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TRANSIENT STATES IN REACTION CENTERS CONTAINING REDUCED BACTERIOPHEOPHYTIN

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Summary

Photosynthetic reaction centers isolated from *Rhodopseudomonas sphaeroides* strain R-26 were excited with non-saturating 7-ps, 600-nm flashes under various conditions, and the resulting absorbance changes were measured. If the quinone electron acceptor (Q) is in the oxidized state, flash excitation generates a transient state (P^F), in which an electron has moved from the primary electron donor (P, a dimer of bacteriochlorophylls) to an acceptor complex involving a special bacteriopheophytin (H) and another bacteriochlorophyll (B). P^F decays in 200 ps as an electron moves from H to Q. If Q and the acceptor complex are reduced photochemically before the excitation, the flash generates a different transient state of P with a high quantum yield. This state decays with a lifetime of 340 ps. There is no indication of electron transfer from P to B under these conditions, but this does not rule out the possibility that B is an intermediate electron carrier between P and H. Measurements of the yield of fluorescence from P under various conditions show that the 340 ps state is not the fluorescent excited singlet state of P. The transient state could be a triplet state, a charge-transfer state of P, or another excited singlet state that is not fluorescent.

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Abbreviations: B, a special molecule of bacteriochlorophyll in the reaction center; BChl, bacteriochlorophyll. BPh, bacteriopheophytin; H, a special molecule of bacteriopheophytin in the reaction center; P, a special dimer of bacteriochlorophylls in the reaction center; Tris, tris(hydroxymethyl)aminomethane.

Introduction

Reaction centers of photosynthetic bacteria contain a special bacteriochlorophyll (BChl) dimer called 'P'. P has a strong absorption band near 870 nm in reaction centers from bacterial species containing BChl *a*, or near 960 nm in those with BChl *b* [1—3]. Excitation of the reaction centers with light causes P to undergo photooxidation to a radical cation (P^+). The electron acceptor appears to involve a complex of at least two molecules . One of these (B) is another BChl; the other (H) is a bacteriopheophytin (BPh) [3—15]. B absorbs near 800 nm in BChl *a*-containing reaction centers, and near 830 nm in BChl *b*-containing reaction centers. H absorbs near 760 nm in the former, and 800 nm in the latter. Recent studies on BChl *a*-containing species [16—18] suggest that B acts as an intermediary electron carrier between P and H. If this is correct, the initial movement of an electron from an excited singlet state of P (P^*) to B evidently is complete within 1—2 ps [16,17,19]. Electron density appears on H with a time constant of approx. 4 ps [16].

Electron transfer from H^- to Q can be blocked if Q is reduced before the excitation, or extracted from the reaction centers. Under these conditions, the initial photooxidation of P still occurs, generating a transient radical-pair state (state P^F) that decays by reverse electron transfer in approx. 10 ns [13,20]. Flash-induced absorbance changes measured in such reaction centers suggest that P^F is a mixture of two states, $^1[P^+B^-]$ and $^1[P^+H^-]$ [18]. The relative populations of the two depend on the temperature, and on the electronic charge on Q. In the presence of Q^-, $^1[P^+H^-]$ appears to lie approx. 0.025 eV below $^1[P^+B^-]$ in energy [18].

The electron acceptor complex can be trapped in a reduced state if reaction centers are illuminated continuously in the presence of a reduced cytochrome, after electron transfer to Q has been blocked by reduction of Q [7—15]. EPR and ENDOR studies of the reduced complex suggest that most of the added electron density resides on one molecule, which is presumably H [12,14,22]. One might expect that flash excitation under these conditions could still cause electron transfer from P^* to B, generating the state $P^+B^-H^-$. Attempts to detect the formation of this state have been described by two groups of investigators [13, 21], both of whom studied reaction centers from the BChl *b*-containing organism *Rhodopseudomonas viridis*. The results were negative. There was no indication of a transient bleaching at 830 nm, which should occur if B undergoes reduction, or of a transient absorbance increase at 1310 nm, which should occur if P undergoes oxidation. These results were interpreted as indicating that B does not act as an intermediate electron carrier between P^* and H. But this is not a necessary conclusion. If the interaction between B and H is strong (as it probably is, in view of the fact that reducing the electron acceptor complex perturbs the absorption spectra of both molecules severely [15]), reduction of H could prevent electron transfer from P^* to B.

When *Rps. viridis* reaction centers were excited after the reduction of the acceptor complex, Holten et al. [13] did observe the formation of a transient state that decayed with a lifetime of approx. 20 ps. The absorbance changes that accompanied the formation of the transient state suggested that the state was P^*. The decay of the transient state was unexpectedly rapid, considering that

the photooxidation of P appeared to have been blocked, but this was attributed to reactions between P^* and the reduced acceptor. In the present work, we performed similar studies on reaction centers from the BChl *a*-containing species *Rps. sphaeroides*. The near-infrared absorption band of BPh is better resolved from other components in these than it is in reaction centers from *Rps. viridis*, and more is known about the various states that can result from flash excitation. We found that a transient state of P is formed if reaction centers are excited after the reduction of the acceptor complex, but that the transient state is not the fluorescent excited singlet state P^*.

Materials and Methods

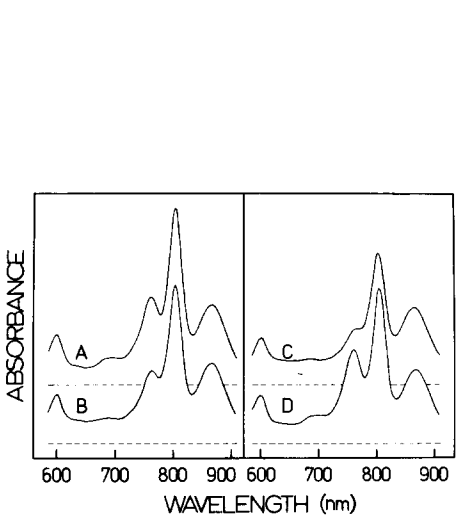
Reaction centers from *Rps. sphaeroides* strain R-26 were prepared by treating chromatophores with lauryldimethylamine oxide, followed by chromatography on DEAE-Sephacel, as described elsewhere [36], and were suspended in 50 mM Tris-HCl, pH 8.0, 0.05% Triton X-100. Concentrations were determined using an extinction coefficient of 288 $mM^{-1} \cdot cm^{-1}$ at 802 nm [22]. Low redox potentials (below −450 mV with respect to H^+/H_2) were obtained by the addition of $Na_2S_2O_4$ (approx. 1 mg/ml). To reduce BPh, reaction centers containing $Na_2S_2O_4$ and 0.2 mM horse heart cytochrome *c* were illuminated with 80 $mW \cdot cm^{-2}$ near-infrared light from a tungsten lamp filtered by a Corning 2600 glass filter and 5 cm of water. The reaction centers were held in a magnetically stirred, stoppered, water-jacketed fluorescence cell, with a 1×0.4 cm sample chamber. The water in the jacket was kept at approx. 20°C. The same cell was used for all of the measurements.

Picosecond absorbance measurements were performed as described previously [19]. The 7-ps, 600-nm excitation pulses were produced by stimulated Raman scattering from 530-nm pulses in a 5 cm cell of perdeuterocyclohexane. They had an energy of approx. 0.2 mJ, and were focused to a 1.4 mm diameter spot at the center of the sample. The excitation and measuring beams intersected at an angle of 10°. The excitation flashes were subsaturating for the photooxidation of P, causing approx. 55% of the calculated maximum bleaching at 870 nm. Because of the off-axis excitation geometry and the finite absorbance of the reaction centers at 600 nm ($A_{600} \approx 0.3$), the samples were not excited uniformly, and the extent of the photooxidation may have been greater than 55% in the central part of the cuvette. The continuous infrared illumination was turned off for a few seconds at the time of each measurement.

For fluorescence measurements, reaction center suspensions were excited with weak, 20-μs, 600-nm flashes from a Xe lamp with an interference filter and complementary glass filters. Fluorescence was detected at 90° through a monochromator and glass filters, using a cooled S-1 photomultiplier (RCA 7102). Signals from the photomultiplier were amplified, digitized with a Tektronix 7912 transient digitizer, and accumulated in computer memory.

Results

Fig. 1 shows absorption spectra of *Rps. sphaeroides* R-26 reaction centers in the presence of cytochrome *c* after the addition of $Na_2S_2O_4$ to reduce Q (curve



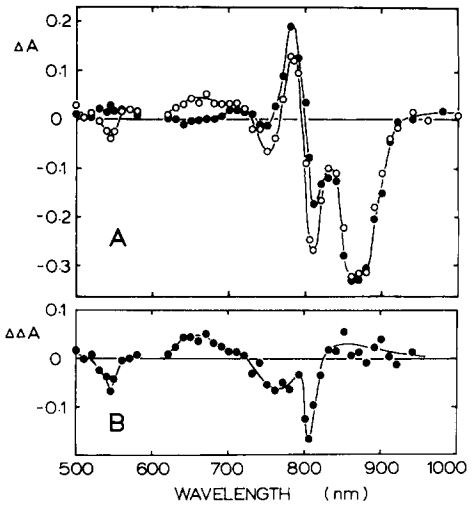


Fig. 1. Absorption spectra of 4.2 μM reaction centers in 50 mM Tris, pH 8, 0.05% Triton X-100, 0.2 mM cytochrome *c*, and 1 mg/ml $Na_2S_2O_4$ (1 cm path), with: A, no illumination; B, 25 min illumination; C, 150 min illumination, and D, 150 min illumination followed by aerobic relaxation in darkness for 6 h. Reference cuvette contained 50 mM Tris, pH 8, 0.05% Triton X-100, 0.2 mM cytochrome *c*, 1 mg/ml $Na_2S_2O_4$. The absorbance at 802 nm in spectrum (A) is 1.2.

Fig. 2. Spectra of absorbance changes caused by flash excitation of 5.2 μM reaction centers at moderate redox potential in 50 mM Tris, pH 8, 0.05% Triton X-100 (1 cm path). Panel A: ○, measurements made 20 ps after the excitation pulse; ●, measurements made 3 ns after the excitation pulse. Each data point is an average of 3—6 measurements. Panel B shows the difference between the filled and open circles of panel A.

A), and after continuous illumination for 25 min (curve B). Illumination at low potential for relatively short periods causes a bleaching of the 760 nm absorption band due to BPh, a partial bleaching of the 800 nm band due to BChl and a broad absorbance increase between 620 and 730 nm. The absorption band that remains near 800 nm is shifted to slightly shorter wavelengths. These absorbance changes are similar to ones that have been described by Okamura et al. [14]. Prolonged illumination results in more extensive bleaching at 760 and 800 nm (Fig. 1, curve C). The extensive bleaching at 760 nm suggests that both of the two BPhs [22] of the reaction centers can be reduced; similar observations have been made in *Rps. viridis* [3]. The absorbance changes caused by the prolonged illumination were almost fully reversible, if the sample was opened to air and allowed to stand several hours in the dark (Fig. 1, curve D). The absorbance at 760 nm returned to a value slightly above its initial level, whereas that at 800 nm remained slightly decreased; a small amount of the bacteriochlorophyll that absorbs at 800 nm may have been converted to BPh.

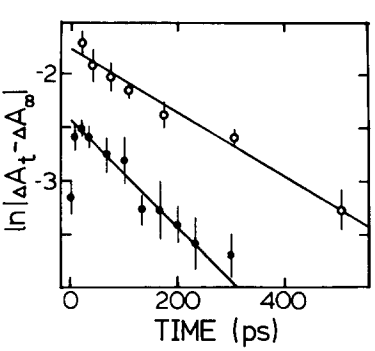
Fig. 2A shows spectra of the absorbance changes that result from the excitation of reaction centers at moderate potential (approx. +200 mV with respect to H^+/H_2) with non-saturating, 7-ps flashes at 600 nm. The spectrum measured at 20 ps after excitation (open circles) reflects the formation of state P^F; that measured at 3 ns (filled circles) is due to P^+Q^- [4,5,13]. Fig. 2B shows the dif-

ference between the measurements at 20 ps and 3 ns; this probably reflects mainly absorbance changes with the transient reduction of B and H. The reduction causes absorbance decreases at 545, 760 and 800 nm, and an absorbance increase in the 620—730 nm region. The difference spectrum is very similar to the difference between curves A and B in Fig. 1.

In agreement with previous work [4,5], the absorbance changes associated with the transient reduction of the acceptor complex decay with a lifetime of approx. 200 ps, as an electron moves on to Q. The filled circles in Fig. 3 show a measurement of kinetics at 802 nm; the decay time is 200 ± 35 ps. There is no indication of the 30 ps decay component that has been seen previously at 802 nm [4] (and at 830 nm in *Rps. viridis* [13]), probably because the excitation flashes used in the present work were relatively weak. This agrees with evidence presented elsewhere [19] that the 30 ps component develops only if one excites the reaction centers with more than one photon.

A different transient state can be detected when *Rps. sphaeroides* reaction centers are excited with non-saturating flashes, after reduction of the acceptor complex by continuous illumination in the presence of $Na_2S_2O_4$ and cytochrome *c*. Fig. 4 shows spectra of the absorbance changes measured 26 ps after excitation under these conditions. Their features are a bleaching of the 870 nm absorption band of P, and small absorbance increases between 620 and 780 nm. In agreement with the previous work in *Rps. viridis* [13,21], there is no indication of a sharp bleaching at 800 nm, which one might expect to see if the excitation generated $P^+B^-H^-$.

The measurements shown in Fig. 4 involved samples that were illuminated



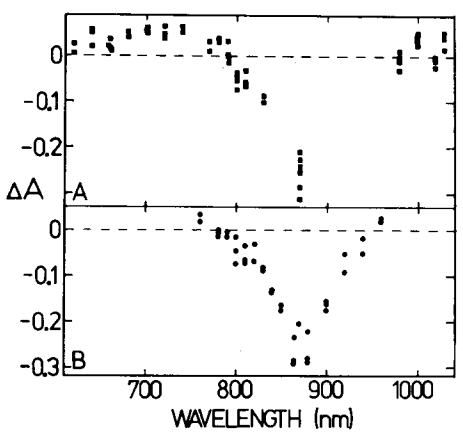


Fig. 3. Decay kinetics of absorbance changes measured with subsaturating laser flashes. •, absorbance changes measured at 802 nm in reaction centers at moderate potential. ○, absorbance changes measured at 865 nm after illumination at low potential under conditions similar to those of Fig. 1C. Each point is an average of 4—6 measurements. The error bars show the S.D.

Fig. 4. Spectra of absorbance changes measured 26 ps after flash excitation of reaction centers, following illumination at low potential. The extent of reduction was similar to that shown in Fig. 1C. Each point represents one laser shot. The panels show two independent sets of measurements; the reaction center concentration was 6.4 μM in panel A and 4.3 μM in panel B (1 cm path).

for periods on the order of 1 h. The second BPh may be partially reduced along with H under these conditions (Fig. 1C). This raises the question of whether both BPhs participate in the normal electron transfer reactions. To explore this point, we measured the flash-induced absorbance changes at 750 and 760 nm in reaction centers that were illuminated for relatively short periods. The measurements were made 17 ps after the excitations. Before the illumination, flash excitation caused a bleaching in the 760 nm absorption band, which we take to reflect the reduction of H (Fig. 2A). After illumination for 5 min, excitation caused a small absorbance increase similar to that shown in Fig. 4. There was thus no indication that the second BPh could be reduced efficiently by flash excitation, following the reduction of H. The reduction seen during prolonged illumination probably occurs with a very low quantum yield.

The decay kinetics of the transient state seen in reaction centers that have BPh reduced are shown with empty circles in Fig. 3. The bleaching at 865 nm decays with a lifetime of 340 ± 35 ps. This is considerably longer than the 20 ps lifetime seen in *Rps. viridis* after excitation with saturating flashes at 530 nm [13]. Because it was difficult to maintain the reaction centers with only one BPh reduced for the long time that was required to complete a set of measurements with the picosecond apparatus, we were not able to determine whether the decay kinetics of the transient state depend on the redox state of the second BPh.

The quantum yield of the transient state appears to be relatively high, because the bleaching at 870 nm caused by weak flashes is similar to the bleaching accompanying the photooxidation of P, when unreduced reaction centers are excited with the same flashes (compare Figs. 2 and 4). The photooxidation occurs with a quantum yield of essentially 1.0 in unreduced reaction centers [23], and full photooxidation causes the 870 nm band to bleach by 88% [24].

To investigate the identity of the transient state, we measured the yields of fluorescence from reaction centers before and after the reduction of Q and the acceptor complex. The excited singlet state that is generated by excitation of reaction centers in their long-wavelengths (870 nm) absorption band fluoresces at 920 nm [24]. The 340 ps lifetime of the transient state seen in reduced reaction centers is at least 50-times longer than the lifetime of the fluorescent state in reaction centers that do not have BPh or Q reduced, and probably more than 100-times longer. The lifetime of the excited state in unreduced reaction centers cannot be greater than 4 ps, because an electron appears on H in this time [19]. As discussed above, it is likely that P^* is quenched by electron transfer to B in less than 2 ps. The population of the fluorescent state could, however, remain significant through the additional 4 ps that is required for the movement of the electron to H. From the quantum yield of fluorescence, Zankel et al. [24] have calculated that the excited state has an effective fluorescence lifetime of approx. 7 ps. An increase in the lifetime of the fluorescent state should result in a comparable increase in the yield of fluorescence, because the natural radiative lifetime probably does not change significantly. The reduction of the acceptor complex has little or no effect on the absorption sepctrum of reaction centers in the 870 nm region (Fig. 1). It also has no significant effect on the fluorescence emission spectrum near 920 nm (Fig. 5).

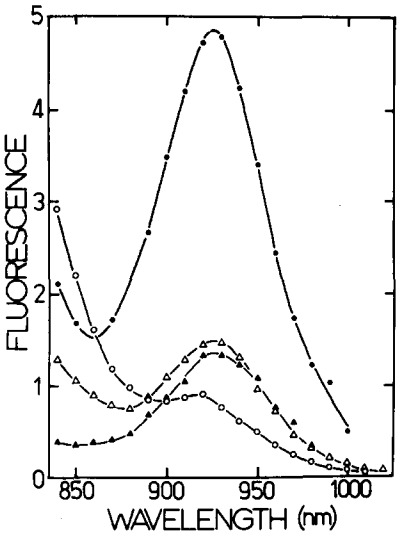


Fig. 5. Fluorescence emission spectra for 6.6 μM reaction centers, uncorrected for the spectral response of the photomultiplier and the monochromator. △, fluorescence from reaction centers under conditions similar to those of Fig. 1A (low potential, no illumination); ▲, reaction centers under conditions intermediate between those of Fig. 1A and B (illumination to give reduction of less than one BPh per reaction center); ●, reaction centers that were more completely reduced than those of Fig. 1C; ○, reaction centers at moderate redox potential. Monochromator bandpass 20 nm; vertical scale arbitrary. During the course of the fluorescence measurements for the experiments with filled symbols there was a small time-dependent increase in apparent fluorescence at 920 nm (less than 5%). This correlated with a slight increase in turbidity of the sample and was corrected for by linear interpolation.

TABLE I

FLUORESCENCE YIELDS FROM REACTION CENTERS UNDER VARIOUS CONDITIONS

Conditions	Relative fluorescence yield (920 nm)	
	Measured *	Corrected **
Moderate redox potential	1.00	1.00
Low redox potential (Q reduced), no illumination	2.27	3.02
Low potential		
5 min illumination	2 04	2.65
25 min illumination	1.70	2.11
90 min illumination	2.65	3.62
150 min illumination	3.43	4.86
150 min illumination then 6 h darkness, aerobic	2.68	3.67
High potential (P oxidized with 1 mM $K_3Fe(CN)_6$)	0.37	0

* This is from the same experiment that gave the data shown in Fig. 1; it is a different series of measurements from those shown in Fig. 5. Yields are expressed relative to the yield at moderate redox potential.

** Fluorescence measured at high potential subtracted.

Table I and Fig. 5 present measurements of the yields of fluorescence under various conditions. As is well known [24], lowering the redox potential so as to reduce Q causes the fluorescence at 920 nm to increase approx. 2-fold. The additional fluorescence is probably largely delayed fluorescence that results from the regeneration of P^* from P^F [8,25—27]. Illumination for several minutes at low redox potentials causes the fluorescence at 920 nm to decrease. Part of the initial decrease is due to an emission component that peaks at shorter wavelengths (Fig. 5); the origin of this component is unclear, but it could involve trace amounts of free BPh, or BPh in partially denatured reaction centers. Continued illumination causes a decrease in the emission due to P^*, probably because the reduction of the acceptor complex blocks the initial formation of P^F [8], and new decay routes for the excited state become important. After 25 min of illumination, the fluorescence at 920 nm is approx. 25% less than that measured before the illumination, or approx. 1.7-times higher than that measured at moderate redox potentials (Table I). The fluorescence increases again, if the illumination is continued for periods of several hours. After 90 min the fluorescence is approx. 2.7-times higher than that measured at moderate redox potentials. If one subtracts from both measurements the fluorescence that remains after oxidation of P with $K_3Fe(CN)_6$, the calculated fluorescence yield after 90 min of illumination is apprcx. 3.6-times the yield at moderate potential (Table I). This increase is still much smaller than the 50-100-fold increase that would be expected if the 340 ps state were the fluorescent excited singlet state. The increase in fluorescence after prolonged illumination is only partially reversible and may reflect secondary changes such as pheophytinization in the reaction centers.

Discussion

The present work agrees with the previous studies of *Rps. viridis* [13,21] that a transient $P^+B^-H^-$ state is not detectable if reaction centers are excited after the reduction of the acceptor complex. If such a state does form, it must decay prior to the time when the spectrum in Fig. 4 was measured, which was at 26 ps after the excitation. We emphasize, however, that these observations do not argue forcefully against the idea that B acts as an intermediate electron carrier between P and H. The reduction of H could prevent the formation of $P^+B^-H^-$. In addition, B could have been partially reduced during the prolonged illumination that we used in the picosecond experiments. (This reservation does not apply to the experiments of Netzel et al. [21], who used conditions that insured that only one electron had been added to the acceptor complex.)

The transient state that forms on excitation of reduced reaction centers must involve P, because it is characterized by a bleaching of the main absorption band of P at 870 nm. The transient state apparently is not the fluorescent excited singlet state of P, because its 340 ps lifetime is inconsistent with the finding that the fluorescence yield increases by only a factor of approx. 4 during prolonged illumination, and actually decreases when H is first reduced. This point could not be established in the previous work on *Rps. viridis*, because the transient state had a much shorter lifetime in that species.

It is possible that P has two excited singlet states, with very different radia-

tive rate constants. Pellin et al. [28] recently have obtained evidence for the existence of two such states in covalently linked dimers of pyrochlorophyll in vitro. The transient state seen in the reduced reaction centers could be a non-fluorescent excited singlet state that lies below the fluorescent state and is formed from it. The idea that P has a singlet exciton state at lower energy than the 920 nm fluorescent state would be inconsistent with recent suggestions [29—33] that the 870 nm absorption band and the 920 nm emission are due to the lower of the two singlet exciton states of P, and that the higher energy absorption band is near 800 nm. However, our observation that the formation of the 340 ps transient state causes little or no bleaching at 800 nm argues that P does not have an absorption band in this region. The conversion of P to a triplet state (^{3}P) also does not cause a bleaching in the 800 nm region; it causes a small absorbance increase [18].

Another possibility is that the transient state is a charge-transfer state of P, in which an electron has moved from one BChl of the dimer to the other. This interpretation is potentially attractive, because a singlet charge-transfer state is a plausible intermediate in the electron transfer reaction between P* and B [34]. Coulombic interactions with nearby charged or dipolar amino acid side chains could stabilize the charge-transfer state and could promote unidirectional electron transfer from P to B [34].

The quantum yield of the transient state appears to be relatively high. This suggests that the state could be an intermediate in the photochemical electron transfer reactions in unreduced reaction centers. On the other hand, the decrease in fluorescence suggests that the reduction of the acceptor complex opens up a new decay path for P*, or for a state that is in equilibrium with P*. The transient state thus could be a side product that forms only when the normal electron transfer sequence is blocked.

Perhaps the most likely side product to form under these conditions is a triplet state. The decay of P^F in reaction centers that have Q, but not H, reduced gives rise to a triplet state (state P^R) in low quantum yield [20]. P^R appears to be a mixture of a locally excited triplet state of P (^{3}P) and a triplet charge-transfer state ($^3[P^+B^-]$) [18]. Recent studies of the effects of magnetic fields on the decay kinetics of P^F and the quantum yield of P^R have suggested the existence of still other triplet states, in addition to these two [36]. The spectrum of the absorbance changes that accompany the formation of P^R at 293 K is very similar to the spectrum associated with the transient state (Fig. 4), except that it includes a more sharply defined (but still relatively small) bleaching at 800 nm [18,20]. The bleaching at 800 nm probably reflects the formation of $^3[P^+B^-]$. The 340 ps lifetime of the transient state is much shorter than the ordinary lifetime of P^R, which is approx. 10 μs at 293 K, but this could be explained by spin exchange interactions with the odd electron on H^- or B^-. The same type of interactions could account for the formation of a triplet state in significant quantum yield during the very short lifetime of P*. Similar processes have been described for metalloporphyrins in vitro [35].

Additional work will be needed to determine which of these interpretations is correct.

Acknowledgements

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References

1 Parson, W.W. and Cogdell, R.J. (1975) Biochim. Biophys. Acta 416, 105—149
2 Feher, G. and Okamura, M.Y. (1978) in The Photosynthetic Bacteria (Clayton, R.K. and Sistrom, W.R., eds.), pp. 349—386, Plenum Press, New York
3 Thornber, J.P., Dutton, P.L., Fajer, J., Forman, A., Holten, D., Olsen, J.M., Parson, W.W., Prince, R.C., Tiede, D.M. and Windsor, M.W. (1978) in Proc. 4th Int. Congr. Photosynth. (Hall, D.O., Coombs, J. and Goodwin, T.W., eds.), pp. 55—70, Spottiswoode Ballantyne Press, London.
4 Rockley, M.G., Windsor, M.W., Cogdell, R.J. and Parson, W.W. (1975) Proc. Natl. Acad. Sci. U.S.A. 72, 2251—2255
5 Kaufman, K.J., Dutton, P.L., Netzel, T.L., Leigh, J.S. and Rentzepis, P.M. (1975) Science 188, 1301—1304
6 Fajer, J., Brune, D.C., Davis, M.S., Forman, A. and Spaulding, L.D. (1975) Proc. Natl. Acad. Sci. U.S.A. 72, 4956—4960
7 Tiede, D.M., Prince, R.C. and Dutton, P.L. (1976) Biochim. Biophys. Acta 449, 447—469
8 Shuvalov, V.A. and Klimov, V.V. (1976) Biochim. Biophys. Acta 440, 587—599
9 Shuvalov, V.A., Krakhmaleva, I.N. and Klimov, V.V. (1976) Biochim Biophys. Acta 449, 597—601
10 Van Grondelle, R., Romijn, J.C. and Holmes, N.G. (1976) FEBS Lett. 72, 187—192
11 Dutton, P.L., Prince, R.C., Tiede, D.M., Petty, K.M., Kaufmann, K.J., Netzel, T.L. and Rentzepis, P.M. (1977) Brookhaven Symp. Biol. 28, 213—237
12 Davis, M.S., Forman, A., Hanson, L.K., Thornber, J.P. and Fajer, J. (1979) J. Phys. Chem. 83, 3325—3332
13 Holten, D., Windsor, M.W., Parson, W.W. and Thornber, J.P. (1978) Biochim. Biophys. Acta 501, 112—126
14 Okamura, M.Y., Isaacson, R.A. and Feher, G. (1979) Biochim. Biophys. Acta 546, 394—417
15 Shuvalov, V.A. and Asadov, A.A. (1979) Biochim. Biophys. Acta 545, 296—308
16 Shuvalov, V.A., Klevanik, A.V., Sharkov, A.V., Matveetz, T.A. and Krukov, P.G. (1978) FEBS Lett. 91, 135—139
17 Akhmanov, S.A., Borisov, A. Yu., Danielius, R.V., Gadonas, R.A., Kozlowski, V.S., Piskarkas, A.S., Razjivin, A.P. and Shuvalov, V.A. (1980) FEBS Lett. 114, 149—152
18 Shuvalov, V.A. and Parson, W.W. (1981) Proc. Natl. Acad. Sci. U.S.A., in the press.
19 Holten, D., Hoganson, C., Windsor, M.W., Schenk, C.C., Parson, W.W., Migus, A., Fork, R.L. and Shank, C.V. (1980) Biochim. Biophys. Acta 592, 461—477
20 Parson, W.W., Clayton, R.K. and Cogdell, R.J. (1975) Biochim. Biophys. Acta 387, 268—278
21 Netzel, T.L., Rentzepis, P.M., Tiede, D.M., Prince, R.C. and Dutton, P.L. (1977) Biochim. Biophys. Acta 460, 467—478
22 Straley, S.C., Parson, W.W., Mauzerall, D.C. and Clayton, R.K. (1973) Biochim. Biophys. Acta 430, 83—93
23 Wraight, C.A. and Clayton, R.K. (1974) Biochim. Biophys. Acta 333, 246—260
24 Zankel, K.L., Reed, D.W. and Clayton, R.K. (1968) Proc. Natl. Acad. Sci. U.S.A. 61, 1243—1249
25 Holmes, N.G., van Grondelle, R., Hoff, A.J. and Duysens, L.N.M. (1976) FEBS Lett. 70, 96—101
26 Van Grondelle, R., Holmes, N.G., Rademaker, H. and Duysens, L.N.M. (1978) Biochim. Biophys. Acta 503, 10—25
27 Godik, V.I. and Borisov, A. Yu. (1979) Biochim. Biophys. Acta 548, 296—308
28 Pellin, M.J., Wasielewski, M.R. and Kaufmann, K.J. (1980) J. Am. Chem. Soc. 102, 1868—1873
29 Vermeglio, A. and Clayton, R.K. (1976) Biochim. Biophys. Acta 449, 500—515
30 Shuvalov, V.A., Asadov, A.A. and Krakhmaleva, I.N. (1977) FEBS Lett. 76, 240—245
31 Vermeglio, A., Breton, J., Paillotin, G. and Cogdell, R.J. (1978) Biochim. Biophys. Acta 501, 514—530
32 Paillotin, G., Vermeglio, A. and Breton, J. (1979) Biochim. Biophys. Acta 545, 249—264
33 Raferty, C. and Clayton, R.K. (1978) Biochim. Biophys. Acta 502, 51—60
34 Warshel, A. (1980) Proc. Natl. Acad. Sci. U.S.A. 77, 3105—3109
35 Ake, R.L. and Gouterman, M. (1969) Theor. Chim. Acta (Berl.) 15, 20—42
36 Schenk, C.C., Blankenship, R.E. and Parson, W.W. (1981) Biochim. Biophys. Acta, in the press