On the long-wavelength component of the light-harvesting complex of some photosynthetic bacteria

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ABSTRACT The effect of the presence of a minor antenna component in light-harvesting complexes of photosynthetic bacteria is investigated with numerical simulation employing the transition probability matrix method. A model antenna system of hexagonal symmetry is adopted, using as a working hypothesis that the minor component forms a ring around the trap. Three cases have been considered: (*a*) the minor component is isoenergetic with the trap, which is at lower energy than the antennas (the "supertrap"), (*b*) the minor component is at lower energy than the trap, which is at lower energy than the antennas (the "asymmetric gutter"), (*c*) the minor component is at lower energy than the trap, which is isoenergetic with the antennas (the "gutter"). It is found that the supertrap speeds up the fluorescence decay and enhances the trapping efficiency, whereas the gutter slows down the fluorescence decay and decreases the trapping efficiency. It is concluded that, in contrast to a recent suggestion (Bergström, H., R. van Grondelle, and V. Sundström. 1989. *FEBS (Fed. Eur. Biochem. Soc.) Lett.* 250:503–508), concentrating excitations in the vicinity of the trap by the so-called long-wavelength minor antenna component purportedly present in *Rhodobacter sphaeroides* and *Rhodospirillum rubrum* instead of improving trapping actually impedes trapping.

INTRODUCTION

In recent studies of some bacterial light-harvesting systems, the existence of a minor antenna component was revealed, whose peak wavelength was some 20 nm red-shifted with regard to the phototrap (1–9). The energy difference, $\sim$260 cm^{-1}, represents at ambient temperature 125% of the thermal energy. This means that the minor component could act as a fairly deep trap, and consequently considerably hamper efficient energy transfer to the reaction center. On the other hand it has been advanced that the minor component is situated close to the reaction center and precisely because it is an energy trap, may help to prevent excitations, once they have reached the confines of the reaction center, getting lost by back diffusion into the outer domains of the antenna complexes (6, 7, 10, 11).

In this communication we wish to explore the idea that the minor component helps concentrating excitations in the neighborhood of the reaction center, by the simulation of excitation migration in a model system. We have used the transition probability matrix method for obtaining accurate curves of the time evolution of excitation trapping as well as values for the trapping yield at infinite time. The major outcome of the present study is, that situating the minor component close to the reaction center hinders rather than promotes photochemical trapping at the reaction center. Only if the minor component and the reaction center both are at a lower energy than the antenna pigments does the minor component enhance the efficiency of photochemical trapping. Since at least at ambient temperature, the latter condition does not reflect the observation that the reaction center absorbs at roughly the same energy as the major antenna pigments, we conclude that there does not seem to be a simple functional explanation for the presence of the minor component.

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METHODS

The transition probability matrix method was first applied to photosynthetic antenna arrays by Robinson (12) and later used to study model antenna systems in plant (13) and bacterial (14, 15) photosynthesis. In this method, the probabilities of excitation transfer among pigments organized in a symmetric array are grouped in a so-called transition probability or transfer matrix and an initial set of excitation probabilities of the pigments is defined. Repeated operation of the transfer matrix represents successive steps of excitation transfer until ultimately a (quasi)equilibrium is reached. For symmetric arrays the method allows very fast simulation of excitation motion, so that the system can be studied under a great variety of conditions, parameter variation et cetera in an interactive way.

Because bacterial and plant antenna systems show quite nice hexagonal symmetry (16–21), we have chosen for our model of the antenna system a two-dimensional triangular lattice of antenna pigments surrounding the photochemical trap (Fig. 1). To represent the "minor antenna component" we label the first hexagonal ring of antennas adjacent to the trap "*b*", and allow them to absorb at an energy different from that of the trap or the major antennas. The next and subsequent rings of antennas are taken to absorb at the same energy, which may be the same as or higher than that of the trap. The system of connected photosynthetic units (PSUs) of hexagonal symmetry is described by adopting periodic (totally reflecting) boundary conditions. Throughout we assume that we have low-light conditions, i.e., on the average less than one excitation per PSU.

The motion of the excitation in the antenna bed can be described by calculating for each step the probability to be on a certain type of pigment, *a*, *b*, et cetera. This probability is given by the product of the probabilities on each previous step:

$$P(n) = M \cdot P(n-1), \quad (n > 0), \tag{1}$$

with M the transition probability matrix transforming the probability vector after $n = 1$ steps into that after n steps, and $P(n)$ a column vector whose components are composed of the probabilities to have the excitation on *a*, *b*, et cetera after the n^{th} step, $P(n) = [a(n)\ b(n)\ c_2(n)\ c_1(n)$

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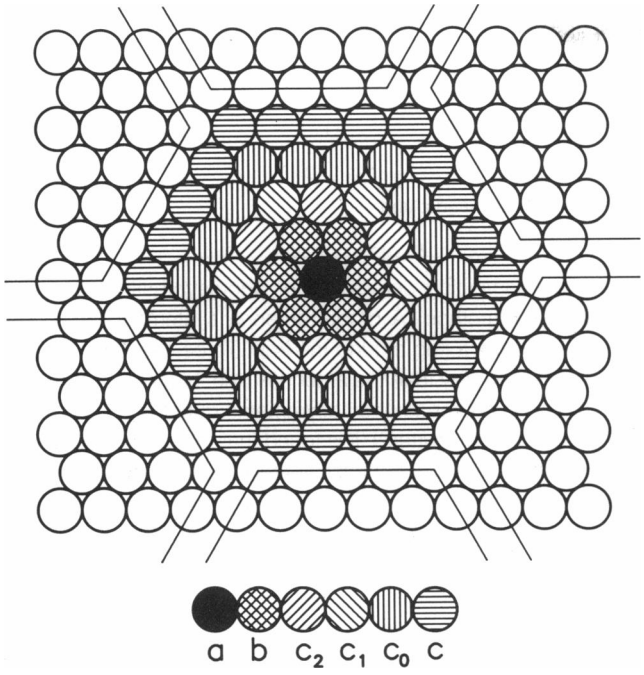

FIGURE 1 Model of a photosynthetic unit. *a* is the trap; *b* a minor antenna component; c_2, c_1, c_0, *c* the major antenna pigments, c_2 and c_1 having two or one connection(s) with antennas of the first *b* ring, respectively. The pigments of the third ring (labeled c_0 because they are not connected to the *b* ring) differ in their connection to the second ring: they are adjacent to either a c_1 or a c_2 antenna. This difference is taken into account in calculating the average probability of energy transfer from a c_0 antenna to a c_1 or a c_2 antenna. All antennas of the outer ring, *c*, are assigned the same, averaged connectivity. Further *c* rings may be added as required.

$c_0(n)\ c(n)]^*$, with the asterisk denoting transposition to the column vector. $P(0)$ is given by the initial conditions; for the excitation motion starting on *b*, for example, $P(0) = [0\ 1\ 0\ 0\ 0\ 0]^*$. The elements of M are easily calculated considering the connectivities of the various types of pigments (22, 23). Nonideal photochemical trapping on *a* is represented by an escape probability f, fluorescent losses by a loss factor l for each transfer step.

RESULTS AND DISCUSSION

Excitation migration in a homogeneous lattice

We have applied our algorithm to calculate the kinetics of fluorescence for a homogeneous lattice and a number of values of f, the probability to escape from the trap and N, the number of pigments in the PSU. To simulate an experimental fluorescence decay curve we have multiplied the number of steps by the hopping time h, which is a known function of the photochemical trapping rate (3 ps^{-1}), the fluorescence lifetime for the PSU with a closed trap (1,100 ps), and the loss factor l (22, 23). To a very good approximation all curves could be fit with single exponentials. This close to single-exponential behavior of the fluorescence decay is largely due to averaging over all starting sites. Curves for starting the motion of the excitation on a *c* pigment, for example, show a pronounced initial sigmoidity (in the first few steps there is no loss of the excitation through trapping), whereas those for a start on a *b* pigment show an initial fast decrease (the probability for trapping is in the first few steps much larger than later on) (22). These higher decay modes persist for times that are roughly proportional to the size of the PSU. The characteristic decay time of the surviving single exponential, however, is independent of the starting condition. The dominance of the zeroth order diffusion mode agrees with earlier observations (24–27). We have found that this dominance persists even for strongly heterogeneous lattices (see below).

Elsewhere (22, 23) we have demonstrated that by combining the theory of random walks and the probability matrix with experimental data on the fluorescence decay in PSUs with open and closed traps, it is possible to determine the value of f that is compatible with the observed fluorescence lifetimes and the known size of the antenna system. The result is $f = 0.8$–0.9. We have used $f = 0.9$ in the simulations of excitation migration discussed below.

Effects of a gutter-type minor antenna component on the efficiency of trapping

The energy transfer rate from *c* to *b* and vice versa will be different from that between the *c* pigments because of the change in Förster overlap integral. This difference is difficult to estimate, as the absorbance and fluorescence bands are broad and asymmetric because of inhomogeneous broadening, and the 0–0 transitions and Stokes shifts are not well known. For the small energy difference considered here (260 cm^{-1}), comparable or less than the bandwidth, the transfer from *c* to *b* will be somewhat faster than that between the *c* pigments because of better overlap between the fluorescence band of *c* and the absorption band of *b*, whereas the transfer from *b* to *c* will be slowed because a reduction in overlap between donor and acceptor bands. The resulting asymmetry is taken into account by an asymmetry factor y ($0 \le y \le 1$) representing the ratio of the total transfer rates from *b* to *c* and vice versa, with which the transition probabilities from *b* to c_2 and c_1 is multiplied. For simplicity we take the downhill transfer rate from *c* to *b* equal to the transfer rate between the *c* pigments and treat y as a variable parameter. This simplification underestimates somewhat the accumulation of excitations on the *b*-ring. To compensate for this the asymmetry factor y can be chosen somewhat smaller (the "trap depth" somewhat larger). It will be seen later that the above simplification does not affect our conclusions.

The energy of the trap *a* will be varied between that of *b* and that of the *c* pigments. Similar reasoning as above for the y factor holds for an x factor that represents the ratio in transfer rates from *b* to *a* and vice versa. Because

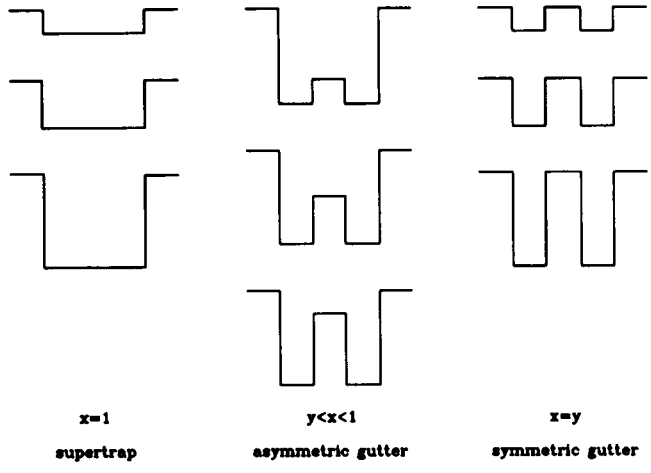

FIGURE 2 Schematic representation of the three types of heterogeneous antenna arrays considered: supertrap, asymmetric gutter and symmetric gutter for various values of x and y (see text). (*Supertrap*) The trap absorbs at the same energy as the minor antenna pigments, which energy is lower than that of the major antennas. (*Asymmetric gutter*) The energy at which the trap absorbs is higher than that of the minor, but lower than that of the major antenna pigments. (*Symmetric gutter*) The trap absorbs at the same energy as the major antenna pigments, which energy is higher than that of the minor antennas.

the total hopping probability equals unity, the probability to hop from b to b has to be increased by a factor z, which is given by $x/6 + y/3 + y/6 + 2z/6 = 1$ or $z = (6 - x - 3y)/2$. Because the b pigments are indistinguishable, this amounts to an increased mean residence time for the b pigments, in keeping with the notion that they constitute an energy trap. We investigate the fluorescence and trapping properties of our model PSU for values of x and y that bracket the Boltzmann factor corresponding to the energy difference between minor and major antennas. We will distinguish three different situations, illustrated in Fig. 2: the "supertrap" ($x = 1, y < 1$), the "asymmetric gutter" ($y < x < 1$), and the "symmetric gutter" ($x = y < 1$).

The supertrap

Fig. 3 shows the fluorescence and trapping curves for a "supertrap" of various depth, starting the excitation motion randomly. Apart from the first 10–20 ps, identical curves are obtained for starting the migration of excitations on a particular pigment (not shown). Two effects of an increasing depth are immediately apparent: the trapping and concomitant fluorescence decay become increasingly faster and the trapping efficiency at infinite time is somewhat enhanced. The extent of both effects is about the same for the two sizes of the PSU considered. Similar results are obtained for $N = 61$ (not shown). The effects are not difficult to explain: the a and b pigments together now form an extended energetic trap; once the excitation has arrived on b it is increasingly difficult to leave with increasing depth of the supertrap and the probability to be photochemically trapped in a is enhanced. A similar conclusion was drawn earlier by Seely (17). These simulations would validate the concept that the minor antenna complex functions to enhance the cross-section for trapping were it not that in reality the pigments a and b are not at the same energy level. To see what happens when we increase the energy of a relative to b we first consider the second case (see below).

The asymmetric gutter

We now let x be smaller than unity but larger than y. Because the probability to transfer to a becomes smaller, this corresponds to raising the energy of a relative to that of b. Fig. 4 shows simulations of the trapping and fluorescence kinetics for three combinations of x and y, again for random excitation and the PSU sizes as in Fig. 3. We now see the opposite effect: raising the energy of a relative to b the trapping and the fluorescence decay become slower, and the trapping efficiency at infinite time is decreased. In other words, increasing the energy of a undoes the trap-promoting property of the supertrap. The more symmetric the gutter becomes with decreasing x the more severe the slowing of the trapping is, until for a symmetric gutter ($x = y$) the fluorescence decay is actually slower and the trapping less efficient than when there is no gutter at all ($x = y = 1$). This corresponds to the third case.

The symmetric gutter

Here, $x = y$ and Fig. 5 shows the time dependence of trapping and fluorescence decay for various depths of the

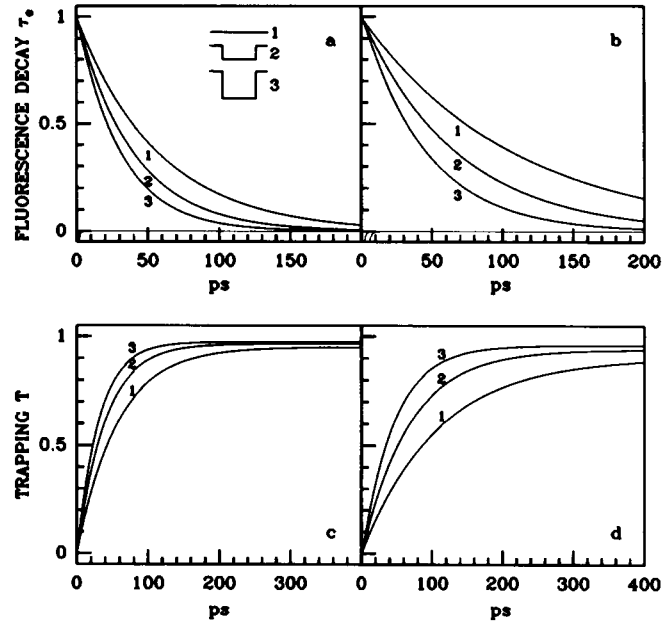

FIGURE 3 Time dependence of the fluorescence (*a, b*) and the trapping yield (*c, d*) for the supertrap ($x = 1$) with random excitation. Curves 1, 2, 3 for $y = 1, 0.5, 0.25$ (see inset *a*). $N = 19$ (*a, c*) and 37 (*b, d*); $f = 0.9$, loss factor $l = 1 - 2.8(1 - f)/1100f = 0.99972$, hopping time $h = 2.8(1 - f)/lf = 0.31$ ps (22, 23). The residuals of a single exponential fit to the fluorescence decay, multiplied by a factor 50, are indicated by fine lines in the lower left corner in the order 1, 2, 3 from left to right.

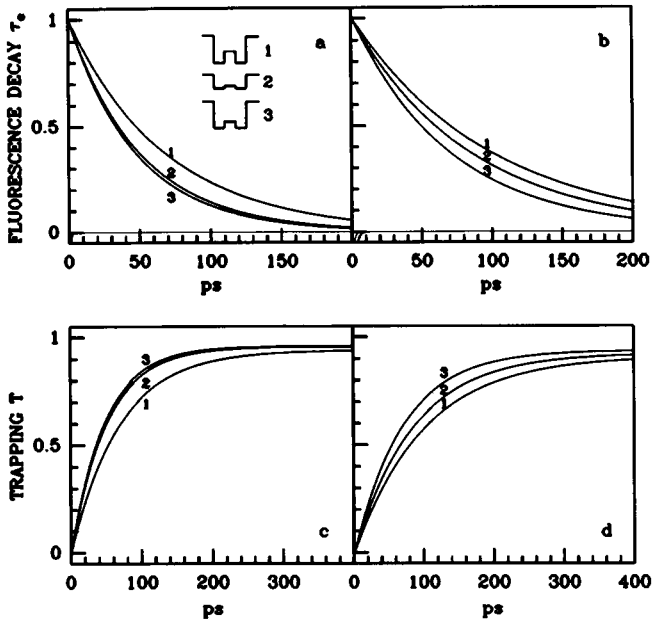

FIGURE 4 Same as Fig. 3 for the asymmetric gutter ($y < x < 1$) with $x = 0.4$, $y = 0.25$ (*curve 1*), $x = 0.75$, $y = 0.5$ (*curve 2*) and $x = 0.6$, $y = 0.25$ (*curve 3*) (see *inset, a*). $N = 19$ (*a, c*) and 37 (*b, d*). The residuals are given in the order 2, 1, 3 from left to right, further conditions as in Fig. 3.

symmetric gutter. The kinetics become considerably slower with increasing depth, whereas the trapping efficiency at infinite time becomes much lower than for a "gutterless" antenna system (all pigments at the same energy, $x = y = 1$). In other words, the more the minor antenna component acts as an energy trap the lower is the efficiency of the PSU! With the above results for the supertrap and the asymmetric gutter the explanation is readily found. Once the excitation is trapped in the ring of *b* pigments, its probability to be photochemically trapped in *a* is lower the deeper the *b* trap is, whereas the probability to be detrapped by transfer to the major antennas is several times higher than that of photochemical trapping. The net result is that the gutter actually impedes excitation transfer to *a* instead of promoting it as was suggested in references 10 and 11.

The above results are in general agreement with those reported by Fetisova and co-workers (14, 15, 28), who used the transition probability matrix method for studying a variety of heterogeneous lattices. We stress, however, that these workers have used an ad hoc trapping efficiency of unity ($f = 0$), whereas we have used the value $f = 0.9$ that was extracted from experimental data on the fluorescence decay with the aid of the relations developed in (22, 23). This makes it rather difficult to make a detailed comparison.

Just as for the homogeneous lattice, the fluorescence decay curves for the heterogeneous lattice deviate initially from a single exponential, especially when the excitation is started on a particular type of pigment. This deviation from a single exponential persists somewhat longer for the heterogeneous lattice than for the homogeneous one, but is still of much shorter duration than the subsequent exponential decay mode (22, 23) (zeroth-mode dominance [24–27]). It follows that the effect of the higher diffusion modes will be difficult to observe, even when exciting in a long-wavelength component (22, 29). For random excitation the higher diffusion modes practically disappear (Figs. 4, 5). It follows that the multiphasic fluorescence decay that is often observed cannot be due to heterogeneity of antenna pigments that form part of the PSU (i.e., that are well connected to the trap). Instead, the nonexponential decay must be due either to antenna components that are not, or very badly connected to the PSU that contains the trap, or to some other, non-energy transfer process, such as radical recombination to the excited singlet state of the primary donor (30).

In our model calculations we have used a uniform hopping time *h*. In actual fact *h* is not uniform, as the residence time of an excitation on a particular pigment depends on the transfer probability, which is site-dependent (*a*, *b*, or *c*-type). For cases of practical interest, however, the excitation is rapidly spreading into the major antennas, even for selective excitation of the *a*, *b* pigments, and after a few initial steps, the site-averaged transfer time is step-independent. This corresponds to the rapid initiation of the zeroth-mode dominance discussed above. Thus, only if one desires to make a detailed study of the higher diffusion modes is it necessary to include step-dependent transfer times into the model.

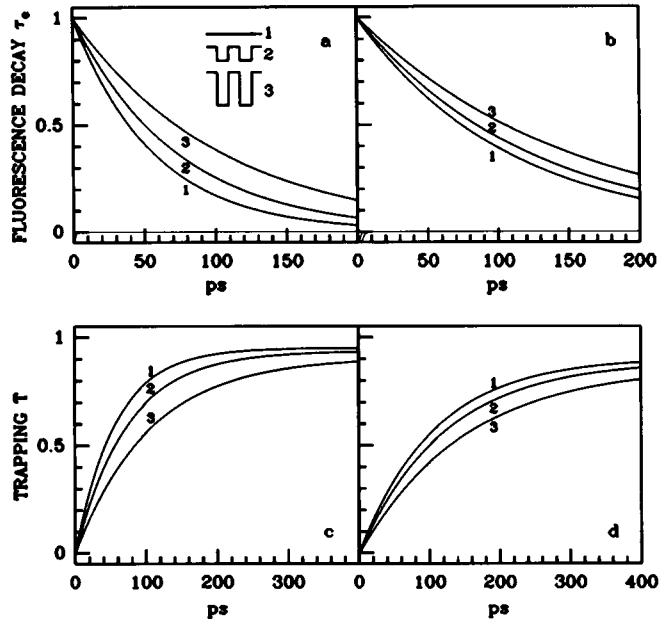

FIGURE 5 Same as Fig. 3 for the symmetric gutter ($x = y$). Curves 1, 2, 3 for $x = y = 1$, 0.5, 0.25 (see *inset, a*). $N = 19$ (*a, c*) and 37 (*b, d*). The residuals are given in the order 1, 2, 3 from left to right; further conditions as in Fig. 3.

Is the long-wavelength minor antenna component real?

Although the above results are found for one particular model of the PSU the conclusion that a symmetric gutter-type minor antenna component impedes photochemical trapping is quite general, because it rests on the topological property that the number of contacts of the minor antennas with the major antennas is always larger than with the single photochemical trap. Only when the gutter formed by the minor antennas is significantly asymmetric will the minor antenna component help trapping. This situation might prevail at cryogenic temperatures where the reaction center absorption has shifted to ~890 nm, i.e., close to that of the minor antennas (which do not shift appreciably with temperature [10]). Then, the minor antennas and the reaction center would form a supertrap, and the trapping rate would be enhanced. At physiological temperatures, however, there is presently no evidence that a supertraplike arrangement occurs, as then the minor antenna component absorption is shifted some 20–30 nm to lower energy from both the reaction center and the major antenna absorptions, and the situation of Fig. 2 *c* applies. It is self-evident that when the minor antennas do not form a contiguous assembly, or are isolated from the photochemical trap, the situation is even worse, because then the ratio of the number of contacts with the major antennas to that with the photochemical trap is even larger.

It has been suggested (11, 31), that the distance between the minor antennas and the trap is appreciably larger than that between the major antennas. In other words, between the *b* pigments and the *a* trap there is a "moat". It is obvious, that such an arrangement always hinders trapping, regardless of the relative energy of the *a* and *b* pigments (i.e., at all temperatures). Photons directly absorbed by the trap will have a higher probability to be photochemically trapped (they have a lower escape probability) than without moat, but excitations in the minor antennas (and in the major if the minor antennas surround the trap) have a lower probability to reach the trap. Since for random excitation the latter are in the majority, trapping will be significantly decreased, i.e., replacing the minor antenna (*b*) pigments by major antenna (*c*) pigments will always help trapping.

We must conclude that at present it is difficult to find a teleological explanation for the presence of a low-energy minor antenna component and that quite possibly, its observation is due to inhomogeneous broadening of the absorption band of the "major" antenna system (see for example recent work by Freiberg and co-workers [32–34]).

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