Energy Migration in the Light-Harvesting Antenna of the Photosynthetic Bacterium *Rhodospirillum rubrum* Studied by Time-Resolved Excitation Annihilation at 77 K

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ABSTRACT The intensity dependence of picosecond kinetics in the light-harvesting antenna of the photosynthetic bacterium *Rhodospirillum rubrum* is studied at 77 K. By changing either the average excitation intensity or the pulse intensity we have been able to discriminate singlet-singlet and singlet-triplet annihilation. It is shown that the kinetics of both annihilation types are well characterized by the concept of percolative excitation dynamics leading to the time-dependent annihilation rates. The time dependence of these two types of annihilation rates is qualitatively different, whereas the dependencies can be related through the same adjustable parameter—a spectral dimension of fractal-like structures. The theoretical dependencies give a good fit to the experimental kinetics if the spectral dimension is equal to 1.5 and the overall singlet-singlet annihilation rate is close to the value obtained at room temperature. The percolative transfer is a consequence of spectral inhomogeneous broadening. The effect is more pronounced at lower temperatures because of the narrowing of homogeneous spectra.

INTRODUCTION

Excitation energy migration in the light-harvesting antenna (LHA) and electron transfer in the reaction center (RC) are the crucial processes of the primary photosynthesis responsible for a very high quantum yield of the charge separation—more than 90% of the absorbed photons eventually cause the transfer of an electron through the membrane. The pathway and rates of electron transfer in the RC are quite well known and understood. Only the very first transfer step away from the special pair (P) is still subject to discussion (see, e.g., Fleming and van Grondelle, 1994). At the same time, the microscopic parameters and even the very nature of the excitation transfer in the antenna systems (exciton relaxation or incoherent hopping) are not entirely established yet. Various experimental methods and conditions have been used to address this problem. At low excitation intensities the main relaxation channel of the antenna excitation is quenching by the RC (Borisov et al., 1985; Sundström et al., 1986; van Grondelle et al., 1987; Freiberg et al., 1989; Timpmann et al., 1991; Werst et al., 1992; Müller et al., 1993). These studies have given a picture of the overall excitation transfer and trapping at conditions close to natural light harvesting. However, from picosecond studies it has been difficult to obtain unambiguous information about the details of the elementary transfer steps. For example, the comparative analysis of time-resolved fluorescence and absorption measurements for the simplest LHA of the photosynthetic bacterium *Rhodospirillum rubrum* (Pullerits et al., 1994b) suggested a single-step pairwise transfer time on the order of a picosecond at room temperature. On the other hand, recent measurements with femtosecond time resolution have revealed kinetic components of a few hundred femtoseconds at room temperature as well as at cryogenic temperatures (Visser et al., 1995), which were also interpreted as energy transfer within the LHA. Furthermore, some ultrafast features in the femtosecond pump-probe signal of LHA of purple bacteria were recently interpreted as exciton relaxation, with a time constant of a few tens of femtoseconds (Pullerits et al., 1994a).

Alternatively, a nonlinear annihilation approach has also been used to study the excitation energy transfer in the LHA. For instance, the process of singlet-singlet (S-S) annihilation involves the interaction of two singlet excitations when they are close to each other. This can be visualized as follows. Because of such an interaction one excitation is transferred to the other excited molecule, creating a doubly excited molecular state, which relaxes very quickly by internal conversion to the singly excited molecular state or to the ground state (den Hollander et al., 1983; Bakker et al., 1983; Trinkunas and Valkunas, 1989; Valkunas, 1989). On the other hand, in experiments with high pulse repetition rates (and consequently high average intensities), triplet state pigment molecules can be accumulated (Valkunas et al., 1991). In that case, during the migration process singlet excitations may encounter a molecule in the triplet state and interact with it. In a manner similar to that of S-S annihilation, the singlet excitation can be transferred to the molecule in the triplet state, creating a higher excited triplet state. This state will very quickly relax to the lowest triplet state, whereby a singlet excitation has been annihilated by the triplet state (singlet-triplet, S-T, annihilation). In both cases one singlet excitation disappears. In earlier works the dependence of the fluorescence quantum yield on the pulse

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intensity was measured (Bakker et al., 1983) and provided direct evidence for the presence of S-S annihilation and enabled the estimate of a hopping time of ~0.5 ps in the antenna systems of purple bacteria.

Comparison of measurements of linear transfer and the nonlinear annihilation of excitations suggested that the structural organization of the LHA and the RC contains a few scaling parameters: one of them determines the mean interpigment distance within the LHA, and another describes the distance from the LHA to the RC (Valkunas et al., 1992; Somsen et al., 1994). Thus, it was proposed that the overall excitation trapping by the RC does not depend significantly on the fast hopping but is mainly determined by the energy transfer from the LHA to the RC. Such a conclusion of fast energy transfer within the LHA is supported by the excitation depolarization kinetics (Bergström et al., 1988), and slow transfer from the LHA to the RC was proved by detrapping studies (Timpmann et al., 1993; Xiao et al., 1994).

The spectrum of the LHA of *R. rubrum* is obviously inhomogeneously broadened (Timpmann et al., 1991; Pullerits and Freiberg, 1992; van Mourik et al., 1992). Being of minor importance at room temperature, the effects of spectral inhomogeneity become more pronounced at lower temperatures. Qualitatively this was already evident from experiments with picosecond time resolution, where energy equilibration among the pigment molecules of the inhomogeneously broadened spectrum was observed as very fast (mostly not resolved) spectral shifts or decay components at 77 K (Freiberg et al., 1987; van Grondelle et al., 1987; Pullerits et al., 1994b). More recent experiments with femtosecond resolution were capable of resolving the very fast dynamics, both at room temperature and at lower temperatures, demonstrating that there are relaxation processes on the femtosecond time scale that presumably occur within a single spectroscopic entity (a pigment molecule, an aggregate of the strongly coupled pigments, etc.; Visser et al., 1995; Hess et al., 1995), and energy equilibration over the whole LHA is basically completed within a few picoseconds. At very low temperatures the excitation may be trapped by energetically low-laying antenna pigments, giving rise to pronounced nonexponential excited state decay. It was the slow part of this decay that was observed in early low-temperature picosecond measurements as a nonexponentiality in the overall antenna decay. Lattice models of the LHA providing analytical expressions for the energy migration generally describe the pigment array as isoenergetic (Pearlstein, 1982; Valkunas et al., 1986). This is obviously an oversimplification, and inhomogeneity has to be included for a more complete description.

Because of the fast energy migration in the LHA, the excitation diffusion radius, which determines the size of the domain of the common LHA, covers more than 500 pigment molecules. In that case, the approximation of small domains (den Hollander et al., 1983), which has been used for the analysis of the intensity dependence of fluorescence quantum yields (Bakker et al., 1983; Deinum et al., 1989), does not seem to be appropriate. Moreover, direct kinetic measurements of the nonlinear relaxation processes contain much more information as compared to the time-integrated quantum yield measurements. The formalism for analyzing such experimental results was recently developed for singlet-singlet annihilation kinetics at room temperature (Valkunas et al., 1995).

In this work we present a time-resolved study of S-S and S-T annihilation in the antenna system of the photosynthetic purple bacterium *R. rubrum* at 77 K. We will focus on the effect of spectral inhomogeneity, which at 77 K is expressed more strongly than at room temperature.

MATERIALS AND METHODS

The excitation annihilation dynamics were measured with a two-color transient absorption pump-probe spectrometer based on a picosecond dye laser system with variable pulse repetition rate (Zhang et al., 1992). Chromatophore samples of *R. rubrum* in a buffered (Tris, pH 8) 3:1 (v/v) glycerol:water glass at 77 K were excited at 867 nm by a ~12-ps pulse, and the recovery of the transient bleaching of the antenna Bchls was monitored with a delayed pulse of similar duration in the wavelength range of 885 to 901 nm. The cross-correlation of this laser system is about 15 ps, resulting in an effective time resolution of about 2 ps when measured kinetics are deconvoluted with the apparatus response function. Because of the high repetition rate of the laser pulses, the reaction centers were rapidly converted to their oxidized state P^+. Kinetics were therefore measured for chromatophores with closed (P^+) RCs with an antenna excited state lifetime of ~200 ps in the absence of annihilation. Closed RC conditions are useful for detecting additional kinetic components due to singlet-singlet and singlet-triplet annihilation, because these expectedly faster kinetics will be more clearly resolved against the background of the slow ~200-ps P^+ quenching of the antenna excitations (as opposed to the 50–60-ps lifetime of active RC; Borisov et al., 1985; Sundström et al., 1986).

With the variable pulse repetition rate of the dye laser/cavity dumper, in combination with pulse intensity attenuation using neutral density filters, we can vary both average and peak power in a controlled manner over a wide range. As described above, S-S annihilation will occur at high instantaneous pulse powers, whereas S-T annihilation will occur at high average powers (high pulse repetition rates), because of build-up of long-lived triplet states. Thus, with this laser system we can selectively prepare the photosynthetic units in a state where either S-T or S-S annihilation occurs or, alternatively, in an annihilation-free state. Low repetition rate (80 kHz) and maximum pulse energy (~2.5 nJ/pulse, in all measurements the diameter of the beam in the sample was ~ 0.1 mm) generate S-S annihilation, high repetition rate and higher average intensity (4 MHz, ~0.7 nJ/pulse) favor S-T annihilation, and for annihilation-free kinetics we have used a pulse frequency of 800 kHz and a pulse energy of ~ 0.2 nJ/pulse. The corresponding average light intensities on the sample were 0.2 mW, 3 mW, and 0.15 mW, respectively (see Table 1).

TABLE 1 Pulse energies, average intensities on the sample, and pulse repetition rates at various experimental conditions for the curves in Fig. 1

	Pulse energy (nJ)	Average intensity at the sample (mW)	Pulse repetition rate (kHz)
A	0.2	0.15	800
B	2.5	0.2	80
C	0.7	3.0	4000

The diameter of the exciting beam is 0.12 mm.

RESULTS

Theoretical background

Singlet-singlet annihilation in the homogeneous LHA results in the following kinetic equation:

$$\frac{dn}{dt} = I(t)(N - n) - kn - \gamma(t)n^2, \quad (1)$$

where n is the concentration of excited pigment molecules in the domain and N is the total concentration of pigments, $I(t)$ is the excitation generation function, k is the linear excitation decay rate (mainly due to the excitation trapping by the RC), and $\gamma(t)$ is the time-dependent singlet-singlet annihilation rate constant. The time dependence of $\gamma(t)$ is due to i) the correlative effects of the excitations (Trinkunas and Valkunas, 1989; Valkunas, 1989; Valkunas et al., 1995) and ii) to the dimensionality effect of the antenna organization (Valkunas et al., 1995; Bunde and Havlin, 1991). Below we show that the latter effect can be caused by the spectral inhomogeneity of the system, where the excitations migrate over the longest-wavelength pigment pool, and the shorter-wavelength pigments can be considered as energy barriers for the excitations. In such a case, the LHA behaves as a fractal structure (Bunde and Havlin, 1991) and by investigating the time window starting from a few picoseconds (the relaxation to the randomly distributed reddest pigments takes a few hundred femtoseconds; van Grondelle et al., 1994; Visser et al., 1995), Eq. 1 is still valid. However, now N and n refer to only the reddest pigments in the domain.

The solution of Eq. 1 for times longer than the pulse duration can be written as

$$n(t) = \frac{n_0 \exp(-kt)}{1 + n_0 \int_0^t \gamma(t') \exp(-kt')dt'}, \quad (2)$$

where n_0 is the initial concentration of excited pigments.

For three-dimensional large systems, γ is time independent (Ovchinikov et al., 1989). In such a case, Eq. 2 can be rewritten as

$$n(t) = \frac{n_0 \exp(-kt)}{1 + n_0\gamma k^{-1}[1 - \exp(-kt)]}. \quad (3)$$

In the general case, $\gamma(t)$ is determined by the pair correlation function of the excitations (Valkunas et al., 1995). For diffusion-limited annihilation the asymptotic time dependence of $\gamma(t)$ corresponds to a power law (Bunde and Havlin, 1991), i.e.,

$$\gamma(t) = \frac{\gamma_0}{t^\alpha}, \quad (4)$$

where

$$\alpha = 1 - \frac{d_s}{2}, \quad \text{if } d_s < 2$$
$$\alpha = 0, \quad \text{if } d_s \geq 2. \quad (5)$$

d_s is the spectral dimension of the fractal structure or is equal to d for Euclidean structures.

For short-time kinetics when $kt < 1$, by substituting Eq. 4 into Eq. 2 the following analytical solution can be obtained:

$$n(t) = \frac{(1 - \alpha)n_0}{1 - \alpha + n_0\gamma_0 t^{1-\alpha}}. \quad (6)$$

In the case of singlet-triplet annihilation, the high pulse repetition rate accumulates part of the Bchl molecules in the triplet state with an average stationary concentration, and the corresponding kinetic equation is

$$\frac{dn}{dt} = -kn - \gamma_{ST} n n_T, \quad (7)$$

which can be rewritten as

$$\frac{dn}{dt} = -K(t)n, \quad (8)$$

where

$$K(t) = k + \gamma_{ST} n_T. \quad (9)$$

The time dependence of $K(t)$ is due to the correlative effects of singlets and triplets via the time dependence of γ_{ST}. The asymptotic kinetics for this process is determined by the decay kinetics of the singlet excitations, which reside in the areas of the LHA domain free of triplets. Thus, the kinetics have to respond to the most probable size of such a volume free of triplets, and therefore becomes sensitive to the dimension of the structure under consideration, i.e. (Bunde and Havlin, 1991),

$$n(t) \propto \exp(-At^{d/(d+2)}), \quad (10)$$

where A is a constant dependent on n_T. Eq. 10 also holds for fractal structures by substituting d by d_s.

Analysis of the experimental data

A representative set of the experimental data at various excitation conditions is presented in Fig. 1 (see Table 1 for pulse energies and average intensities). The annihilation-free low-intensity curve decays with a single-exponential lifetime of 210 ps because of the quenching by closed RCs. The average intensity used to measure the kinetic curve at the lowest pulse repetition rate is only slightly higher than in case of the low-intensity curve. Therefore we can practically exclude the possibility of the accumulation of triplets in this case. At the same time the decay is significantly faster, strongly suggesting the presence of singlet-singlet annihilation. On the other hand, for the curve measured at a high pulse repetition rate, the pulse energy is only about three times higher than that used for the low-intensity curve, whereas it is still about four times less than what was used to obtain the curve with S-S annihilation. The rate of S-S annihilation is proportional to the square of the density of

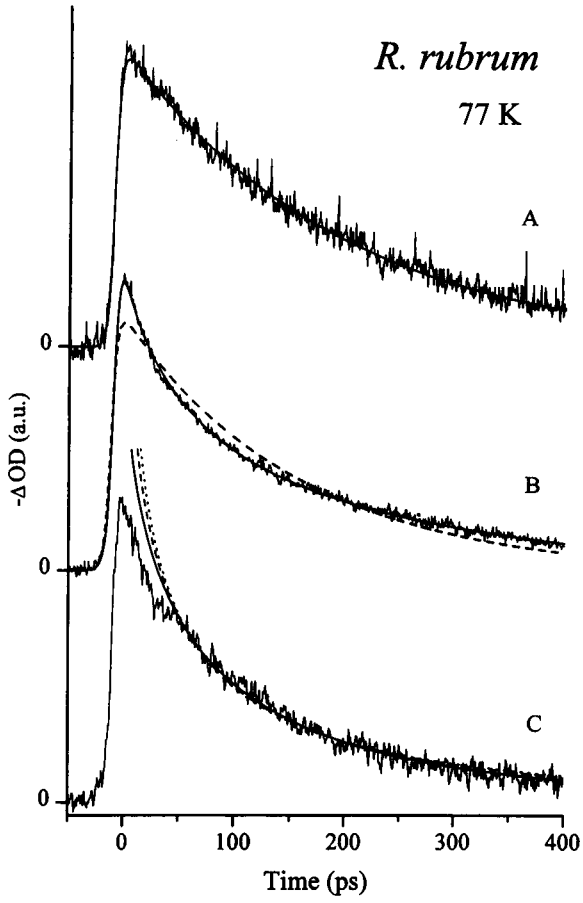

FIGURE 1 Pump-probe kinetics at three different experimental conditions (see Table 1). (*A*) Kinetics without annihilation. The solid line is a single-exponential fit with 210 ps lifetime. (*B*) Kinetics with S-S annihilation. The solid line is constructed from Eq. 6 (see text). The dashed line is a single-exponential fit with 155 ps lifetime. (*C*) Kinetics with S-T annihilation. Lines correspond to the fits in the region from 50 to 350 ps by Eq. 10 with different values of d_s and A. d_s = 2, 1.5, and 1.1 for solid, dashed, and dotted lines, respectively. Corresponding A values are 0.15, 0.26, and 0.46 $ps^{-(ds/(ds+2))}$.

excitations, and consequently as a first approximation we can neglect it in the "high-repetition-rate" kinetics (16 times less S-S annihilation as compared to the "low-repetition-rate" kinetics). At the same time the average excitation intensity used for this measurement was 20 times higher than for the low-intensity curve and about 15 times higher than for the S-S annihilation curve. It can be clearly seen in Fig. 1 that with these conditions, despite the absence of S-S annihilation, the decay was still significantly faster than the low-intensity curve. We assign this shortening of the lifetime to S-T annihilation.

First we take a closer look at the S-S annihilation curve. Eq. 6 can be rewritten as

$$\frac{1}{p(t)} - 1 = \frac{2n_0\gamma_0}{d_s} t^{d_s/2}. \tag{11}$$

Here on the left-hand side we have the reciprocal of the normalized excitation density $p(t) = n(t)/n_0$, which can be directly related to the experimental decay curve. To get $p(t)$, the experimental curve has to be deconvoluted with the apparatus response. The pure deconvolution of experimental data can be performed through the Fourier transform, but this procedure is notoriously sensitive to noise and does not generally give reliable results (Press et al., 1992). Therefore, to construct the approximate pure decay $p(t)$, at first we fitted the experimental curve by a sum of six exponentials convoluted by the apparatus response. To avoid unreasonably short decay components we fixed the shortest lifetime at 5 ps (note that the time resolution of our system is about 2 ps). In that way we were using a reasonable numerical representation of the pure material response, which after normalization gives us $p(t)$. Now we can numerically construct the left-hand side of Eq. 11 and fit it by the corresponding time dependence, which has to be proportional to $t^{d/2}$. In Fig. 2 we have plotted the constructed curve (*solid line*), together with the power law fit (*dashed line*). The expression (Eq. 11) is valid only for the range where $kt < 1$ (k^{-1} = 210 ps), and therefore, we have only fitted the time region up to 50 ps (note that the time scale of Fig. 2 is not linear). Within this time interval the constructed curve has a nearly linear dependence on $t^{d/2}$.

Alternatively, the time-dependent S-S annihilation rate constant $\gamma(t)$ can be directly extracted from Eq. 1 as

$$-n_0\gamma(t) = \frac{1}{p}\left(\frac{1}{p}\frac{dp}{dt} + k\right). \tag{12}$$

Using $p(t)$ as obtained above we can calculate the right-hand side of Eq. 12, which is plotted in Fig. 3 (*solid line*). This curve is fitted by the power law expression according to Eq. 4 (*dashed line*). To construct $\gamma(t)$ we have to perform a number of numerical manipulations with $p(t)$. At longer times this can accumulate large errors. On the other hand, Eq. 4 has a nonphysical singularity if $t \to 0$. Therefore, we have restricted the fitting to the time region from 5 ps to 80 ps.

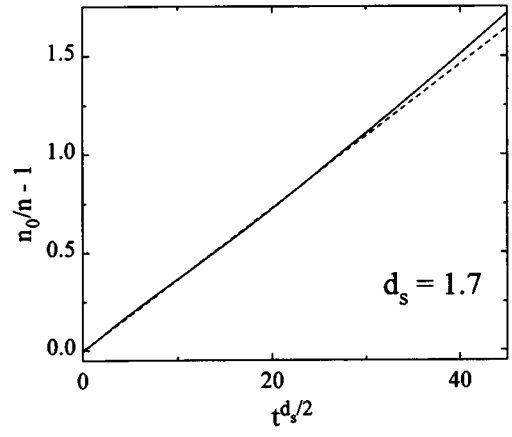

FIGURE 2 Dependence of constructed (see text, Eq. 11) $n_o/n(t) - 1$ on $t^{0.85}$. One can see that the constructed curve (*solid line*) is very close to being linear on this scale, giving d_s = 1.7. The dashed line corresponds to an exact power law dependence $t^{0.85}$.

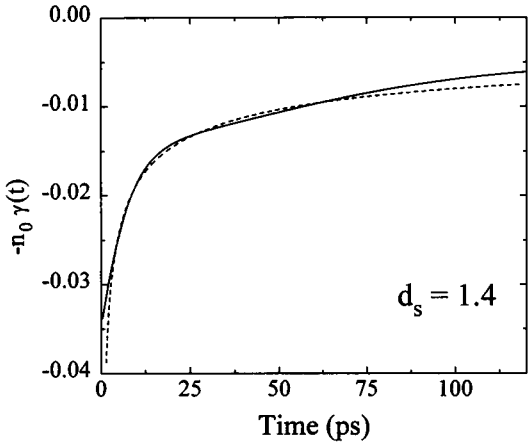

FIGURE 3 Time dependence of the S-S annihilation constant as extracted (see text) from experimental traces (*solid line*), together with the power law fit (*dashed line*), which yields $d_s = 1.4$.

In both schemes of analysis there are two fitting parameters, d_s and $n_0\gamma_0$. We have analyzed a number of independent experimental curves, and for the dimensionality parameter we obtain the average value $d_s = 1.5^{+0.3}_{-0.2}$. For the other parameter we obtained $n_0\gamma_0 = 0.04(\pm 0.005)ps^{-ds/2}$, which with our estimation $n_0 = 0.01$ (one excitation per 100 molecules per pulse) at the given excitation intensity leads to the conclusion that $\gamma_0 = 4\ ps^{-ds/2}$. Using these parameters and Eq. 6 we have reconstructed $n(t)$ and added one exponential component of 210 ps to fit the long time region where Eq. 6 fails. As can be seen from Fig. 1, the reconstructed curve convoluted with the apparatus response (smooth solid line in S-S annihilation) fits the experimental kinetics perfectly.

The experimental curves with S-T annihilation were directly fitted by Eq. 10. As has already been mentioned, this equation is valid for long time asymptotics, and thus we performed the fitting over the time interval from 50 to 350 ps. It is noteworthy that if some singlet-singlet annihilation is present under these conditions, then it would mainly affect the initial part of the decay curves ($t \leq 50$ ps), and not so much the region that we have used for the fitting. We can obtain a very good fit in the selected region by using the d_s obtained from the singlet-singlet annihilation analysis. The fit is plotted as a dashed line in Fig. 1, curve C. However, it turns out that the two fitting parameters in Eq. 10, A and d_s, compensate each other in the above region, and we can get a good fit also with $d_s = 2$ (*solid line*) and $d_s = 1.1$ (*dotted line*). Thus, we can conclude that the value of d_s obtained from the S-S annihilation analysis is consistent with the S-T annihilation data, but we would not have been able to obtain this parameter on the basis of only S-T annihilation data.

DISCUSSION

The theoretical approach we are using does not involve explicitly spectral inhomogeneity, and the question may arise, to what extent are our results biased by this simplification? The picosecond absorption (van Grondelle et al., 1987) and fluorescence (Timpmann et al., 1991) kinetics at low intensities demonstrate that spectral inhomogeneity has quite a small influence on the excitation dynamics at 77 K at time scales longer than a few picoseconds. Consequently, inhomogeneity can be considered to be the perturbation to the homogeneous system. Thus, by investigating the nonlinear annihilation kinetics at times longer than that required to reach a spectrally equilibrated state, i.e., $t \geq 1$ ps, we can use the theoretical basis developed for the homogeneous systems. This assumption is further supported by fluorescence quantum yield measurements at various excitation intensities, which clearly indicate the absence of any excitation wavelength dependence at room temperature and only a small wavelength dependence at 77 K (Deinum et al., 1989). This implies that the mean pairwise hopping time of the excitations through the LHA does not change much with temperature, and this value is still about $\tau_{hop} = 0.5$ (Valkunas et al., 1992; Somsen et al., 1994) at 77 K. Therefore, within the time window starting from a few picoseconds up to hundreds of picoseconds the asymptotic expressions 4 and 6 are applicable.

In our analysis we have used a time-dependent S-S annihilation rate constant. On the other hand, the analysis of room temperature experimental kinetics with 30-ps time resolution suggested that γ is practically time independent. The kinetics calculated with γ = constant were indistinguishable from the kinetics with time-dependent γ (Valkunas et al., 1995). This might be due partly to the limited time resolution used by Valkunas et al. (1995), because the time dependence of γ in our analysis is most pronounced within the time resolution of these measurements (see Fig. 3). Still, the time dependence of γ appears to be more pronounced at 77 K.

The value of the parameter d_s, which we have obtained from analysis of S-S annihilation data, holds also for S-T annihilation. Despite the broad error limits in the latter case, it still gives an important independent support for the S-S annihilation results. The physical meaning of d_s can be related to the spectral dimension, which is due to the fractality of the structure (Bunde and Havlin, 1991). The value of this parameter contains information about the changes in the symmetry and structure of the paths along which the excitation moves. For an ideal two-dimensional LHA array the most straightforward expectation would be $d_s = d = 2$ (nonfractal structure). The value obtained from our analysis is somewhat smaller than that. The LHA of *R. rubrum* is a ring of 32 Bchl *a* molecules surrounding the RC (Karrasch et al., 1995). Earlier annihilation studies have shown that excitation visits several (>10) such units (LHA + RC) before being lost or trapped (Bakker et al., 1983; van Grondelle, 1985). Thus, a number of these elementary rings are interconnected and form a bigger antenna array to which excitation can migrate. If this array has a fractal-like secondary structure then one would expect it to have a spectral dimension of less than 2. However, $d_s < 2$ at 77 K is most

likely due to the spectral inhomogeneity of the LHA. The inhomogeneous distribution of site energies forms a potential surface for the excitation, a random landscape of "valleys" and "ridges." At 77 K the excitation preferably migrates through the "valleys," and consequently the pathway of excitation migration has a fractal-like structure. This interpretation is supported by the lack of time dependence of γ at room temperature (Valkunas et al., 1995). (However, it should be noted that these room temperature experiments were performed with lower time resolution, which implies that some weak and short-lived time dependence of γ, due to a secondary fractal structure of LH1 rings, cannot be entirely excluded.) At room temperature excitation is not any more "forced" to move only through the "valleys" and therefore fully covers the two-dimensional antenna. Consequently, at $d_s = 2$, γ is constant (see Eqs. 4 and 5). Moreover, the value of d_s obtained from our analysis is close to 4/3—the spectral dimension of percolative-like structures (Bunde and Havlin, 1991; Aharony and Stauffer, 1984). One can see that despite the fact that we have not included the spectral inhomogeneity explicitly in our formalism (see previous paragraph), the effect indirectly appears through the spectral dimension d_s.

The characterization of energy migration through the LHA network, extending over the time scale from subpicoseconds to ~100 ps at 77 K, can now be summarized in the following way. Interpigment excitation transfer occurs in the subpicosecond time scale. This equilibrates the excitation within the nearest local minima around the initially excited pigment molecules. On the picosecond time scale the excitation then moves over the whole LHA, making tens or hundreds of hops, and therefore the excitation transfer over larger distances can be considered as a diffusion-like process. Our formalism of singlet-singlet and singlet-triplet annihilation processes is applicable for the longer time scale, and the corresponding kinetics represent the average excitation migration. Analysis of singlet-singlet annihilation kinetics at room temperature is well described by γ = constant (Valkunas et al., 1995), whereas at 77 K γ becomes time dependent. We attribute this time dependence to the spectral inhomogeneity of the LHA, which restricts the possible pathways of the excitation in the antenna to fractal-like structures.

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