}\right)} = \frac{1 - \frac{\rho B}{A}}{\left[1 + \frac{\rho B}{A}\left(1 + \frac{h\nu}{kT} + \frac{C_1}{kT} + \ln\frac{\rho B}{A}\right)\right]^2} \tag{11}$$

which takes the value zero only for

$$f = \frac{\rho B}{A} = 1, \tag{12}$$

or

$$y = 2. \tag{13}$$

This indicates that the system stabilizes, as soon as enough energy is available, around a state with $f = 1$ (or $y = 2$), where one half of the molecular units are unexcited and the other half are in the excited exciplex state.

From Eq. (8) we get the differential equation

$$\left(\frac{\mu + C_1}{kT} - 2\right)\frac{dy}{y + 2} = \frac{df}{f + f^2}, \tag{14}$$

with a solution in the form

$$\frac{\mu + C_1}{kT} - 2 = \frac{\ln\left(\frac{2f}{1 + f}\right)}{\ln(2 + y)}. \tag{15}$$

For the case of a stationary state, where $y = 1 + \frac{1}{f}$, the term $\left(\frac{\mu + C_1}{kT} - 2\right)$ follows the f-dependence as hown in figure 4.

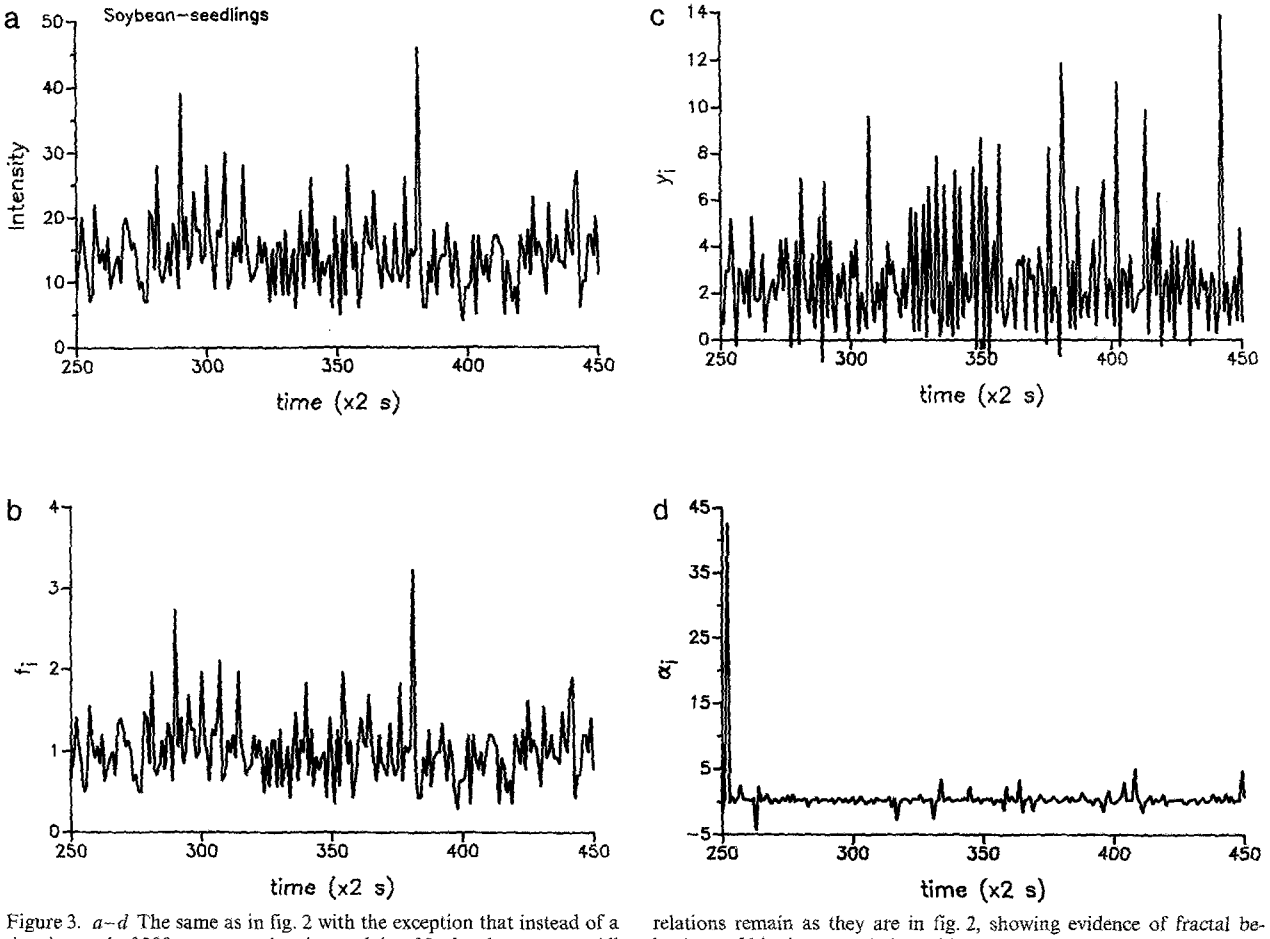

Figure 3. *a–d* The same as in fig. 2 with the exception that instead of a time interval of 200 ms now a time interval Δt of 2 s has been preset. All relations remain as they are in fig. 2, showing evidence of fractal behaviour of biophoton emission with respect to Δt.

Around $\frac{\mu + C_1}{kT} - 2 = 0$, which is obtained for $f = 1$, the system becomes subject to a negative feed-back coupling, since

$$\frac{df}{dy} > 0 \quad \text{for} \quad f > 1, \tag{16}$$

and

$$\frac{df}{dy} < 0 \quad \text{for} \quad f < 1. \tag{17}$$

This means that as soon as f increases above the threshold ($f > 1$), y increases too, indicating a process of stimulated emission of radiation. The system gets deexcited by its own photon emission. On the other hand, below threshold ($f < 1$) a further decrease of f results again in an increase of y, opening the possibility of higher absorbance of the system. If enough energy is available, it will induce a decrease of y, resulting at the same time in an increase of f as long as $f < 1$.

Let us apply this model to a concrete case, where *Acetabularia* have been poisoned with different concentrations

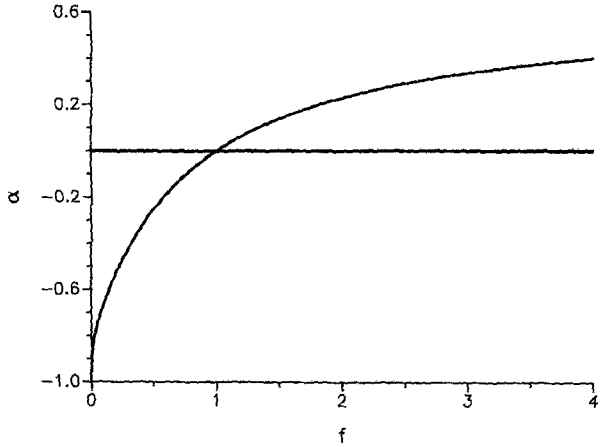

Figure 4. The chemical potential μ (expressed here in terms of $\alpha = \frac{\mu + C_1}{kT} - 2$) depends sensitively on the f-value. At $f = 1$, α changes its sign, thus providing a negative feed-back coupling (see text and formulae (14) and (15)).

of atrazine. It turns out that the photon re-emission of *Acetabularia* ('delayed luminescence') after light illumination reacts sensitively to exposure to a poison, e.g. it

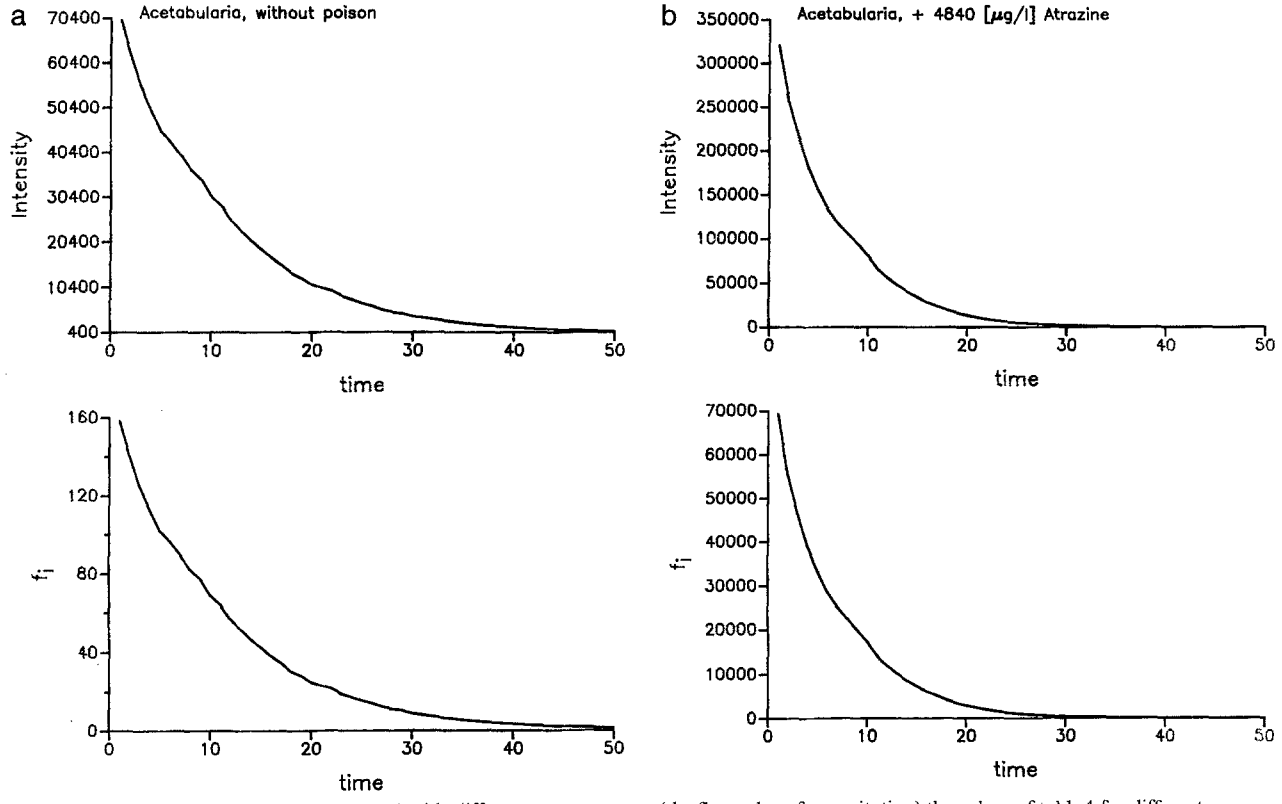

Figure 5. *a, b Acetabularia* which are treated with different concentrations of a poison display different excitation after exposure to white-light illumination (detailed description of the experiments, see reference 56). From the measured curves the f_i-values can be calculated, where for f_1 (the first value after excitation) the values of table 1 for different concentrations of the poison in two series of measurements have been obtained [56].

Table 1

Series of measurements	f_1-experimental	Concentration [µg/l]
1	144.62	0
	4615.2	200
	44774.96	1000
	99846.01	4000
2	158.45	0
	148.08	0.7
	227.31	7.9
	950.28	60
	5586.77	194
	69328.17	4840

increases considerably and immediately after the addition of atrazine (see table 1 and fig. 5).

The experimental data I_i (photon intensity at time t_i, $i = 1, 2, \ldots$) can be used for evaluation of f_i, where $f_i = \frac{I_i}{I_\infty}$. This provides that the relaxation of the system arrives finally at $f_\infty = 1$. From f_i we obtain y_i ($i = 1, 2, \ldots$), where Eq. (7) is used. Then the values $\frac{(\mu + C_1)}{kT}$ are calculated, where the set f_i, y_i ($i = 1, 2, \ldots$), which is obtained from Eq. (8), is inserted into Eq. (8). These values can be examined, in order to get some criteria for the validity of the model, e.g. the relation $\frac{(\mu + C_1)}{kT} - 2 \approx 0$. On the other hand, the transformation of the experimental values into the parameters of the chaos-model serves as a basis for modelling the effect of the agent. This procedure does not lead to an unequivocal description of the efficacy of the poison. However, one can find a way of understanding it in very simple terms which can be more and more improved after experimental re-examination.

As is already known, delayed luminescence follows a hyperbolic relaxation behaviour (see, for instance, ref. 66). This means that

$$f_i \propto (i + i_0)^{-\beta}, \tag{18}$$

where β is a constant.

From Eq. (18) we get, after a straightforward calculation,

$$\beta = \frac{\ln\left(\frac{f_i + 1}{f_i}\right)}{\ln\left(\frac{i + i_0}{i + 1 + i_0}\right)}. \tag{19}$$

By the use of the experimental data, approximate values of β and i_0 are obtained. Table 2 shows these values. In figure 6 the dependence of β on the concentration c of the poison is displayed.

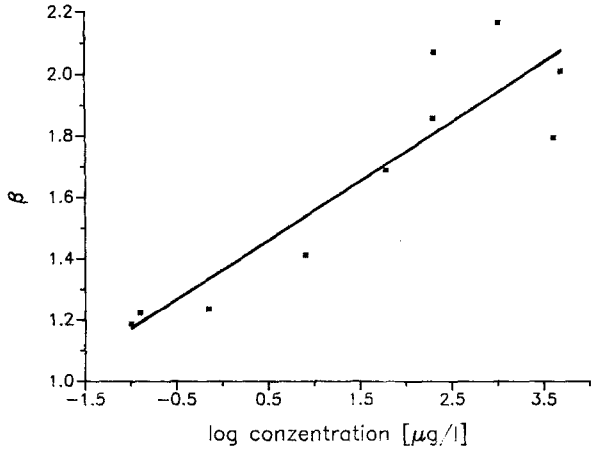

Figure 6. The values β according to formula (19) display a linear increase with increasing concentration c of the poison. This indicates a continuous loss of the degree of coherence in the system with increasing c.

Table 2

Series of measurements	$\beta \pm \Delta\beta$	i_0	Concentration [μg/l]
1	1.187 ± 0.304	11	0
	2.0699 ± 0.543	11	200
	2.164 ± 0.357	11	1000
	1.793 ± 0.496	11	4000
2	1.223 ± 0.367	12	0
	1.235 ± 0.272	11	0.7
	1.412 ± 0.182	11	7.9
	1.687 ± 0.395	11	60
	1.856 ± 0.233	11	194
	2.008 ± 0.261	11	4840

Table 3

Series of measurements	f_1-theoretical	σ	Concentration [μg/l]
1	143.1	59.2	0
	4617.8	110.1	200
	44696.7	143.6	1000
	99563.7	155.4	4000
2	155.04	60.4	0
	148.9	59.8	0.7
	227.3	66	7.9
	954.1	86.9	60
	5804.6	113	194
	69137.1	150	4840

The results show that the poisoning immediately induces an increase of β, indicating a considerable decrease of the degree of coherence within the system.

However, there must be a connection to the strong increase of the f-values by the addition of atrazine. Consequently, it may be worthwhile to simulate the increase of photon re-emission during the exposure to light illumination, and not only after switching off the external light source. A rather reasonable method has been used for describing this effect in terms of Eqs (7 and 8). Of course, we can alsways write

$$y_i = 1 + \frac{1-\delta_i}{f_i}, \tag{20}$$

where δ_i represents a measure of the deviation of y_i from its attractor $y_i = 1 + \frac{1}{f_i}$. Since at the beginning and at the end of light exposure we can simply assume that y_i takes the value of its attractor, the Gaussian-like deviation

$$\delta_i = f_i \exp\left(-\frac{(i+i_0)^2}{2\sigma^2}\right) \tag{21}$$

may account for the occupation of excited states by absorption of light. The lower the value of σ, the faster and more cooperative is this effect. One may expect that there will be a positive correlation between $\beta(c)$ and $\sigma(c)$. This is actually the case. Table 3 shows the values of σ which have to be chosen in order to get the final values $f(c)$ for different concentrations. In figure 7 some examples have been displayed. Immediately after poisoning, the biological system needs a longer time to store the absorbed light, while after switching off the external source, the coherent rescattering of light within the system is significantly lowered compared to that in the untreated system. In both cases the distribution of light and its interaction with the exciplex system become more and more diffused.

Non-equilibrium phase transition in biophoton emission

In this section we investigate the non-equilibrium phase transition in biophoton emission using the quantum theory of the interaction of radiation with matter. Let us consider the three-level exciplex system as shown in figure 1 (c) and assume that the system consists of N molecules, which occupy a region which is small compared to the wavelength of the relevant radiation (the Dicke model [12]). The system interacts with two pumping fields of the resonance frequences Ω_1 and Ω_2. The Hamiltonian describing the interaction can be written in the rotating-wave approximation in the form ($\hbar = 1$):

$$H = \sum_{i=1}^{3} \omega_i A_{ii} + (G_1 A_{12} e^{-i\Omega_1 t} + G_2 A_{32} e^{-i\Omega_2 t} + HC), \tag{22}$$

where ω_i and $A_{ij} = \sum_{k=1}^{N} |\alpha_i\rangle_k {}_k\langle\alpha_j|$ $(i,j = 1, 2, 3)$ represent the energy levels of the system and its collective operators, respectively, and G_1 and G_2 are the Rabi frequencies of the two pumping fields. The master equation of the density operator ρ of the system is then expressed by

$$\frac{\partial\rho}{\partial t} = -i[H,\rho] + \left.\frac{\partial\rho}{\partial t}\right|_{dt} + \left.\frac{\partial\rho}{\partial t}\right|_{a} + \left.\frac{\partial\rho}{\partial t}\right|_{n} + \left.\frac{\partial\rho}{\partial t}\right|_{b}, \tag{23}$$

where the last four terms describe, respectively, 1) the molecular decays and the influences of thermal fields, 2) the absorptions, 3) the nonradiative transitions between $|\alpha_2\rangle$ and $|\alpha_3\rangle$, and 4) the influence of the heat bath,

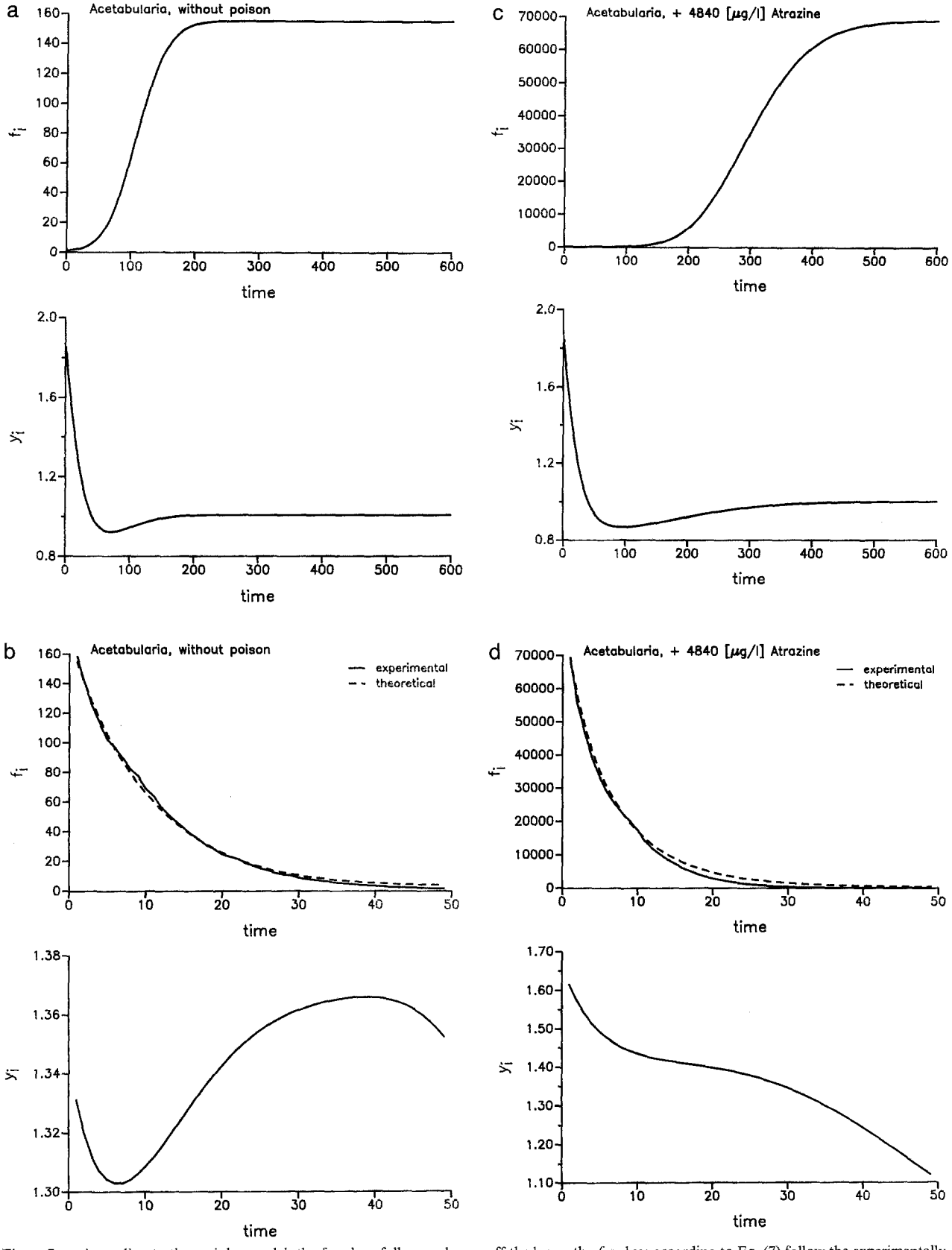

Figure 7. *a* According to the exciplex model, the δ_i-values follow under light-exposure formula (21), where i_0 (= 25) is fixed, while σ depends on the concentration c of the poison according to table 3. Starting with $f_1 = 1$, the δ_i-values determine y_i according to formula (20), thus yielding f_{i+1} according to Eq. (7). Again δ_{i+1} can be deduced from equation (21), and so on. For $c = 0$, one obtains f_i and y_i of fig. 7a. *b* After switching off the lamp, the f_i-values according to Eq. (7) follow the experimentally observed data. The y_i-values, representing the ratio of unexcited to excited exciplex-states, are drawn in the lower part. *c, d* The same as in figs 7a, b with the exception that the *Acetabularia* have been poisoned with 4840 [µg/l] atrazine.

which can be given by

$$\left.\frac{\partial\rho}{\partial t}\right|_b = -\gamma_1|m_1|(2A_{21}\rho A_{21} - A_{21}^2\rho - \rho A_{21}^2)\,e^{i\Phi_1} - \gamma_1|m_1|(2A_{12}\rho A_{12} - A_{12}^2\rho - \rho A_{12}^2)\,e^{-i\Phi_1} - \gamma_2|m_2|(2A_{23}\rho A_{23} - A_{23}^2\rho - \rho A_{23}^2)\,e^{i\Phi_2} - \gamma_2|m_2|(2A_{32}\rho A_{32} - A_{32}^2\rho - \rho A_{32}^2)\,e^{-i\Phi_2}, \quad (24)$$

where γ_1 and γ_2 are the coefficients of single-molecular spontaneous emission from $|\alpha_2\rangle$ to $|\alpha_1\rangle$ and from $|\alpha_2\rangle$ to $|\alpha_3\rangle$, and $m_i = |m_i|e^{i\Phi_i}$ are the bath parameters[35, 112] such that $|m_i|^2 \leq \bar{n}_i(\bar{n}_i + 1)$ with $\bar{n}_i$ as the mean photon numbers of the thermal fields corresponding to frequencies Ω_i. The master equation (23) in the presence of the heat bath is obtained for the first time, to our knowledge. When we put $m_i = 0$, the master equation reduces to the form obtained previously[21].

Under the secular approximation, one can get the steady-state solutions of Eq. (23) in the form

$$\rho_s = \frac{1}{C}\sum_{m=0}^{N}\sum_{n=-m,\,-m+2,\ldots}^{m} x^{-m}|m,n\rangle\langle n,m|, \quad (25)$$

where C is the normalization constant, $|m,n\rangle$ are the collective states, and x is a parameter related to all factors of the three-level exciplex system interacting with both the pumping fields and the biophoton fields.

By using the steady-state solutions (25) the molecular populations in the excited state $|\alpha_2\rangle$ and their fluctuations can be obtained in the form

$$\langle A_{22}\rangle_S = \tfrac{1}{2}\langle E\rangle_S, \quad (26)$$

and

$$\sigma_2^2 = \langle A_{22}^2\rangle_S - \langle A_{22}\rangle_S^2 = \tfrac{1}{3}\langle E^2\rangle_S - \tfrac{1}{4}\langle E\rangle_S^2 + \tfrac{1}{6}\langle E\rangle_S, \quad (27)$$

where

$$\langle E^k\rangle_S = \frac{1}{C}\sum_{m=0}^{N} m^k(m+1)x^{-m}\,(k = 1, 2, 3, \ldots). \quad (28)$$

In particular, at $x = 1$, one has

$$\langle A_{11}\rangle = \langle A_{22}\rangle = \langle A_{33}\rangle = \frac{N}{3} \quad \text{(for any } N\text{)}, \quad (29)$$

and

$$\lim_{N\to\infty}\frac{\sigma^2}{N^2} = \frac{1}{18}. \quad (30)$$

Both the fractional populations $\langle A_{22}\rangle_S/N$ and the relative fluctuations σ_2^2/N^2 as functions of the parameter x are shown in figure 8. For the analytical and numerical results obtained here one can execute the discussion as follows:

1. There are the quasi-stationary excited states of molecules in the region $x < 1$, in particular, for $N \to \infty$ the fractional population $\langle A_{22}\rangle_S/N$ in the excited state $|\alpha_2\rangle$ is $\frac{1}{2}$ and the relative fluctuation σ^2/N^2 is finite and non-zero. It is characteristic of cooperativity of biological systems as open systems far away from thermal equilibrium. Therefore, this region may be regarded as a 'cooperative region' (or an 'ordered' regime).
2. In the region $x > 1$, both the fractional population in the excited state $|\alpha_2\rangle$ and the corresponding relative fluctuation tend to zero for $N \to \infty$, which means that the molecules behave as a group of individuals without any correlation with each other. Therefore, this region may be considered as an 'individual region' (or a 'chaotic' regime).
3. In the limit $N \to \infty$, both the fractional population in the excited state $|\alpha_2\rangle$ and the corresponding relative fluctuation show a discontinuous behaviour through $x = 1$, which may be considered as a non-equilibrium first-order phase transition at the critical point $x = 1$. At this point, the molecular populations are equally divided among the three states for any value of N, as has been shown in Eq. (29), which agrees with the result obtained in the case of the two-level exciplex model (see Eq. (12)).

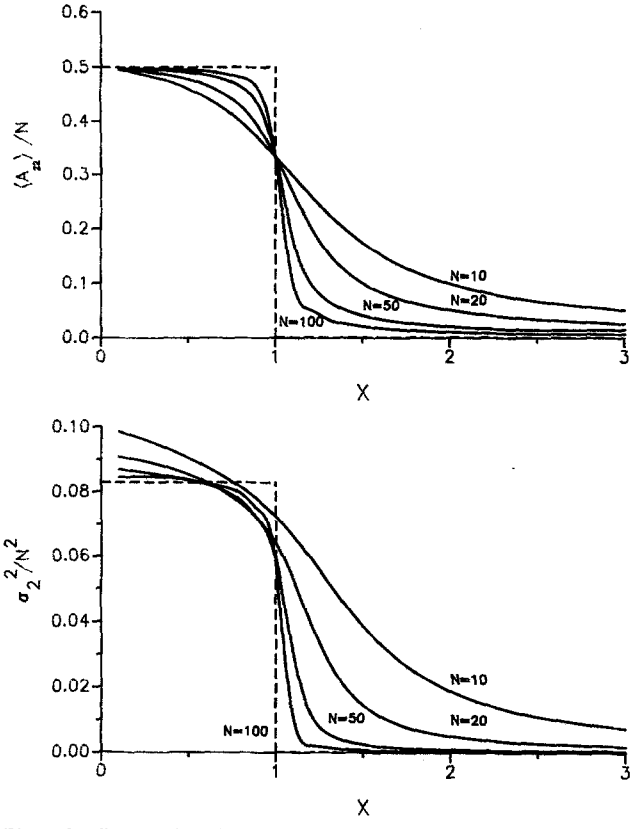

Figure 8. The quasi-stationary fractional populations $\langle A_{22}\rangle_S/N$ and their relative fluctuations σ_2^2/N^2 as functions of the parameter x, where $g_1^2 = \frac{2}{3}$ and $g_2^2 = \frac{1}{3}$ correspond to the experimental data[56]. The dashed curves represent the non-equilibrium phase transitions at $x = 1$, for $N \to \infty$. The critical behaviour agrees with that of Eq. (12) and Eq. (29).

Furthermore, the quasi-stationary intensity of biophoton emission, corresponding to transitions from $|\alpha_2\rangle$ to both

$|\alpha_1\rangle$ and $|\alpha_3\rangle$, can be given by

$$I = \langle A_{21}A_{12}\rangle_S + \langle A_{23}A_{32}\rangle_S$$
$$= \tfrac{1}{6}[(3N+5)\langle E\rangle_S - 2\langle E^2\rangle_S], \tag{31}$$

and its limiting values read

$$\lim_{N\to\infty}\frac{I}{N^2} = \begin{cases}\frac{1}{6}, & x \leqq 1\\ 0, & x > 1\end{cases}. \tag{32}$$

Obviously, in the region $x \leq 1$ the intensity is proportional to N^2 for large values of N, which exhibits a phase-locking in the sequences of molecules in the exciplex system[81]. Above the threshold $x = 1$, the intensity varies as N and the system behaves as a group of individuals radiating independently. One can see again that the threshold $x = 1$ marks a phase transition from an ordered to a chaotic regime. The N^2-dependence of biophoton intensity has been found to be supported by experiment[29].

At the threshold $x = 1$, the spectral distribution of biophoton emission, corresponding, for instance, to the transition from $|\alpha_2\rangle$ to $|\alpha_3\rangle$, can be written as

$$S(\omega - \Omega_2) = \gamma_2 \int_0^\infty d\tau \langle A_{23}(\tau) A_{32}\rangle_S \exp[-i(\omega-\Omega_2)\tau] + HC. \tag{33}$$

Eq. (33) results in a symmetrical structure with five peaks[21] located at the frequencies

$$\omega - \Omega_2 = 0,\ \pm G,\ \pm 2G\ (G = \sqrt{G_1^2 + G_2^2}),$$

which is found to be consistent with some experimental observations[59].

The coherence of biophoton fields can be understood from the normalized second-order intensity correlation function defined by[36]

$$g_{ij}^{(2)}(\tau) = \frac{\langle :\hat{I}_i(t+\tau)\hat{I}_j(t):\rangle}{\langle \hat{I}_i(t+\tau)\rangle\langle \hat{I}_j(t)\rangle}. \tag{34}$$

We are interested in the steady-state correlation function $g_{ij}^{(2)}(0)$, in particular, in

$$g_{22}^{(2)}(0) = \frac{\langle A_{23}A_{32}A_{32}A_{23}\rangle_S}{\langle A_{23}A_{32}\rangle_S^2}, \tag{35}$$

$$g_{21}^{(2)}(0) = \frac{\langle A_{23}A_{21}A_{12}A_{32}\rangle_S}{\langle A_{23}A_{32}\rangle_S\langle A_{21}A_{12}\rangle_S}. \tag{36}$$

One can get the analytical expressions[21] for $g_{22}^{(2)}(0)$ and $g_{21}^{(2)}(0)$ with respect to $\langle E^k\rangle_S$, and their limiting values read

$$\lim_{N\to\infty} g_{22}^{(2)}(0) = \begin{cases}1.2, & x<1\\ 1.6, & x=1,\\ 2.0, & x>1\end{cases} \tag{37}$$

$$\lim_{N\to\infty} g_{21}^{(2)}(0) = \begin{cases}1.2, & x<1\\ 0.8, & x=1.\\ 2.0, & x>1\end{cases} \tag{38}$$

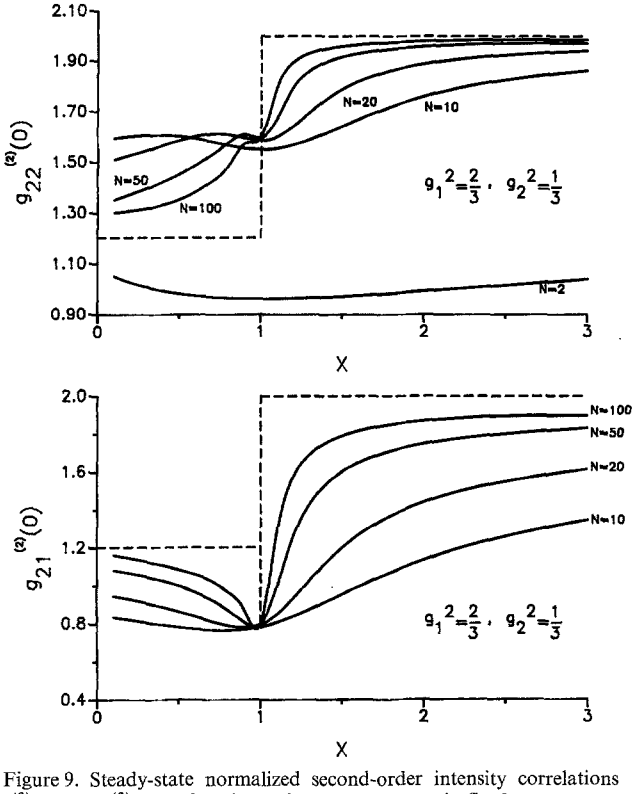

Figure 9. Steady-state normalized second-order intensity correlations $g_{22}^{(2)}(0)$ and $g_{21}^{(2)}(0)$ as functions of x. Data same as in fig. 8.

The plots of $g_{22}^{(2)}(0)$ and $g_{21}^{(2)}(0)$ as functions of x are shown in figure 9. In the region $x > 1$ and for $N \to \infty$, the value of $g_{22}^{(2)}(0) = g_{21}^{(2)}(0) = 2.0$ makes the emitted field fully incoherent. This is a manifestation of the fact that the molecules tend to independent radiation in this region. In the region $x < 1$ and for $N \to \infty$, one has $g_{22}^{(2)}(0) = g_{21}^{(2)}(0) = 1.2$, which means that the emitted field has quite a high degree of coherence. At the critical point $x = 1$, the value of $g_{22}^{(2)}(0) = 1.6$ indicates a partially coherent field, while the value of $g_{21}^{(2)}(0) = 0.8$ represents a non-classical field with an antibunching effect. By the way, the antibunching effect has already been observed in the experiments on biophoton emission[74, 82].

In order to study the dynamic behaviour of the system after excitation, one can write down the motion equation for $\langle E(t)\rangle (= 2\langle A_{22}(t)\rangle)$, starting from Eq. (23), in the form

$$\frac{d}{dt}\langle E(t)\rangle = a - b\langle E(t)\rangle + x_0\langle E^2(t)\rangle, \tag{39}$$

where a and b are the parameters of the system and $x_0 < 0$, $x_0 = 0$, $x_0 > 0$ correspond to $x < 1$, $x = 1$, $x_0 > 1$, respectively. Evidently, for the cooperative region ($x_0 < 0$), the critical point ($x_0 = 0$), and the individual region ($x_0 > 0$), the system displays different dynamics.

Considering the cooperative region and using the decorrelation approximation, one gets from Eq. (39) that

$$\frac{d}{dt}\langle A_{22}(t)\rangle = a - c\langle A_{22}(t)\rangle - 2|x_0|\langle A_{22}(t)\rangle^2, \quad (40)$$

with c as a parameter of the system.
Furthermore, the intensity emitted by the system after excitation is given, in terms of photocount rate, by

$$I(t) = -\frac{d}{dt}\langle A_{22}(t)\rangle + I_S, \quad (41)$$

where I_S describes the non-zero stationary intensity as $t \to \infty$. One can get from Eqs (40, 41) that

$$I(t) = R\,\mathrm{csch}^2\left(\frac{t+T_R}{\tau_R}\right) - A\,\mathrm{sech}^2\left(\frac{t+T_A}{\tau_A}\right) + I_S, \quad (42)$$

where R, T_R, τ_R, A, T_A and τ_A are the parameters of the system[21]. In Eq. (42), R-term and A-term describe the photon emission and absorption, respectively. Provided that the absorption term can be neglected, expanding Eq. (42) leads to the approximation:

$$I(t) = \frac{I_1}{(t+T_R)^\beta} + I_S, \quad (43)$$

where I_1 is related to the initial intensity, and $\beta = 2 + \delta$ with δ as a small quantity. Eq. (43) displays a hyperbolic decay (see, for instance, ref. 81).
Figure 10 shows a comparison of the dynamic decay according to the theoretical formula (42) and that obtained in the experimental results. From figure 10, τ_R, which reflects a certain coherence time of the biophoton field, can be estimated to be of the order of 1 s and then $|x_0|$ is of the order of $10^{-3}\,s^{-1}$. These results indicate that there is high-degree coherence within the biological systems and that they emit biophotons in such a way that their working points are around the threshold $x = 1$.

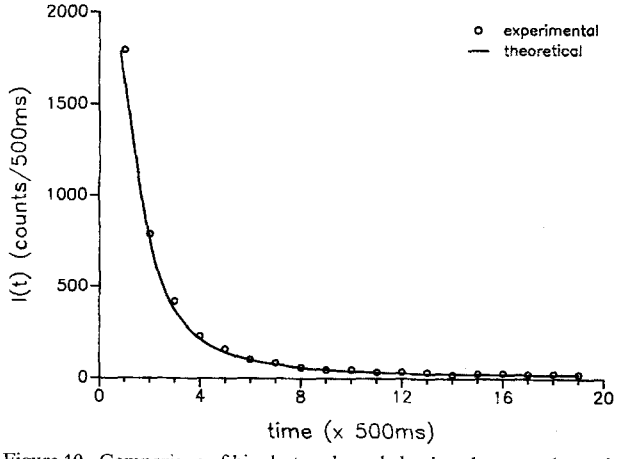

Figure 10. Comparison of biophoton decay behaviour between theoretical formulation Eq. (42) and the experimental results from rye grains which are excited initially by white-light-illumination. The measured intensities range from about 200–800 nm.

Concluding remarks

In this paper, using the exciplex models, we study the nonlinear effects in biophoton emission, such as chaos, fractal behaviour and non-equilibrium phase transition. These effects as responses of biophoton emission to external perturbations characterize the coherence nature of living systems. There are two important quantities in our approaches:

f (for the two-level exciplex model),
x (for the three-level exciplex model).

They play the roles of working points of two-level and three-level exciplex systems interacting with both the pumping and biophoton fields, respectively.
In our models, the influences of external perturbations on the exciplex systems, e.g., the change of temperature, the addition of agents, the exposure to light, etc., can be interpreted as shifts of the working points (f or x), leading to the presence of various kinds of nonlinear effects, as have been discussed above.
However, usually the living systems work within a very narrow region of variations of the parameters f or x around $f = 1$ or $x = 1$, which marks a critical point of non-equilibrium phase transition between an 'ordered' and 'chaotic' regime. Around the critical point biophotons display a diversity of their aspects, for instance, their photon statistical properties involve both partial coherence ($g_{22}^{(2)}(0) = 1.6$) and non-classical coherence ($g_{21}^{(2)}(0) = 0.8$). Such a diversity of behaviours of biophotons from living systems remaining at a phase-transition threshold guarantees the optimal flexibility of living tissues[81].

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0014-4754/92/11-12/1069-14$1.50 + 0.20/0

Photon emission of phagocytes in relation to stress and disease

E.-M. Lilius* and P. Marnila

**Department of Biochemistry and Department of Biology, University of Turku, SF-20500 Turku (Finland)*

Abstract. Phagocytes, the first-line cells of the body's defence mechanisms against invading pathogens, kill microorganisms by means of lysosomal degradative enzymes and highly toxic reactive oxygen intermediates. The reactive oxygen compounds are produced, in a process called the 'respiratory burst', by the NADPH oxidase complex in plasma membranes, and by myeloperoxidase in phagolysosomes after degranulation. These processes generate electronically excited states which, on relaxation, emit photons, giving rise to phagocyte chemiluminescence (CL). This paper describes the conditions for the measurement of CL, and reviews the activity of phagocytes from individuals undergoing stress or disease. The capability of phagocytes to emit photons reflects remarkably well the pathophysiological state of the host. In many cases even the magnitude of the stress, the presence of a pathogen in the body, or the activity of the disease can be estimated. Physiological changes, e.g. in the reproductive cycle, can also be predicted.
Key words. Chemiluminescence; phagocyte; stress; disease.

Introduction

Phagocytosing leukocytes constitute the first line of the body's defence mechanism against invading microbial pathogens. Neutrophils (polymorphonuclear leukocytes, PMNL) are the first cells to invade a site of inflammation following an infection. In an inflammatory response the neutrophils are followed later by activated monocytes, macrophages and – especially in the case of parasitic infection – also by eosinophils.

Phagocytes kill microorganisms by means of lysosomal degradative enzymes, such as proteases, and highly toxic reactive oxygen metabolites. Killing processes can take place inside the cell in phagolysosomes as well as outside the phagocyte.

In a process called the 'respiratory burst' activated phagocytes reduce molecular oxygen to superoxide via a special electron transport system (NADPH-oxidase). Superoxide radicals form hydrogen peroxide in a dismutase reaction catalyzed by the superoxide dismutase enzyme (SOD). Hydrogen peroxide serves as a substrate for the myeloperoxidase (MPO) reaction, in which a variety of highly toxic metabolites, including hypochlorite, are generated. These processes produce electronically excited