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A picosecond-absorption study on bacteriochlorophyll excitation, trapping and primary-charge separation in chromatophores of *Rhodospirillum rubrum*

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Absorbance-difference spectra and kinetics of absorbance changes were measured of chromatophores of *Rhodospirillum rubrum* by means of picosecond-absorption spectroscopy. A 35 ps excitation pulse at 532 nm produced absorbance changes due to the formation and decay of excited states of antenna pigments (Nuijs, A.M., Van Grondelle, R., Joppe, H.L.P., Van Bochove, A.C. and Duysens, L.N.M. (1985) Biochim. Biophys. Acta 810, 94–105), and, when open reaction centers were present, also those due to charge separation and primary electron transport. At low excitation energy density the lifetime of singlet-excited antenna bacteriochlorophyll was 80 ± 10 ps when the reaction centers were initially open and 200–400 ps when the primary electron donor was oxidized. Under the former conditions photooxidation of the primary donor occurred with a time constant of 70 ± 10 ps. Reduction of an electron-acceptor complex in the reaction center, probably involving both bacteriochlorophyll and bacteriopheophytin, was observed. Reoxidation of this acceptor occurred with a time constant of 200–300 ps. When the ubiquinone acceptor was reduced chemically, the primary radical pair decayed by recombination with a time constant of about 4 ns at high flash-energy densities, and of about 10 ns at lower energy densities. This dependence of the lifetime of the radical pair on the flash intensity was explained in terms of quenching processes by carotenoid triplet states in the antenna, and indicated a standard free-energy difference between the radical pair and the singlet-excited state of antenna bacteriochlorophyll of about 160 meV.

Introduction

The photosystem of the purple bacterium *Rhodospirillum rubrum* consists of reaction-center complexes embedded in a light-harvesting antenna, denoted as B880 [1]. Bacteriochlorophyll (BChl) *a* and carotenoid (Car) together serve as the pigments of the B880 complex, and about 50 BChl *a* molecules are present per reaction center. Singlet excitations on antenna BChl, either formed by direct excitation of BChl or by energy transfer from excited Car, are rapidly transferred among the BChl molecules, until trapping by the reaction centers occurs. In the reaction centers a charge separation takes place, followed by further electron-transfer steps that stabilize the separated

Abbreviations: BChl, bacteriochlorophyll; B880, antenna complex with an absorption maximum at 880 nm; Car, carotenoid; P, primary electron donor bacteriochlorophyll; I, intermediary electron acceptor complex; BPh, bacteriopheophytin; P-800, reaction center bacteriochlorophyll molecule absorbing around 800 nm; PMS, *N*-methylphenazoniummethosulfate; RC, reaction center.

charges. So far the charge separation and the early electron-transport steps in purple bacteria have mainly been studied in isolated reaction centers. It was established that bacteriopheophytin (BPh) functions as an intermediary electron acceptor between the primary donor, P, and the traditional primary quinone acceptor Q [2]. The role of a bacteriochlorophyll molecule, P-800, as an even earlier acceptor than BPh is still controversial. Possibly P-800 and BPh interact strongly and together constitute an acceptor complex (see Ref. 3 and references therein.

In intact photosystems the quantum yield of charge separation is high. At low-excitation intensity about 90–95% of the excitations are trapped in the reaction centers [4]; the remaining ones are lost by emission of fluorescence or by radiationless decay processes. The experimentally observed fluorescence lifetime under these conditions is about 60 ps [5]. When the traps are closed (state $P^+Q^{(-)}$) the fluorescence lifetime is lengthened to about 200 ps [5] and the fluorescence yield increases 3-fold [6]. When the excitation density is increased drastically by applying an intense picosecond laser flash, a large decrease is observed in both the mean fluorescence lifetime [7] and the fluorescence yield [6]. These data can be quantitatively interpreted on the basis of competition between trapping, loss and singlet–singlet annihilation [8]. From fluorescence yield measurements on *R. rubrum* it was estimated that the fluorescence lifetime, when all the reaction centers are in the state P Q is about 100 ps, and in the state $P^+Q^{(-)}$ about 300–350 ps [6].

Recently, we reported a picosecond absorbance difference study on chromatophores of *R. rubrum* in which the formation and decay of singlet-excited BChl (BChl*) were detected via spectral changes in the near-infrared [9]. From these experiments, in which the reaction centers were kept in the state $P^+Q^{(-)}$, it was concluded that the average lifetime of BChl* decreased from 200–400 ps at low excitation intensity to 10 ps or less when about 10 excitations are generated per reaction center. In this work we report similar experiments but now for different redox states of the reaction center. At low intensity the lifetime of BChl* decreases from 200–400 ps in the state $P^+Q^{(-)}$ to 80 ps or less in the state P Q. Under the latter conditions the oxidation of P is observed with a time constant of 70 ± 10 ps. The kinetic and spectral data concerning the charge separation are in general agreement with those obtained for isolated reaction centers.

Materials and Methods

R. rubrum was grown anaerobically in a continuous culture as described in Ref. 10. Chromatophores were prepared using a French press and were diluted in a buffer medium containing 250 mM Tricine/5 mM KH_2PO_4/4 mM $MgCl_2$ (pH 8.0). The redox state of the reaction centers was controlled by the addition of either ferricyanide together with continuous back-ground illumination, or of dithionite, or of PMS and ascorbate. All measurements were performed at room temperature.

The picosecond absorbance difference measurements were performed by means of the apparatus described in Ref. 9. A 35 ps mode-locked Nd-YAG laser served as a light source. The frequency-doubled 532 nm output of the laser (maximum energy density, about 3 mJ/cm^2) was used as an excitation pulse. The 1064 nm radiation that remained after frequency-doubling was focused into a cell filled with water, in order to generate a broad wavelength continuum. The 35 ps probe pulse, that was used for measuring the absorbance changes, was obtained from this continuum by means of a monochromator placed before the sample (bandwidth, about 3 nm). Schott KV 550 filters were used to prevent stray excitation light from reaching the photodiodes.

Results

Fig. 1 shows the absorbance difference spectra of chromatophores of *R. rubrum* in the region 550–900 nm measured at 2 ns after the 35 ps excitation pulse for different initial redox conditions of the reaction centers. In the presence of the donor system PMS and ascorbate (open circles) the absorbance changes above 600 nm are due to the formation of the state P^+Q^-. They are similar to those obtained in isolated reaction centers [2]. The bleaching around 870 nm is caused by the disappearance of the Q_y band of P, while the

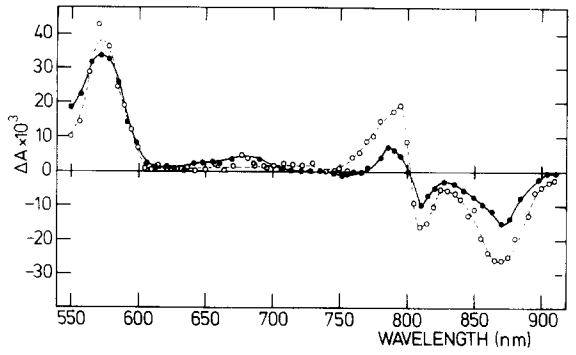

Fig. 1. Absorbance difference spectra of chromatophores of *R. rubrum* at 2 ns after the flash in the presence of 15 μM PMS and 10 mM ascorbate (○) and in the presence of dithionite (●), at a flash excitation energy density of 1.3 mJ/cm^2. The absorbance of the sample was 0.57 at 532 nm in a 2 mm cell.

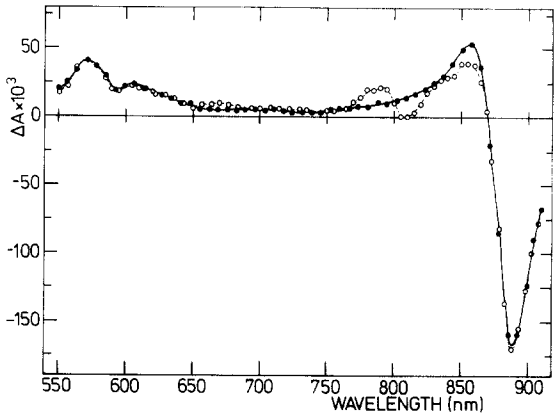

Fig. 2. Absorbance difference spectra of chromatophores of *R. rubrum* with coincident excitation and probe pulses (0 ps) in the presence of 2 mM ferricyanide and continuous background illumination at 528 nm (●) and in the presence of 15 μM PMS and 10 mM ascorbate (○). Further conditions as in Fig. 1.

maximum and minimum at 790 and 810 nm, respectively, reflect the shift of a BChl molecule in the reaction center which absorbs at about 800 nm. The absorbance increase centered at 570 nm can be ascribed to the formation of triplet states of antenna carotenoid molecules (Car^T) [11]. The formation of these Car^T states probably results from a fission of the singlet excited state of Car (Car*), produced by direct excitation of Car at 532 nm [12,13]. Car^T states have been shown to be present within 100 ps after the formation of Car* [9].

In the presence of dithionite (solid circles) to reduce Q chemically, the absorbance changes due to formation of P^+ and Car^T can be observed as well, albeit that the changes related to oxidation of P are smaller in amplitude (see below). In addition, however, an increase in absorbance between 640 and 700 nm is present, together with a small bleaching around 750 nm. These additional changes have also been observed in isolated reaction centers of purple bacteria under similar reducing conditions, and have been ascribed to the reduction of BPh [2]. A comparison of the two spectra of Fig. 1 in the region around 800 nm after normalization at 870 nm shows that the band shift around 800 nm is not symmetric under reducing conditions, in contrast to the band shift in the P^+Q^- spectrum. A similar asymmetry has been observed earlier by Van Bochove et al. [14] and Van Grondelle et al. [15], which could originate from P-800 that forms a complex with BPh and therefore loses oscillator strength upon the reduction of the complex, in a similar way as has been proposed for isolated reaction centers of *Rhodobacter (Rhodopseudomonas) sphaeroides* (see Ref. 3 and references therein). We will denote the complex by I.

Absorbance difference spectra of chromatophores of *R. rubrum* at 0 ps (i.e., with coincident excitation and probe pulses) are shown in Fig. 2. In the presence of ferricyanide and continuous background illumination (solid circles) only absorbance changes due to formation and decay of antenna-excited states are brought about, since P is permanently oxidized under these conditions [9]. The bleaching around 888 nm is related to the disappearance of ground-state absorbance of antenna BChl *a* molecules upon excitation [16]. In a previous publication [9] we have suggested that the presence of a singlet excitation on a BChl *a* molecule causes the bleaching of its Q_y absorption band at 880 nm and induces a blue shift of about six or more neighboring BChl *a* molecules. The combined spectral effects result in a shift of the maximal bleaching from 880 to 888 nm and an increase in absorption between about 800 and 867 nm. The absorbance increase between 550 and 650 nm is predominantly due to formation of Car* and, possibly, of Car^T. The small trough around 590 nm is probably caused by the bleaching of the Q_x band of the antenna BChl *a* [9]. In the pres-

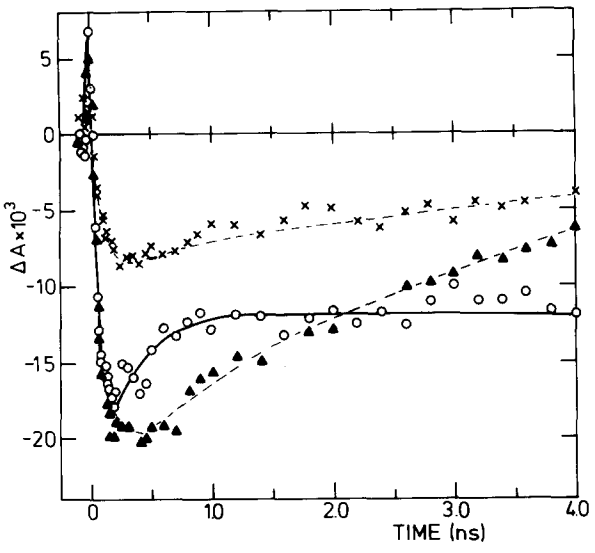

Fig. 3. Kinetics of absorbance changes at 810 nm in chromatophores of *R. rubrum* in the presence of 15 μM PMS and 10 mM ascorbate at an excitation density of 1.1 mJ/cm² (○) and in the presence of dithionite at 1.1 mJ/cm² (▲) and at 0.1 mJ/cm² (×). Further conditions as for Fig. 1.

ence of PMS and ascorbate (Fig. 2, open circles), or dithionite (not shown) similar absorbance changes can be observed. However, the shape of the spectrum between 770 and 820 nm now indicates the electrochromic shift of a reaction center BChl molecule resulting from the oxidation of P. The reduction of I can be deduced from the absorbance increase between 650 and 700 nm. A comparison of the amplitudes of the absorbance changes in the region 790–810 nm of Fig. 1 (open circles) and Fig. 2 (open circles) shows that at 0 ps (i.e., during the flash) in 50–60% of the traps that will ultimately be closed, a radical pair has already been formed. The rate at which the traps are closed, however, depends upon the excitation energy density as will be shown below.

Fig. 3 depicts the kinetics of the absorbance changes at 810 nm for a number of conditions. In the presence of PMS and ascorbate (open circles) a constant bleaching is observed from about 1 ns after the flash onwards. The bleaching arises from the shift of the absorption spectrum of a reaction center BChl molecule (cf. Fig. 1) due to the formation of P^+. At earlier times a short-lived increase in absorbance can be discerned which is probably due to antenna BChl* *a* (cf. Fig. 2). An additional phase is observed after the disappearance of BChl* *a* that decays with a time constant of 200–300 ps to the above-mentioned constant level. During the time that I is reduced an additional bleaching is present around 810 nm [2], and thus the 200–300 ps phase presumably represents the electron transfer from I^- to Q, resulting in a partial recovery of the oscillator strength of the P-800 molecule. Attempts to monitor this step in the electron transfer in the region of absorption of BPh failed as a result of a too small signal-to-noise ratio. In agreement with this assignment, the 200–300 ps phase is lacking in the kinetics with dithionite. Instead, a decay lasting at least 4 ns can be observed after the deactivation of BChl* *a*, which is caused by the recombination of the radical pair P^+I^- [2,14]. The lifetime of the pair increases from about 4 ns at an excitation energy density of 1.1 mJ/cm² (4.4 excitations absorbed per reaction center) (Fig. 3, triangles) to about 10 ns at an energy density of 0.1 mJ/cm² (0.4 excitations absorbed per reaction center) (Fig. 3, crosses). Such a dependence of the lifetime of the radical pair upon the flash intensity has not been observed for isolated reaction centers. A possible explanation for this effect in terms of quenching processes in the antenna will be given in the Discussion.

The kinetics of the absorbance changes at 898 nm are shown in Fig. 4 for various excitation energy densities. At this wavelength a bleaching is observed caused by the formation of BChl* *a* (cf. Fig. 2). The development of the bleaching in all traces is determined by the convolution of the temporal profiles of the excitation and probe pulses. In the presence of ferricyanide (i.e., state P^+Q) (solid circles) the tail in the decay of the bleaching from about 200 ps onwards yields a lifetime of 200–400 ps for BChl* *a*, that has been ascribed to the mono-excitation decay [9], comparable to the 200 ps lifetime of the fluorescence in the state $P^+Q^{(-)}$ [5]. At earlier times contributions of singlet–singlet annihilation to the decay are present. At decreasing excitation-energy density the relative contribution of the annihilation to the total decay diminishes, as is indicated by the longer decay times for top compared to bottom traces (solid circles). For a more complete discussion of these kinetics we refer to Ref. 9. The kinetics monitored in the presence of PMS and

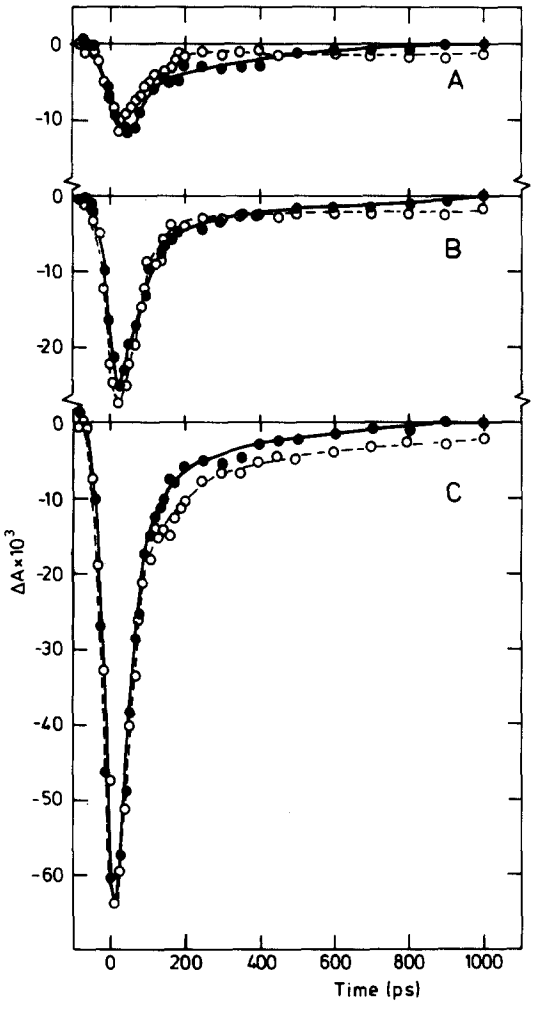

Fig. 4. Kinetics of absorbance changes at 898 nm in chromatophores of *R. rubrum* in the presence of ferricyanide and continuous background illumination (●) and in the presence of 15 μM PMS and 10 mM ascorbate (○), for different excitation densities. (A) 0.031 mJ/cm^2; (B) 0.081mJ/cm^2; (C) 0.34 mJ/cm^2.

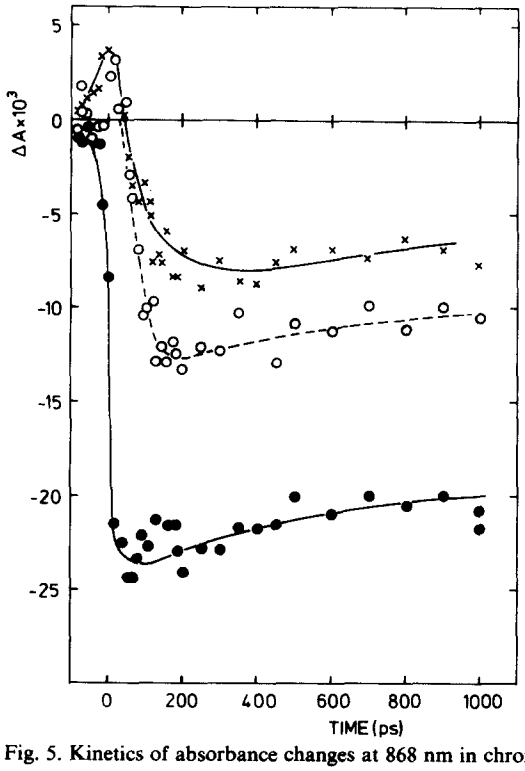

Fig. 5. Kinetics of absorbance changes at 868 nm in chromatophores of *R. rubrum* in the presence of 15 μM PMS and 10 mM ascorbate for different excitation densities. (×): 0.1 mJ/cm^2 (0.4 photons absorbed per RC); (○): 0.23 mJ/cm^2 (0.9 photons per RC); (●):0.8 mJ/cm^2 (3.2 photons per RC).

ascorbate (open circles) differ in some respects. At low excitation energy density (Fig. 6A) the 200–400 ps decay is lacking, and a shorter decay of about 80 ± 10 ps can be observed. Furthermore, from about 200 ps onwards a small remaining bleaching is present due to the formation of P^+ (Fig. 1). We ascribe the 80 ps phase to the mono-excitation decay when the traps are in the state P Q. This lifetime compares reasonably well with the 60 ps decay of the fluorescence as observed by Freiberg et al. for open traps [5]. At higher energy densities (Fig. 6C) the slower 200–400 ps phase reappears. This effect results from the fact that most of the traps are closed during the excitation pulse at those flash energies (see below), which implies that the excitations still present after 200 ps will decay with the traps predominantly in the state $P^+Q^{(-)}$.

The kinetics of the absorbance changes at 868 nm are shown in Fig. 5. At this wavelength the changes due to formation of BChl* *a* have an isobestic point (Fig. 2). The bleaching in Fig. 5 is due to the oxidation of P (Fig. 1). It is observed that the development of the bleaching depends upon the excitation intensity. At relatively low intensity (0.4 photons absorbed per reaction center) the rise-time of the oxidation of P is 70 ± 10 ps. At higher intensities the rise-time becomes shorter and ultimately cannot be resolved due to convolution with the temporal profile of the pulses. The initial small absorbance increase in some of

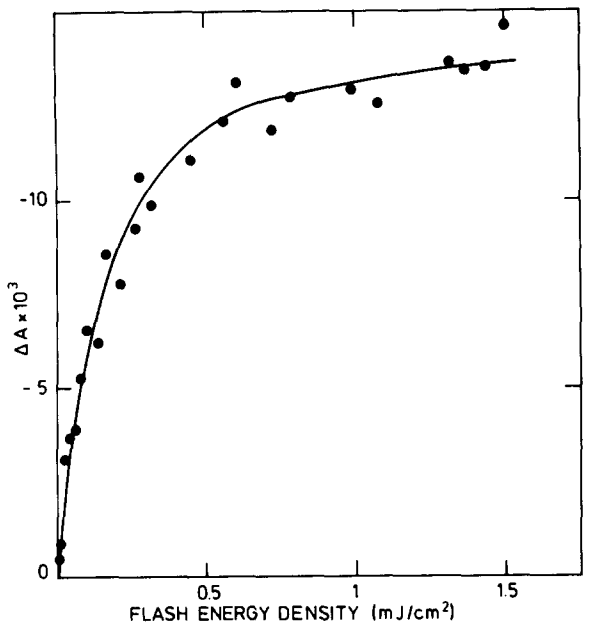

Fig. 6. Absorbance change at 810 nm at 1 ns after the pulse versus the excitation energy density in the presence of 15 μM PMS and 10 mM ascorbate. Further conditions as for Fig. 1.

the traces is presumably due to the excitation of antenna BChl, since the mentioned isobestic point shifts somewhat upon changing the flash density [9]. The low intensity time constant for oxidation of P agrees with the low-intensity lifetime of BChl* *a* which suggests that this lifetime is determined by trapping.

Fig. 6 depicts the saturation behaviour of the formation of P^+ monitored at 810 nm at 1 ns after the pulse. From the amplitude of the maximal bleaching as compared to that under continuous illumination (not shown) it was calculated that 60–70% of the traps can be closed. The value for the maximum amount of P^+ that can be formed after excitation with a picosecond pulse agrees very well with the value of 65% obtained by measurement of the fluorescence yield, and can be explained on the basis of a competition between trapping and annihilation [6].

Discussion

Excitation of chromatophores of *R. rubrum* produces absorbance changes due to formation and decay of excited states of antenna pigments and, in the case of active reaction centers, also those due to the primary charge separation and the ensuing electron transport. In the antenna, direct excitation of Car gives rise to formation and decay of Car* and BChl* *a*, and to formation of Car^T. The absorbance changes caused by these processes have been extensively dealt with in Ref. 9.

The lifetime of BChl* *a* depends upon the state of the traps and upon the number of excitations generated per domain. At low intensities the decay time of BChl* *a* is 80 ± 10 ps in the state P Q and 200–400 ps in the state $P^+Q^{(-)}$. These decay times are in reasonable agreement with the fluorescence lifetimes of 60 ps and 200 ps respectively, obtained by Freiberg et al. [5]. Furthermore, our data suggest that the 80 ps lifetime is determined by the trapping, since the time constant for the oxidation of P was found to be 70 ± 10 ps.

In an experiment similar to ours, Borisov et al. [16] found a decay time of about 60 ps for BChl* *a* and a risetime of about 60 ps for the oxidation of P. However, they claimed that the lifetime of BChl* *a* was independent of the state of the traps [17]. This discrepancy with our results is presumably due to the fact that their kinetics only ranged to 300 ps and to a neglect of the distortion of their kinetics by the convolution with the shape of the pulses. A mono-exponential fit of our first 300 ps of the decay of BChl* *a* (Fig. 4) at a comparable flash-energy density yields an average lifetime of BChl* *a* of 70–100 ps, almost independent of the state of the traps, and would thus lead to a misinterpretation of the data.

Upon increasing the excitation density, the average lifetime of BChl* *a* in the state $P^+Q^{(-)}$ decreases from about 200–400 ps to about 10 ps when about ten excitations are generated per reaction center as a result of efficient singlet–singlet annihilation [9]. An analogous effect of annihilation can be observed when the traps are originally in the state PQ. Due to the efficient competition between annihilation and trapping at high-excitation densities only 60–70% of the traps can be closed (Fig. 6 and Ref. 6).

In the reaction center a charge separation takes place between P and a complex, I, involving both BPh and P-800, analogous to the situation in

isolated reaction centers of *Rb. sphaeroides* [2,3]. The electron transfer from I^- to Q proceeds with a time constant of 200–300 ps, which number agrees well with the 250 ps observed in isolated reaction centers of *R. rubrum* [18]. When Q is chemically reduced the lifetime of I^- is increased and the radical pair decays with a time constant of at least 4 ns, depending on the excitation energy density, in clear contrast to the 10–15 ns reported for isolated reaction centers of *Rb. sphaeroides* [19]. As will be explained below, this reduction of the decay time may be due to the presence of antenna Car^T states in our experiments.

When in chromatophores of *R. rubrum* the acceptor Q is chemically reduced, the emission of luminescence can be observed with a lifetime of 8–11 ns, probably resulting from a recombination of P^+ and I^- [20]. Thus, during the lifetime of the radical pair a certain amount of BChl* *a*, depending on the energy difference between P^+I^- and BChl* *a* is present. In our spectrum of the radical pair (Fig. 1, solid circles) no additional bleaching around 888 nm was observed. Since the amplitude of the absorbance changes upon the creation of one excitation per reaction center is known [9], we can estimate that less than one excitation per 20 reaction centers contributes to the spectrum of Fig. 1. Taking the presence of 0.6 radical pairs per reaction center in Fig. 1, and assuming that one reaction center is in thermal equilibrium with 50 antenna BChl *a* molecules, it is then easily calculated that the standard free-energy difference, ΔG, between P^+I^- and BChl* *a* is at least 160 meV, which compares well with the value of 160 meV derived by Woodbury and Parson from luminescence measurements [21].

We can now estimate the lifetime of the radical pair when it is in thermal equilibrium with the antenna, in which Car^T states act as quenchers of the fluorescence [12]. If $k_1 = 5 \cdot 10^8\ s^{-1}$ [6] and $\bar{k}_t^c = 2.9 \cdot 10^9\ s^{-1}$ [6] are the rate constants for loss of excitations in the antenna and quenching by the closed reaction centers, respectively, then a lifetime of BChl* *a* at low excitation densities is expected of $1/(k_1 + \bar{k}_t^c) \approx 300$ ps, in good agreement with our results. When chromatophores of *R. rubrum* were excited by means of a 532 nm pulse of 1.3 mJ/cm^2, Rademaker et al. observed a decrease in the fluorescence yield of about 30%, which they attributed to the quenching by Car^T states [12]. It thus follows that the rate constant of quenching, $\bar{k}_q$, should be about $0.5\ (k_1 + \bar{k}_t^c) = 1.7 \cdot 10^9\ s^{-1}$. If f_a and f_r are the fractions of excitations that exist as BChl* *a* and P^+I^-, respectively, then, from a value of $\Delta G = 160$ meV and from an antenna size of 50 BChl *a* per reaction center it is calculated that $f_a \approx 0.08$ and $f_r \approx 0.92$. If we further take a lifetime of 12 ns for the radical pair when no luminescence can occur, as in isolated reaction centers, i.e., $k_{P^+I^-} \approx 8.5 \cdot 10^{-7} s^{-1}$ [19], then at low excitation density, when the amount of Car^T is low, a lifetime of the radical pair is expected of $1/(f_a k_1 + f_r k_{P^+I^-}) \approx 9$ ns. At higher excitation densities, when quenching by the Car^T states occurs, the lifetime of P^+I^- can be expected to decrease to $1/\{f_a(k_1 + \bar{k}_q) + f_r k_{P^+I^-}\} \approx 4$ ns. Both values are in good agreement with the observed lifetimes of about 10 ns and 4 ns, respectively.

In conclusion, we may say that the application of sensitive picosecond absorbance difference spectroscopy to chromatophores of *R. rubrum* yields information about antenna excited states, trapping and properties of the primary radical pair in a relatively intact photosystem.

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References

1 Thornber, J.P., Trosper, T.L. and Strouse, C.E. (1978) in The Photosynthetic Bacteria (Clayton, R.K. and Sistrom, W.R., eds.), pp. 113–160, Plenum Press, New York
2 Parson, W.W. and Ke, B. (1982) in Photosynthesis, Volume I: Energy Conversion by Plants and Bacteria (Govindjee, ed.) pp. 331–385, Academic Press, New York
3 Kirmaier, C., Holten, D. and Parson, W.W. (1985) FEBS Lett. 185, 76–82
4 Kingma, H., Duysens, L.N.M. and Van Grondelle, R. (1983), Biochim. Biophys. Acta 725, 434–443
5 Freiberg, A., Godik, V.I. and Timpmann, K. (1984) in Advances in Photosynthesis Research (Sybesma, C., ed.), Vol. I, pp. 45–48, Martinus Nijhoff/Dr. W. Junk Publishers, Dordrecht, The Netherlands

6 Bakker, J.G.C., Van Grondelle, R. and Den Hollander, W.T.F. (1983) Biochim. Biophys. Acta 725, 508–518
7 Campillo, A.J., Hyer, R.C., Monger, T.G., Parson, W.W. and Shapiro, S.L. (1977) Proc. Natl. Acad. Sci. USA 74, 1997–2001
8 Den Hollander, W.T.F., Bakker, J.G.C. and Van Grondelle, R. (1983) Biochim. Biophys. Acta 725, 492–507
9 Nuijs, A.M., Van Grondelle, R., Joppe, H.L.P., Van Bochove, A.C. and Duysens, L.N.M. (1985) Biochim. Biophys. Acta 810, 94–105
10 Kingma, H. (1983) Ph.D. Thesis, State University of Leiden
11 Monger, T.G., Cogdell, R.J. and Parson, W.W. (1976) Biochim. Biophys. Acta 449, 136–153
12 Rademaker, H., Hoff, A.J., Van Grondelle, R. and Duysens, L.N.M. (1980) Biochim. Biophys. Acta 592, 240–257
13 Kingma, H., Van Grondelle, R. and Duysens, L.N.M. (1985) Biochim. Biophys. Acta 808, 383–399
14 Van Bochove, A.C., Van Grondelle, R., Hof, R.M. and Duysens, L.N.M. (1984) in Advances in Photosynthesis Research (Sybesma, C., ed.), Vol. I, pp. 219–222, Martinus Nijhoff/Dr. W. Junk Publishers, Dordrecht, The Netherlands
15 Van Grondelle, R., Romijn, J.C. and Holmes, N.G. (1976) FEBS Lett. 72, 187–192
16 Borisov, A.Yu., Gadonas, R.A., Danielius, R.V., Piskarskas, A.S. and Razjivin, A.P. (1982) FEBS Lett. 138, 25–28
17 Danielius, R.V., Gadonas, R.A., Piskaraskas, A.S., Razjivin, A.P. and Rotomskis, R.I. (1983) in Abstracts of the 6th International Congress on Photosynthesis, Brussels, Vol. I, p. 245
18 Shuvalov, V.A., Klevanik, A.V., Sharkov, A.V., Matveetz, Y.A. and Kryukov, P.G. (1978) FEBS Lett. 91, 135–139
19 Schenck, C.C., Blankenship, R.E. and Parson, W.W. (1982) Biochim. Biophys. Acta 680, 44–59
20 Van Bochove, A.C., Van Grondelle, R. and Duysens, L.N.M. (1981) in Photosynthesis (Akoyunoglou, G., ed.), Vol. III, pp. 989–996, Balaban International Science Services, Philadelphia, PA
21 Woodbury, N.W.T. and Parson, W.W. (1984) Biochim. Biophys. Acta 767, 345–361