acta phys. slov. 43 (1993), No. 1, 114-129

COMPUTER MODELING OF COHERENCE EFFECTS IN EXCITATION TRANSFER IN HEXAGONAL PSU

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Received 27 January 1993
Accepted 15 February 1993

The time development of the site occupation probabilities in hexagonal model of photosynthetic units is investigated. The method based on Stochastic Liouville Equations allows to describe the excition transfer with a trap (reaction center) in coherent, quasicoherent and incoherent regimes.

I. INTRODUCTION

An energy captured in an antenna system (AS) of the photosynthetic unit (PSU) is transferred to a reaction centre (RC) in a form of localized Frenkel excitons.

The motion of excitons is permitted by electronic interactions between antenna pigment molecules. The excitation energy transfer has been investigated in a number of theoretical works, mostly supposing the incoherent regime [1,2].

In the last years the structure of variety antenna systems has been experimentally established [3]. The most striking property is their prevailing regularity. The photosynthetic membrane of purple bacteries shows well ordered two-dimensional arrays of photosynthetic units in which RC complex is surrounded by a ring like structure. The cyclic assembly is hexameric or dodecameric ring of $\alpha - \beta$ polypeptide pairs and their associated bacteriochlorophylles [3,4].

The simplest model of such antenna system contains globulas ($n = 1, \ldots, 6$) placed on the regular hexagonal with reaction center (RC) in the center ($n = 7$).

Kudzmauskas supposed [5] that the exciton within a single globula propagates coherently, while the exciton motion between globulas occurs by means of the mechanism of incoherent excitons. They used PME with homogeneous initial condition to calculate quantum yields and fluorescence decay time.

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Pearlstein and Zuber discussed the unability of the incoherent picture which uses the Förster mechanism for exciton transfer to explain regularity of cyclic antenna systems [4]. The necessity of incorporating of coherent and intermadiate regime was suggested on following grounds:

a) intrinsic, homogenous, linewidth of the photosynthetic pigments may be by many orders of magnitude smaller than the observed, inhomogenous, widths of the absorption bands,

b) new theories [6] and also [7–10] showed that the dynamics of exciton phonon interaction is much less randomizing than previously supposed.

These are new arguments against conclusions given in [11], which were already criticized by Nedbal [12], that the coherence time in antenna system is about 10^{-2}–10^{-1} psec.

Recently Borisov investigated several interaction mechanisms (resonant, exciton and exchange) and tried to point out a principal role of an intermolecular distance. He also questioned a common use of the Forster resonant mechanism in a description of the exciton transfer in PSU. He indicated the possibility of an application of the coherent picture for the exciton motion [13,14].

The character of the exciton motion (coherent or incoherent) is determined, beside the electronic intermolecular interaction, also by the exciton-bath (phonons) interaction. This fact introduces many difficulties. It is necessary to admit some simplifications. We are interested only in a propagation of the exciton but not in the dynamics of the bath. There are many theoretical methods solving the problem of finding the time dependence of the exciton occupation probability $P_n(t)$. We mentioned some of them in our previous papers discussing the role of coherence effects [15,16].

The most important one from the methodological point of view is the convolution Generalized Master Equation method (GME). The transition from the coherent regime to the incoherent one is well pronounced in a behaviour of the time dependence of the memory functions $w_{mn}(t)$ (MFs) entering convolution GME [7–9,17].

GME method can be connected with the Haken-Strobl Stochastic Liouville Equation method (SLE) (for a review see [18]), which can also describe coherent as well as incoherent regime and which is in this respect comparable with GME. The microscopic nature of parameters can be introduced for instance by the connection to GME [10]. A generalization to energetically heterogeneouse systems was given in [7,19].

On the other hand, the memory functions (MFs), which are the crucial quantities for convolution GME, are known only for some simple examples. (Henceforth, by GME, we understand convolution Generalized Master Equations for probability only.) Their use in the practical calculations of the time development of the site occupation probabilities $P_n(t)$ is rather cumbersome. A general analytical expression for calculation of coherent memory functions in finite systems can be found in [20], where we demonstrated the destructive role of the site-energy difference for the coherent regime.

The aim of the present paper is to model the time dependence of the exciton site-occupation probabilities $P_n(t)$ in hexagonal PSU and follow consequences of different kinds of excitation transfer regimes. We have chosen the SLE method for its capability of describing the coherent regime as well as incoherent one and owing to its proper description of the role of the trap.

Excitation energy is transfered through the antenna system to RC where primary processes take place. In our treatment, RC is modeled as a sink. The consequences of the sink in energy transfer processes are different in the coherent and incoherent regime [21]. Is it necessary to admit that in the exciton transfer treatment by GME method this fact has not been for a long time taken properly into account [17]. Čápek and Szöcs [22] were the first who pointed out the necessity of a "transformation" of MF. Absence of such a transformation could lead to completely errorenous results. They gave a prescription for a proper inclusion of the trap into the SLE method, too [22]). There is no such unique prescription for GME method.

In the following section we shall review shortly the theoretical description of GME and SLE methods and the transition between the coherent regime and incoherent one. In section 4, the parameters for SLE treatment of our model including a trap will be shown and in section 5, the results are presented. At the end the conclusions will be given.

II. GME AND SLE METHODS

Exciton motion in PSU can be described by the site occupation probabilities $P_n(t)$. Exciton-bath interaction and exciton trapping at the reaction center (RC), however, render an exact solution impossible.

The most general approach, starting from the first principles, is the Liouville equation

$$i\hbar\frac{\partial}{\partial t}\rho_{mr,ns} = [H,\rho]_{mrns}. \qquad (1)$$

Here H is the Hamiltonian of PSU including phonons, ρ is the density matrix, $m, n, (r, s)$ are molecular (vibrational) indices. The solution of (1), if found, would give a full information on the probability of any excitonic and vibrational state for any pigment molecule at any time.

Looking only for the exciton site occupation probabili ties $P_n(t)$ (relevant but reduced information) it is, however, much easier to solve GME [17]

$$\frac{\partial}{\partial t}P_m(t) = \sum_n \int_0^t [w_{mn}(t-\tau)P_n(\tau) - w_{nm}(t-\tau)P_m(\tau)]d\tau + I_m(t) \qquad (2)$$

$$P_m = \sum_r \rho_{mr,mr}.$$

From now on, for physical reason connected with initial conditions [17], the initial condition term $I_m(t)$ will be ignored.

The memory functions w_{mn} in (2) are complicated functions of hamiltonian matrix elements and temperature and, in most cases, can not be found explicitly. They reflect the character of the exciton motion – coherent or incoherent – in their decay for long times.

Only in exceptional cases (see the next section) GME can be further simplified to the Pauli Master Equations (PME)

$$\frac{\partial}{\partial t}P_m(t) = \sum_m (F_{mn}P_n(t) - F_{nm}P_m(t)) \qquad (3)$$

with transition rates

$$F_{mn} = \int w_{mn}(\tau)d\tau.$$

There is another model which can describe both coherent and incoherent motion and which is in this respect comparable with GME. This is the Haken-Strobl Stochastic Liouville Equation (SLE) method. The basic equations of the model read [18]

$$i\hbar\frac{\partial}{\partial t}\rho_{mn} = [H_0,\rho]_{mn} - i\hbar\delta_{mn}\sum_p 2\gamma_{m-p}(\rho_{mm} - \rho_{pp}) -$$
$$- i\hbar(1-\delta_{mn})[(\Gamma_m + \Gamma_n)\rho_{mn} - 2\gamma_{m-n}\rho_{nm}] \qquad (4)$$
$$\Gamma_m = \sum_n \gamma_{m-n}$$

Here ρ_{mn} is the density matrix for the exciton system, $P_m = \rho_{mm}$, H_0 relates to the exciton system unperturbed by the exciton-phonon coupling. The off-diagonal parameters γ describe the phonon induced hopping of excitons, the Γ's control the decay of the off-diagonal elements ρ_{mn} $(m \neq n)$ which parallels the loss of coherence of the exciton motion.

III. COHERENT AND INCOHERENT EXCITON MOTION

Transition from the coherent to incoherent character of the exciton motion could be well demonstrated on a simple example, namely homogeneous dimer with an intermolecular distance R.

Without interaction with a bath, the exciton motion between two molecules with an intermolecular interaction $J(R)$ has the coherent (oscillating) character, which is governed by the Schrödinger equation. Let us create, at $t = 0$, the excitation with $P_1(0) = 1$. Solving the Schrödinger equation, one can obtain

$$P_2(t) = (1 - \cos(Jt/\hbar))/2.$$

Let us allow an interaction between the exciton and a bath (phonons). In our recent papers we succeded in calculation of the long time asymptotics of the MFs for the exciton interacting locallly with phonons (bath) [8–10,23]. We demonstrated the two-channel character of MFs and we were able to connect phenomenological parameters in SLE with microscopic parameters entering the Hamiltonian.

For instance the memory function for a symmetric dimer takes for long time scale a form

$$w_{21} = w_{12} = Ae^{-Bt} + B\delta(t) = w^{qcoh} + w^{ic}$$

where A and B were given [8,10] for initialy relaxed lattice. We also determined [10] the role of parameters like temperature, strength of exciton-phonon interaction etc. in the exciton motion character in a linear chain.

From microscopically derived w_{mn} one can obtain the microscopic meaning of parameters γ in SLE. The following cases have been discussed [18]:

Purely coherent exciton motion

There is no exciton-bath interaction. Then $w_{12} = w_{12}^{qcoh}$ without damping and $\Gamma = \sum \gamma_{n-m} = 0$. There is no possibility of application of PME to this type of exciton motion.

Purely incoherent exciton motion

This limiting regime takes place when the exciton motion is owing to the second channel with negligible memory effects. One can use the PME with transition rates

$$F_{12} = 2\gamma_{12} = \int w_{12}^{ic}(t)dt.$$

Quasi-incoherent exciton motion

In this case $\Gamma \gg J$, and memory functions w^{qcoh} decay rapidly with time. On the long time scale, one can use PME with transition rates

$$F_{12} = 2\gamma_{12} + J^2/(\Gamma\hbar^2)$$

Quasi-coherent transfer

In this case the memory functions w^{qcoh} are damped, but this damping is not so strong, that the use of PME would be justified. One has to solve the GME or SLE.

Kenkre was the first who pointed out the non-local character of memory functions , previously obtained also by Sokolov [24], even for only nearest neighbor interaction J in H_0, and discussed consequences [25]. He tried also to introduce effective rates.

Pearlstein [21] was the first who discovered another role of the trap (sink) in the coherent exciton transfer regime than in the incoherent one. He demonstrated on finite chain with not very strong trap that in the coherent case the transfer of the excitation to the trap is slower. In the following development of theories based on GME method, Pearlstein's conclusion has received, however, no response.

To take into account the role of a trap (sink) on the exciton transfer one has to complete the SLE (2) by a term

$$-\sigma(\delta_{ms} + \delta_{ns})\rho_{mn}/2$$

Analysing carefully the time dependence of $P_m(t)$ Čápek and Szöcs pointed [22] the necessity of "transformation" of memory functions MF. As shown on a simple example, absence of such a transformation could lead to completely erroneous results. General prescription for such transformation of memory functions in more general cases has not yet appeared. This is also one of the reasons to use the SLE method in our model here.

IV. MODEL

Experiments analysing the structure of PSU has led to heterogeneity in models working with a planar cyclic hexagonal arrangement of globulas with RC in the center.

In our model the exciton motion within a globula is supposed coherent and we shall not calculate the occupation probabilities of finding the exciton on individual intraglobula molecules [5,15]. Our probabilities $P_n(t)$ refer to globulas as a whole. To describe the interglobula exciton transfer we shall use the SLE .

The Hamiltonian H_0 relates to the unperturbed exciton system and includes only the interaction between neighboring globulas. The symmetry of the problem leads to the following form of H_0

$$\begin{matrix} 0 & J & 0 & 0 & 0 & J & I \\ J & 0 & J & 0 & 0 & 0 & I \\ 0 & J & 0 & J & 0 & 0 & I \\ 0 & 0 & J & 0 & J & 0 & I \\ 0 & 0 & 0 & J & 0 & J & I \\ J & 0 & 0 & 0 & J & 0 & I \\ I & I & I & I & I & I & 0 \end{matrix}$$

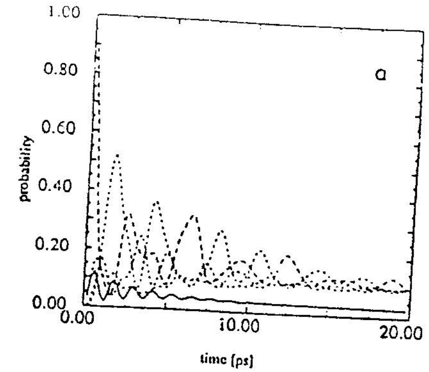
The off-diagonal elements of the Hamiltonian govern the coherent part in the exciton motion. I is the resonance (hopping) integral between RC and individual globulas, J is the resonance integral between antenna globulas.

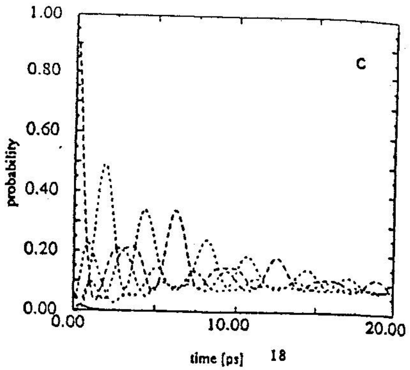
We shall concentrate on the coherence effects in the exciton transfer and their consequences. We shall include in our model the transfer through the quasicoherent channel [10] and we shall neglect a strong temperature dependent [8–10] incoherent channel ($\gamma_{mn} = 0$ for $m \neq n$) for which there is no direct experimental evidence. The coherent memory functions are then damped with factor $\Gamma = \gamma_0$.

SLE (2) is completed by the "sink" term

$$-\sigma(\delta_{m7} + \delta_{n7})\rho_{mn}/2,$$

which describes the capture of the exciton in RC by the help of a trapping rate σ.





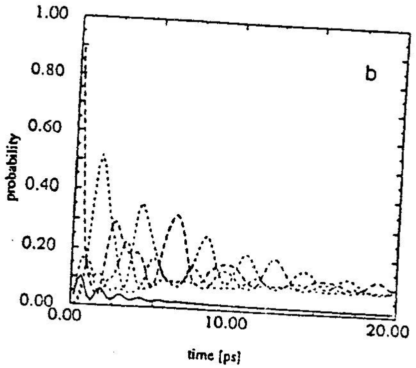


Fig. 1. Time dependence of the occupation probabilities

$P_1(t)$	AS	— — — —
$P_2(t), \ldots, P_6(t)$	AS	- - - - - - - -
$P_7(t)$	RC	————

for $\Gamma = 0.1$ and

$\sigma = 0.1$	a)
1.0	b)
10.0	c)

V. RESULTS AND DISCUSSION

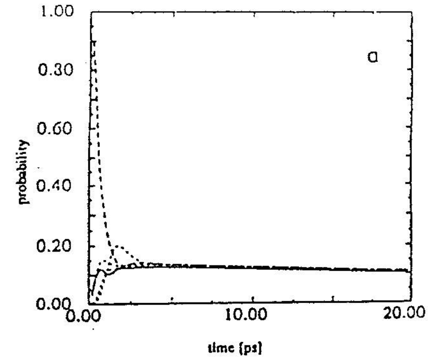
We are interested in the short time behaviour of the exciton. We shall not take into account the finite life time in the antenna system.

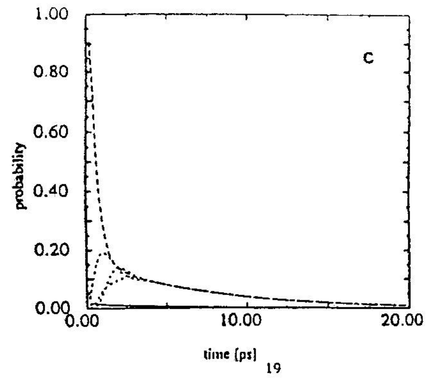
The computer modeling of the time development of the occupation probabilities $P_n(t)$ is shown on following figures for various initial conditions and a broad range of parameters. The time scale is given by $J/\hbar$ in psec^{-1}.

At first the initial condition corresponds to the excitation of one of antenna molecules at $t = 0$, it means that $P_n(0) = \delta_{n1}$. We are interested in the time development of

$P_1(t)$	AS	— — — —
$P_2(t), \ldots, P_6(t)$	AS	- - - - - - - -
$P_7(t)$	RC	————

We demonstrate the influence of the loss of coherence allowing the increase of the





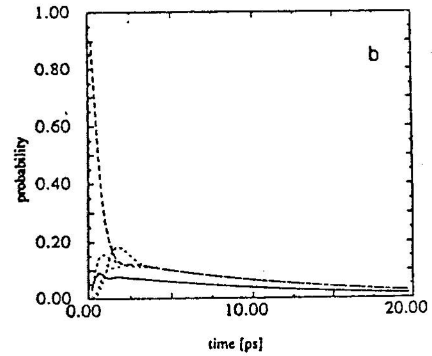


Fig. 2. Time dependence of the occupation probabilities

$P_1(t)$	AS	— — — —
$P_2(t), \ldots, P_6(t)$	AS	- - - - - - - -
$P_7(t)$	RC	————

for $\Gamma = 1.0$ and

$\sigma = 0.1$	a)
1.0	b)
10.0	c)

damping parameter in MF

$\Gamma = \gamma_0 = 0.1$	Fig.1
1.0	Fig.2
10.0	Fig.3

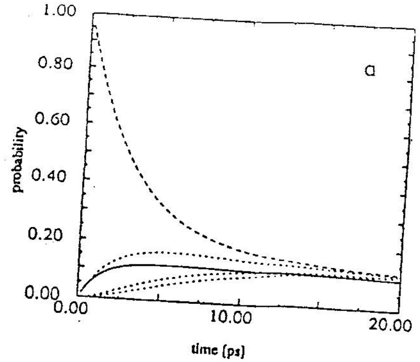
and the influence of the increase of the trapping rate σ

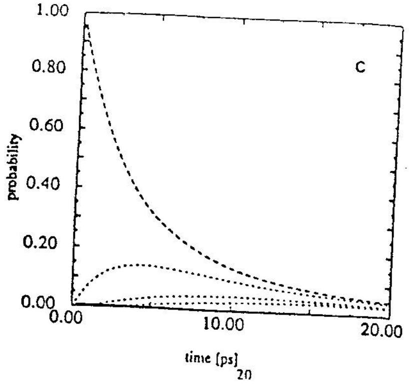
$\sigma = 0.1$	a)
1.0	b)
10.0	c)

for $J/\hbar = 1$ and $I/\hbar = 1$ [14].

In case of the coherent regime the probabilities $P_n(t)$ oscillate. With increasing Γ the oscillatory (coherent) regime is preserved only in initial stages.

We see that the increase of the trapping rate σ from zero leads to the capture of the exciton on the RC. On the other hand, for large σ, the exciton prefers to move around the decoupled AS avoiding the RC before being trapped at RC





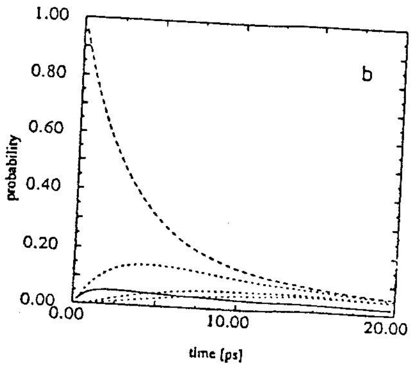


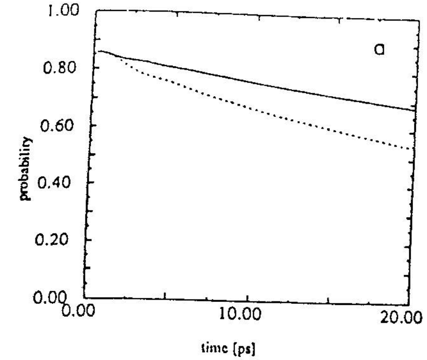
Fig. 3. Time dependence of the occupation probabilities

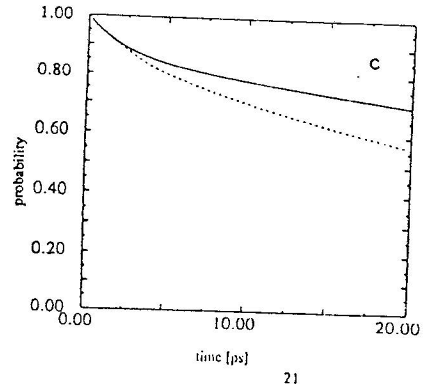
$P_1(t)$	AS	— — — —
$P_2(t),\dots,P_6(t)$	AS	- - - - - - - -
$P_7(t)$	RC	———

for $\Gamma = 10.0$ and
$\sigma = 0.1$ a)
1.0 b)
10.0 c)

[16,21]. These results support the conclusion, that the mutual relation between the incoherent and coherent regime in the presence of a trap is nontrivial [21] and the use of not-properly-transformed memory functions [16,17] as in [15] could be wrong. With the increase of coherence effects the excitation is stored in antenna system more pronouncedly and the transfer rate to RC is suprisingly suppressed [4,21].

It is very often argued [2] that after the initial period of several psec the occupation probabilities should reach a homogenous distribution in PSU and one can consequently use PME for their further time development. As it is seen from our results on Fig. 1-3 this conclusion is for presumably realistic higher degree of coherence in exciton transfer unjustified. The oscillatory behaviour may be preserved in the antenna system for long times. Here, one should realize that decay of memory after some time t_c does not mean necessarily that after that time, the transfer is already incoherent. The reason is that after any individual transfer (hop) the memory effects enter once more into game. Neglecting this fact means to distort e.g. the above mentioned suppressing role of increasing σ as in





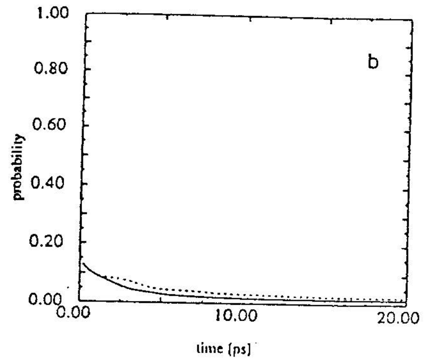


Fig. 4. Time dependence of the occupation probabilities:

antenna globulas	a)
raction center	b)
photosynthetic unit	c)

for $J/\hbar = I/\hbar = .3, \sigma = 0.5$ and $\Gamma = 0.1$.
Initially is excited PSU.

the Pauli-like theories.

In previous pictures we followed the excitation coming to the PSU for instance from neighboring units. This corresponded to the initial condition $P_1(0) = 1$. We supposed equal site energies of globulas in AS and RC. To be nearer to the experimental situation after (mostly optical) excitation we shall simulate the possible energy heterogenity of the initial distribution in PSU showing on next figures the time development of the occupation probabilities of

antenna globulas	a)
reaction center	b)
photosynthetic unit	c)

for $J/\hbar = I/\hbar = 0.3$ [14], and $\sigma = 0.5$ [ps^{-1}], [15] with more-general initial distribution. The initial excitation at $t = 0$ is

homogenous	$P_n(0) = 1/7$	$n = 1,\cdots,7,$	Fig.4.,5.
AS	$P_n(0) = 1/6$	$n = 1,\cdots,6,$	Fig.6.,7.
RC	$P_7(0) = 1$		Fig.8.,9.

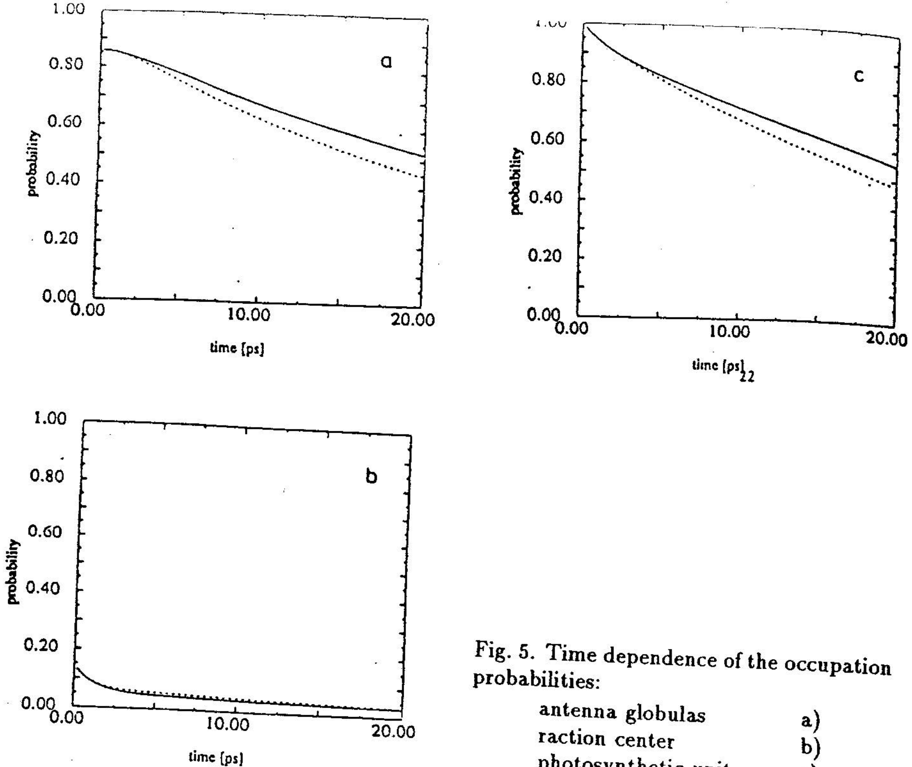


Fig. 5. Time dependence of the occupation probabilities:

antenna globulas	a)
raction center	b)
photosynthetic unit	c)

for $J/\hbar = I/\hbar = .3, \sigma = 0.5$ and $\Gamma = 1.0$. Initially is excited PSU.

and $\Gamma = 0.1$ or 1.0.

It should be mentioned here that the initially inhomogeneous distribution of $P_n(0)$ is necessarily connected with non-equivalence of site-energies; the latter feature may be, however, negligible for dynamical properties in later times, ie. it is neglected in this study limited to coherent effects in PSU.

We see that in the more coherent regime, $\Gamma = 0.1$, the most probable observation of coherent effects is in the case of initial excitation of RC.

Up to now we have used the homogenous Hamiltonian H_0. To demonstrate the influence of the difference of interantenna and antenna-reaction center transfer ($J = I$) the results for two times greater $J/\hbar$ are also shown. In any case the results do not coincide as believed up to now [2,5].

VI. CONCLUSIONS

Let us start with the description of the exciton motion in hexagonal PSU by the Pauli Master equation. Thorough discussion of the incoherent exciton motion

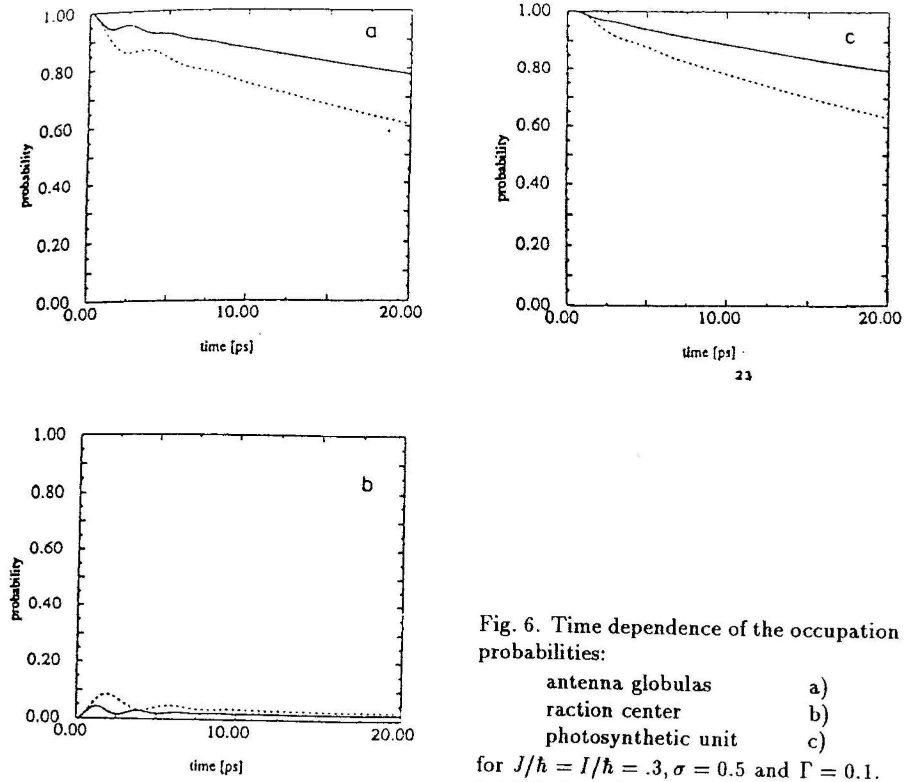


Fig. 6. Time dependence of the occupation probabilities:

antenna globulas	a)
raction center	b)
photosynthetic unit	c)

for $J/\hbar = I/\hbar = .3, \sigma = 0.5$ and $\Gamma = 0.1$. Initially is excited AS.

in the hexagonal model of PSU has been given in papers by Kudzmauskas et al. [5], Valkunas et al. [26] and Borisov [14].

Using the homogeneous initial condition, a two-exponential decay law in the long time development of antenna and reaction center probabilities was obtained. The crucial point is that none of the decay times is connected with the exciton transfer within an antenna system. Two exciton kinetics are connected with the symmetry of the system. One characteristic period is the decay of the exciton within the AS (here not taken into account) and the other – the seizure of the exciton by the RC.

The aim of our work was a computer modeling of coherence effects in exciton transfer in hexagonal PSU. We used the Stochastic Liouville equation method with a proper inclusion of a trap.

We developed a method of solving SLE which allows us to obtain the solution for any initial conditions solving system of 49 differential equations for density matrix elements ρ_{mn}. We did not try to fit input parameters in SLE to experimental data (also in PME treatment the obtained parameters are very ambiguous). On

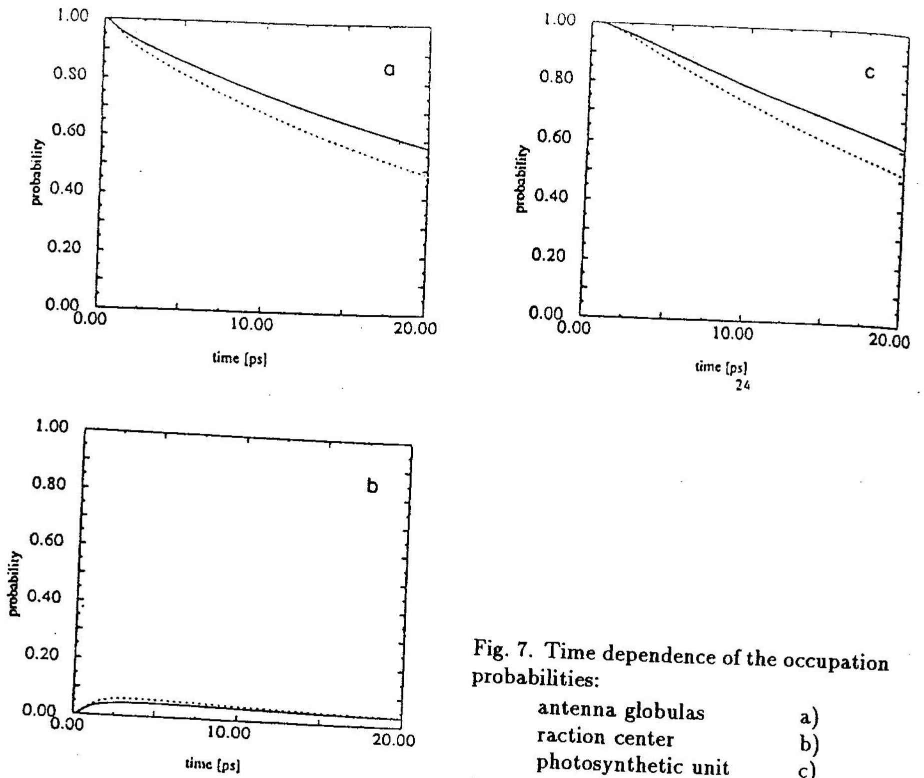


Fig. 7. Time dependence of the occupation probabilities:

antenna globulas a)
raction center b)
photosynthetic unit c)

for $J/\hbar = I/\hbar = .3, \sigma = 0.5$ and $\Gamma = 1.0$.
Initially is excited AS.

the other hand, we wanted to demonstrate, on a short time scale, consequences of the coherent exciton transfer regime, as suggested recently [14,21].

The obtained data were calculated and graphicaly presented for $J/\hbar$ scaled to 1 psec. Qualitative conclusions are not changed for another time scaling.

We used up the advantage of the SLE in capability of describing, besides the exponential (incoherent), as PME method, also oscillatory (coherent or incoherent) time development of the globula occupation probabilities $P_n(t)$. The coherence effects in the sense of time oscillations are preserved for shorter times with increasing incoherence, requiring the ultra-short time optical measurements for their discovery. The oscillatory character of $P_n(t)$ is smeared (averaged) in antenna and reaction center probabilities (Figs. 4–9) in the case of homogenous excitation of either AS or RC. The most probable opportunity to discover the oscillatory coherence effects are seen in the case of the initial excitation of RC (Figs.8–9). On the other hand the non oscillatory behaviour of RC does not rule out the coherent or quasicoherent regime in AS.

The second important result is the effective decoupling of RC from the AS

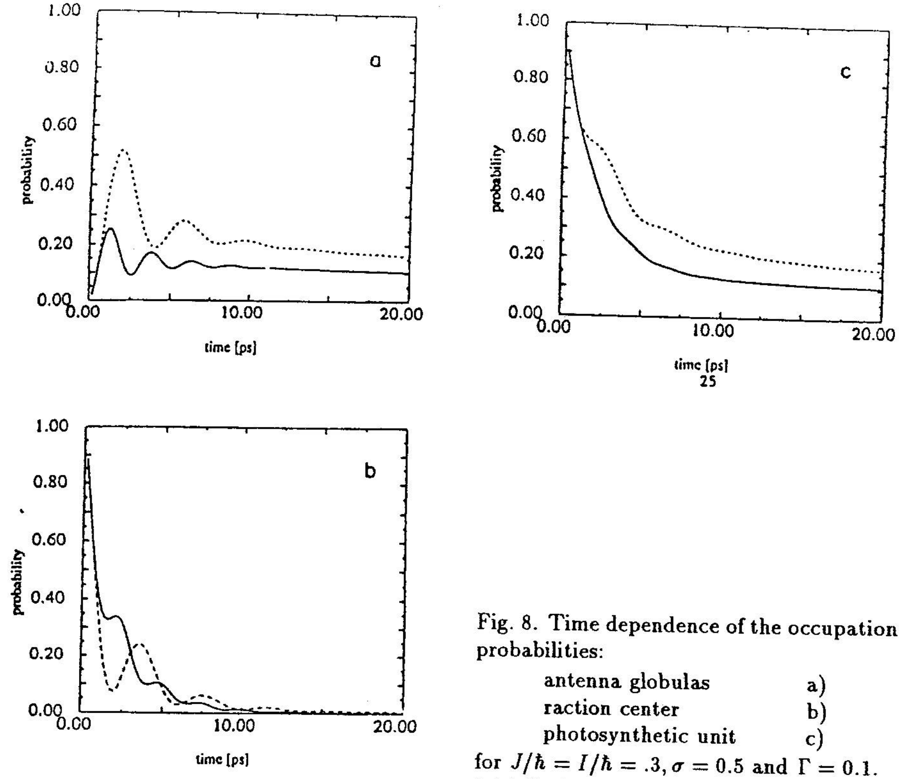


Fig. 8. Time dependence of the occupation probabilities:

antenna globulas a)
raction center b)
photosynthetic unit c)

for $J/\hbar = I/\hbar = .3, \sigma = 0.5$ and $\Gamma = 0.1$.
Initially is excited RC.

with increasing the trapping rate σ. The excitation remains stored in AS avoiding the transfer to RC.

The obtained results are the first step in our understanding of coherence effects in hexagonal cyclic PSU. A more complete model would require further steps in a more complete set of input parameters for SLE method which includes:

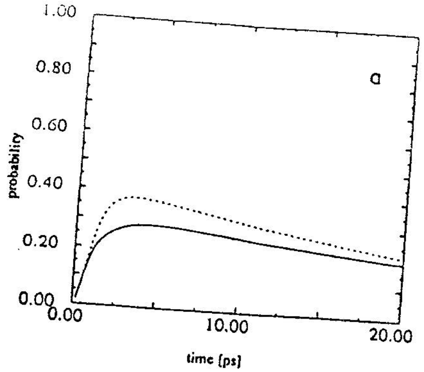
a) site-energy distribution in PSU

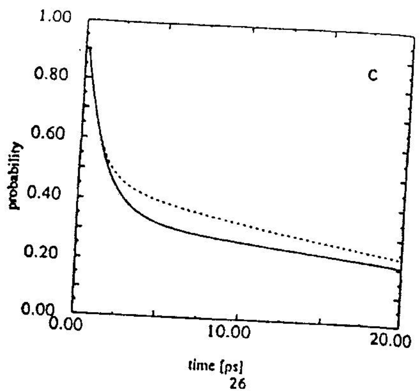
b) hopping assisted channel ($\gamma_{ij} \neq 0$)

For inclusion of the energy-inhomogeneity, we can use the extended SLE method which was developed by Čápek and Szöcs [19].

Acknowledgment

I am grateful to the International Centre for Theoretical Physics, Trieste for hospitality at the Research Workshop in Solid State, Atomic and Molecular Physics 1989, during which a part of this work has been done.





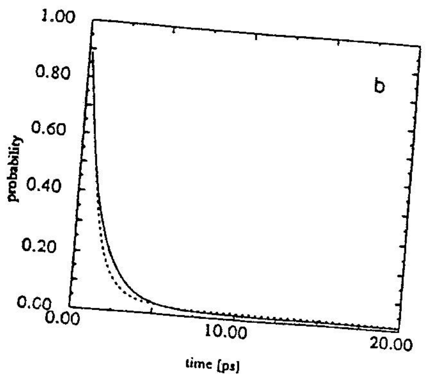


Fig. 9. Time dependence of the occupation probabilities:

antenna globulas a)
raction center b)
photosynthetic unit c)

for $J/\hbar = I/\hbar = .3, \sigma = 0.5$ and $\Gamma = 1.0$. Initially is excited RC.

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