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INFLUENCE OF STRUCTURAL HETEROGENEITY ON ENERGY MIGRATION IN PHOTOSYNTHESIS

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The picosecond laser spectroscopy was applied in biology first of all in the studies of the primary stages of photosynthesis. With the help of multiple investigations which were carried out during the last 15 years a number of kinetic and spectroscopical parametres of the photosynthetic objects were defined. Partly, the mean times of the excitation energy transfer in the light harvesting antenna (LHA) as well as the charge separation in the reaction centre (RC) are known. The spectral forms of the chlorophyll molecules and their sequence of a participation in the evolution of the absorbed solar energy are also known. However, in spite of apparent evidence of the role played by each spectral form, the cause as well as the mechanicm of such effective light-harvesting are not completely clear. The measurement of the quantum yield of the charge separation in the RC, for instance, gets $\mu > 90\%$. The very primitive valuation of this quantity while modeling the LHA as homogeneous matrix of chlorophyll molecules and the RC being the trap for an excitation, shows that such high magnitude of the quantum yield of photosynthesis could be reached while supposing only the extremely short excitation trapping mean time.

It can be demonstrated in the following way. Let us assume that the excitation migration rate in the LHA is very fast and the

excitation lifetime (T) in the system is limited by the excitation trapping rate (τ_{RC}^{-1}) on the RC, i.e. $T^{-1} = \tau_o^{-1} + \tau_{RC}^{-1}/N$ (τ_o being the excitation lifetime on the isolated pigment molecule, N number of molecules which take part in the energy transfer). Then the excitation lifetime on the RC states which are in resonance with the excited states of antenna equals to T/N and the charge separation quantum yield can be calculated thus:

$$\mu = \tau_{RC}^{-1}/(N/T) \equiv (1 + \tau_{RC}\,\tau_o^{-1}N)^{-1} \qquad (1)$$

From formula (1) it follows that in the case of N being large, the high (near 100%) quantum yield is possible only assuming rather strict requirements to the ratio τ_{RC}/τ_o.

The present report is devoted to the discussion of the structural heterogeneity of the energy migration process in the LHA and the excitation traping on the RC. The accepted analysis lets us give unequivocal interpretation to the experimentally defined parametres as well as solve some contradictions while explaining the mechanism of the considered processes.

1. STRUCTURAL DATA

It is known that a photosynthetic apparatus both of bacteria and plants is build up by pigment-protein complexes (PPC) which are situated in the lipid membrane. The above-mentioned is approved by the following data.

The X-ray crystallographic investigations of the green bacteria (Prosthesochloris aestuarii) antenna complexes[1,2] enable us to establish the mean distance between the centres of chlorophyll molecules: within the PPC it equals to 12 Å and between them 24 Å. This fact points to the inhomogeneous location of antenna chlorophyll molecules. The experiments carried out with bacteria Rhodopseudomonas sphaeroides[3], corresponding to 880 nm and 800-850 nm respectively testify weak intercomplex pigment interaction. The change of the PPC relation shows the spectrum variations being additive. Otherwise the experiments on lin-

near and circular dichroism of individual comlexes[4] point to the strong (exciton-like) interaction between pigments.

It is possible to judge about the interlocation of the PPC in photosynthetic system from the experiments on electron microscopy. The measurements of bacteria[5], for instance, show a fine hexagonal structure. Besides, it must be mentioned, that each RC together with surrounding antenna forms so called photosynthetic unit (PSU).

2. THEORETICAL MODEL

On the grounds of the above-mentioned experimental data it is natural to propose the existance of two energy migration processes which are determined by intercomplex and intracomplex interactions. The assumption that the excitation decay time within the PPC (τ_{ex}) being much less than the intercomplex energy migration mean time (τ_{hop})[6],

$$\tau_{ex} \ll \tau_{hop}, \tag{2}$$

expresses the strong (exciton-like) pigment interaction within the PPC and the weak intercomplex interaction. In this case τ_{ex} is conditioned by electron-phonon interation and τ_{hop} incoherent excitation transfer between the PPC time (hopping time). The assertion of the intercomplex energy transfer being incoherent, is based on the comparison of the kinetic constants and the overlap of the chlorophyll molecules spectra[7]. Naturally, it turns out, that the excitation energy transfer to the molecule of the distance of 25 Å and even more, proceeds the Foerster-like mechanism. Now, when inequality (2) is satisfied, each of the PPC can be characterized approximately by a single level. In this case the evolution of the excitation can be described by the following balance-type equation set:

$$\frac{d}{dt} a_n^i(t) = \sum_j \sum_m^{N_c} H_{nm}^{ij} a_m^j(t) \tag{3}$$

where a_n^i is the probability of n-th PPC to be in the i-th excited state, N_c number of complexes in the PSU, H_{nm}^{ij} matrix describing the relaxation constants as well as the excitation transfer probabilities. It is enough to characterize the LHA complexes by a single level (i=1) and the RC - by three (i=0,1,2). If we take into consideration the symmetry of the PSU structure[5] the kinetic problem becomes and can be expressed as an energy transfer problem in one-dimensional structure where nodes are RC and its corresponding surrounding spheres[8]. It must be mentioned that the excitation transfer within a certain sphere does not play any role.

The essential moment of such consideration is the fact that N_c is not a large parametre ($N_c \cong 10$). Therefore effects of the local anisotropy of kinetic constants in the vicinity of the RC can influence on the excitation lifetime in the LHA. On the contrary, the homogeneous antenna model where a number of nodes taking part in the energy transfer process is fairly large ($N \gtrsim 100$), in case of 2- and 3-dimensional structures is characterized practically by one-exponential decay kinetics the same time being insensitive to the local anisotropy effect[9]. Direct solution of set (3) in the case of the PSU with one or two surrounding spheres[8] indicates to a two-exponential excitation decay kinetics. Besides, at any relation of the kinetic constants, these exponents practically are connected separately with the excitation transfer processes to the RC and with the trapping. Consequently, for such structures with a small number of nodes N_c, where the excitation proceeds the random walk, its decay kinetics can not be described by a single mean time T, to formula (1) is not applicable.

In fact the quantum yield of photosynthesis is determined by the occupation of state 2 (see Figure 1) at extremely large time (t), when t is much more than characteristic time of passing processes in the PSU:

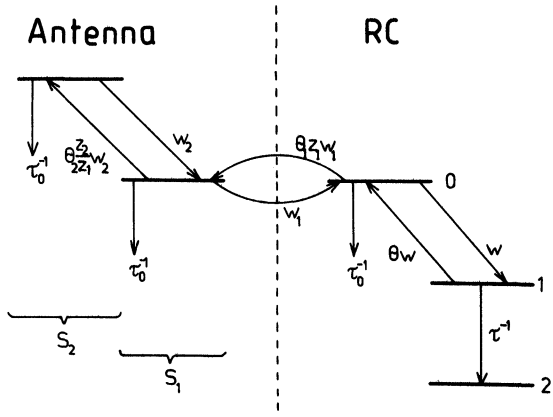

FIGURE 1 Scheme of energy levels of the PSU containing two surrounding spheres.
RC being characterized by three levels (0,1,2), S_1 near sphere to the RC, S_2 extrenal sphere. Rows point out the energy transfer and charge separation processes. W_i is the rate of intercomplex energy transfer from S_i sphere to the RC direction, z_i the PPC number in S_i sphere, τ_o the excited PPC lifetime, W the rate of primary charge separation in the RC, τ the separate charge fixation time, θ_i and θ parametres characrerizing migration anisotropy

$$\mu = \int_0^\infty a_o^1(t)/\tau \, dt \qquad (4)$$

τ being the separate charge fixation time in the RC.

Let us restrict ourselves for the case of two surrounding spheres. Naturally, such a model expresses qualitatively the process considered. As it can be seen directly from Figure 1, we will have 4 equations for the mathematical descrition, 2 of them describing the excitation evolution in the LHA and two others - on the RC.

While determining the quantum yield of photosynthesis, there is no need to solve the set of equations. So it is possible to express μ via coefficients at various power of the characteristic

equation of the eigenvalue:

$$\mu = \frac{1 + Q/(N_C \tau_o)}{1 + (\tau_{RC} N_{ef} + F)/\tau_o} \qquad (5)$$

where τ_o is the excitation decay time in the PPC, $\tau_{RC} = \theta\tau + W^{-1}$ the state of separate charge stabilization time in the RC, $N_{ef} = 1 + \theta_1 z_1 + \theta_1 \theta_2 z_2$, z_i the PPC number in the S_i-th sphere, $\theta_i = \exp(-\Omega_i/kT_o)$ parametres describing anisotropy of the kinetics in the direction to the RC and back, Ω_i the energy distance between corresponding levels, kT_o the temperature (in energy units), Q is determined by the initial conditions:

$$Q = \begin{cases} 0 & \text{excited only sphere } S_2 \\ z_1/W_2, & \text{excited both } S_1 \text{ and } S_2 \\ z_1/W_2 + F, & \text{uniform initial condition} \end{cases} \qquad (6)$$

$$F = W_2^{-1} + (1 + \theta_2 z_2/z_1)/W_1$$

In case of more than two surrounding spheres the value μ is exprssed by the analogical formula as (5).

If we assume the excitation migration rate being extremely high, i.e. $W_1, W_2 \longrightarrow \infty$, the value μ can be expressed thus:

$$\mu = (1 + \tau_{RC} \tau_o^{-1} N_{ef})^{-1} \qquad (7)$$

In the case of $N = N_{ef}$ formula (7) coincides with expression (1).

If we assume now τ_o = 1 ns, τ = 200 ps, W^{-1} = 7 ps, Ω = 0.1 eV, τ_{RC} = 10 ps, μ = 0.9 (parametres of the bacterial photosynthesis) so from formula (7) we get N_{ef} = 11. Since $\theta_1 \approx 1$ and if we assume that all the spheres are spectrally homogeneous (θ_2 = 1), so in this case the system should contain the RC and only a single surrounding sphere.

Let us analyze a more complicated situation of two surrounding spheres, assuming that $\theta_2 < 1$ which is inherent in real photosynthesis. Then, while changing the parametres we get μ = 0.82 to 0.99 (see Figure 2).

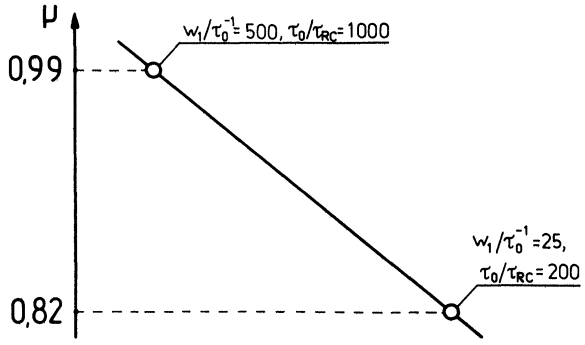

FIGURE 2 Schematical representation of change limits of the quantum yield of photosynthesis

It must be mentioned that, while taking into account the structural heterogeneity of the PSU, the high quantum yield of photosynthesis could be reached without any limitation for the kinetic parametres. The infinite rate of the excitation migration is not the propitions case for the increase of effectivity. The quantum yield of process essentally increases if the "focussing" effect, which manifests itself both in the process of rapid relaxation on the lowest excited state within the PPC parametres as well as in the anisotropy of the kinetics of intercomplex transfer takes place.

3. ANALYSIS OF KINETIC PARAMETRES

The presented model of the PSU gives a good explanation to the excitation transfer mechanism in the LHA. The comparison of the calculated fluorescence decay time[6] with the experimental measurements lets us determine a single intercomplex hopping time (τ_{hop}). From the experiment with bacteria Rhodospirillum rubrum it follows that T_{of} = 50 ps [10]. Then the single hopping time is of the order of 10 ps. Such a result completely coincides with the initial assumption of incoherent process of the excitation migration between

the PPC. Contrary, in the case of the homogeneous model of the PSU, the analogical estimation of the single hopping gives value less than 0.5 ps. Such a result testifies to impossibility of the energy migration incoherent-ixciton-like[11]. Naturally, it could be assumed that the energy migration is determined by coherent exciton[12]. However, this idea is not without contradictions. Partly, in this case we have to speak about a very close-packed structure of chlorophyll molecules in the PSU with a mean distance of order 10 A° which contradicts to the above-mintioned structural data.

The essential point of the given model containing one or two surrounding spheres is the fact that fluorescence decay kinetics should be two-exponential like. However, the majority experiments were carried out up to this time with exciting pulses of 20 ps duration or more. Therefore the kinetic curves difference from a single exponential function is difficult to determine. The recent experiments with 5 ps pulse duration[10] point out the single exponential kinetics could be explained by: i) a number of surrounding spheres being more than two, ii) the meantime of one of exponents being less than 5 ps. The very fact that the excitation decay time does not depend upon priority the LHA or the RC to excite[10], could testify to a very small parametre, of one of exponents (< 5 ps). Besides, there is no necessity to assume that the excitation migration in tha LHA should be very fast. That is because the number of the PPC of the first surrounding sphere z_1 and the ratio z_2/z_1 are the factors which exceed the excitation departure from the RC to the LHA. In the case of hexagonal structure ($z_1 = 6$), for instance, while assuming the single hopping time to be 10 ps, the time of the excitation transfer from the RC to the first surrounding sphere, would be equal to 1.6 ps. Consequently, it is possible that the fluorescence decay kinetics satisfies the two exponential law at any initial conditions that influence the relative

contribution of each of them.

The fluorescence quantum yield measurements with the increase of the excitation intensity witness the manifestation of annihilation effects. During the LHA excitation, the occupation of the first singlet state (S_1) of the PPC is created. The process of the singlet-singlet annihilation makes excitation pass from one excited PPC to the other excited one, letting the latter get to a higher excited state (e.g. in bacterial photosynthesis these states correspond to the Soret band of the absorption spectrum). Besides, such a process takes part not in an isolated PSU but in the domain being a set of the PSU on which free hopping of the excitation occurs. From that high energy state a very rapid intramolecular relaxation of excitation to the S_2 state of the LHA and subsequent relaxation from the S_2 to the S_1 state occurs, the lifetime of the former apparantly being relatively considerable[13,14]. Particularly, as it is evident from the analysis of bacterial photosynthesis (Rhodospirillum rubrum) [15] the dimension parameter r equals 1.1 ($r=2\gamma_1/\gamma_2$, γ_1 and γ_2 being the rates of the linear decay of S_1 excitation and annihilation of a pair of excitations correspondingly in the domain). Then we get $\gamma_2^{-1} \simeq 200$ ps. Assuming the fact that in the whole thickness of the sample the excited molecules are distributed homogeneously, one can estimate the mean time of the annihilation process by the formula:

$$\tau_a^{-1} = \frac{1}{2}\gamma_2 \lambda n_{PSU} \qquad (8)$$

n_{PSU} being the average number of absorbed photons per PSU in the whole thickness of the sample, λ the number of PSU in the domain. While inserting the value $\lambda = 16$ [15] and $n_{PSU} = 1$ we get $\tau_a = 25$ ps. However, straight from the measurements of the value γ_2 (as well as τ_a) it is difficult to judge about the parametres of the migration process though some authors have undertaken such attempts. Naturally, the annihilation time of a pair of excitation as it is seen from the analysis[15,16] is of order or less than 5 ps. There-

fore the supposition of the diffusion limited approximation to the annihilation process in this case is not applicable.

Recent investigations of the spectral and kinetic properties of the PSU by the difference absorption spectroscopy have appeared [17,18]. They show certain deviations from the fluorescence data:

1. In the difference spectrum of chromatophores absorption the minor bacteriochlorophyll spectral form having a longer wavelength than the absorption band of the RC photodonor is observed, and the excitation energy transfer occuring through the minor form to the RC is shown.

2. The mean time of the excitation decay in the LHA is essentially less than the corresponding time obtained from the fluorescence measurements. Besides, the kinetics of the excitation decay does not depend on the state of the RC.

In connection with minor spectral form of the LHA chlorophyll the hypothesis of the pericentral complex which is responsible for such a form was presented. Such an assumption casts out the doubt on the generally accepted conception of the multicentricity in the frame of the globular model of the LHA. That's why it is necessary to discuss these results in a more detailed way.

The measurements of the difference absorbance of the chromatophores[17,18] testify different temporal courses as well as light dependencies of the optical density of the sample at low ($I_o \simeq \simeq 10^{14}$ $h\nu/cm^2$) and high ($I_o \geqslant 10^{16}$ $h\nu/cm^2$) excitation intensities (normalization of the intensity per single pulse of the duration $\tau_o \simeq 25$ ps is used everywhere in text). The main argument, that the kinetics of the spectral changes in the case of $I_o \simeq \simeq 10^{14}$ $h\nu/cm^2$ reflects the picture of the natural photosynthesis, lies in the fact that the quantum yield of the charge separation is $\mu > 0.5$. In the case of used experimental parametres (the samples thickness $\sim$ 1 mm, the optical density of about a unit on

the wavelength ≈ 880 nm and the number of bacteriochlorophyll per RC ≈ 50) such an intensity corresponds to $n_{PSU} \approx 1$. However, as it follows from the above-presented discussion and formula (8) such intensities get $\tau_a \simeq 25$ ps. So the kinetics of the difference absorption can be explained in another way.

As it was mentioned above, the fluorescence decay time in the case of the open RC approximately equals to $\tau^o = 50$ ps [10]. Therefore, while all the RCs are opened, the initial excitation decay kinetics is mainly determined only by the singlet-singlet annihilation process because $\tau_a < \tau^o$. In course of time the contribution of annihilation reduces and the quenching of excitation by the open RC prevails. At still longer times the closed RCs start to influence the excitation decay process (the mean time of fluorescence quenching by the closed RCs $\tau^c = 180$ ps [10]). Actually, it is not difficult to show numerically that three exponential kinetics with indexes mentioned above ($\tau_a = 25$ ps, $\tau^o = 50$ ps and $\tau^c = 180$ ps) easily approximates the experimental kinetic curves [17,18]. Besides, it must be mentioned that the pre-exponential factor of the "rapid" exponent occurs to be essential (≥ 0.5) while the calculation of the quantum yield of charge separation for $n_{PSU} = 1$ giving the value $\mu = 0.5$.

The presented three-exponential kinetics of the excitation decay in the LHA enables us easily to understand its independence of the redox state of the RC. The excitation decay kinetics with the increase of the fraction of the closed RCs changes by the interplay of relative weights of excitation quenching by the open (τ^o) and the closed (τ^c) RCs. In the limit of all RCs closed the "rapid" (annihilation) and the "slow" (excitation quenching by the closed RC) parts of the kinetics remain only. Consequently, within the limits of the time resolution restricted by the duration of the probbing pulse ($\tau_o \simeq 25$ ps) the kinetics for the open or the

closed RC must differ only in the relative contribution of the "slow" exponent the fact being noticed in the experiment.

The origin of the minor spectral form could be also understood while analizing the difference absorption spectrum which could be derived thus[19]:

$$\Delta A(\lambda_{pr}) = \alpha[n_o \sigma_o(\lambda_{pr}) + n_1 \sigma_1(\lambda_{pr}) + n_2 \sigma_2(\lambda_{pr}) - \sigma_o(\lambda_{pr})] \quad (9)$$

α being the numerical coefficient, n_i the occupation of the ground (i=0) the first (i=1) and the second (i=2) excited singlet states, $\sigma_i(\lambda_{pr})$ is the cross-section of the light absorption (and stimulated emission in case of $I \neq 0$) of the i state at the wavelength λ_{pr} of the probbing pulse. The temporal dependence of ΔA is determined by the state occupations n_i, the exact values of which can be obtained by solving the corresponding kinetic equations.

In case of low intensities of the exciting light pulse $n_2 \ll 1$, hence, $n_1 + n_o \simeq 1$. Then from (9) it follows that:

$$\Delta A(\lambda_{pr}) = \alpha n_1(t)[\sigma_1(\lambda_{pr}) - \sigma_o(\lambda_{pr})] \quad (10)$$

σ_1 dependence on λ_{pr} is unknown. However, it should be pointed out that the transition from the first singlet state to the Soret band (in case of bacterial photosynthesis) is slightly blue-shifted in comparison with the maximum of the LHA absorption from the ground state σ_o. Bearing in mind that σ_1 in order of magnitude is equal to σ_o[13], and the above said, it follows directly that the minor spectral form is caused by the difference of two absorption spectra: $\sigma_1(\lambda_{pr}) - \sigma_o(\lambda_{pr})$. It must be pointed out, that while taking into account the heterogeneous structure of the LHA the additional phenomenon should be displayed in the difference absorption spectrum. Particularly, for one excitation in the dimer owing to the resonance interaction the optical transition to the

lowest singlet state could be red-shifted in comparison with that in the monomer. However, for the absorbed second light quantum the resonance interaction is absent, therefore on the dimer (as well as in the oligomers) at low excitation intensities the difference absorption spectrum will always possess the minor component in the longwavelength side of the spectrum. In such a case the light curves (e.g. Figure 3 in[17]) also become comprehensible. With the growth of the excitation intensity the process of singlet-singlet annihilation becomes more extensive, which leads to the nonlinear n_1 dependence on I_o. Besides, at very high intensities, when $n_{PSU} \gg 10$, it becomes possible for a new non-linear mechamism to come - the absorption of the second light quantum with the transition from the first singlet state to a higher one as well as to the ground state during the action of the exciting pulse. The nonlinear processes tend to diminish the difference between the occupations of the ground and the first singlet states, but at very high intensities owing to the relatively long lifetime of excitation in the second singlet state (see, e.g.[13,14]) a considerable occupation of the state S_2 occurs. Thus the change of difference absorption of the main band in the case of a very high excitation intensity could be explained. The lifetime of the state S_2 determines then the delay of the appearance of the minor component in the difference absorption spectrum.

An analytical solution of the kinetic equations for the occupations of the singlet states of the LHA while taking into account the pulse duration of the right-angled form with the width of 25 ps in case of excitation intensities $I_o = 10^{14}$ to 10^{16} $h\nu/cm^2$ gives the dependencies $n_1^{max} \sim I_o^{1/2}$ and $n_2^{max} \sim I_o$. Then from formula (9) it follows directly that with the increase of excitation intensity the occupation number n_2 starts to grow as soon as augmentation of n_1 (the signal of the minor spectral component) slackens. In the discussed region of the spectrum (880 nm) the absorption cross-section σ_2 apparently does not possess resonances and therefore the signal of the main component of the difference

absorption spectrum increases, thus qualitatively reproducing the experimental light curves.

We must point out two moments which can decisively affect while reaching the quantitative agreement between the model calculations and the experimental data. Firstly, the duration and the shape of the exciting and probbing pulses must be included. The use of the δ - like exciting pulse in the consideration mentioned above gives for $n_1(t)$ the dependence on I_o in a state of saturation, where t is the time passed after excitation. Secondly, as the nonlinear quenching should be taken into account, the inhomogeneous distribution of excitation in the PPC in the sample as well as the fluctuations in the domain can influence the result.

Thus the observable discrepancies of the results obtained by fluorescence and difference absorption spectroscopy in fact are apparent and qualitatively could be solved while taking into account the absorption of the probing pulse from the excited states of the LHA as well as the singlet-singlet annihilation in the description of the excitation evolution. Besides, the description did not contain any fitting parametres - all kinetic constants are taken from the fluorescence data.

In conclusion we will mention that the characteristic changes of the transitions $S_o - S_2$, $S_1 - S_2$, S_2 - the Soret band which must appear in the fluorescence and difference absorption spectra (of the regions 600 nm, 2000 nm, and 1240 nm respectively) can serve as confirmation of the explanation presented above. The investigation of their kinetics at the difference excitation intensities might give an adequate answer.

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