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Charge recombination at low temperature in photosynthetic bacteria reaction centers: evidence for two conformational states

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The kinetics of the decay of state $P^+Q_A^-$ at low temperature for bacterial reaction centers present a complex wavelength dependence around the crossing points of the light-induced difference spectrum (approx. 801 and approx. 757 nm). This phenomenon is observed for chromatophores of *Rhodospirillum rubrum* G9 and *Rhodobacter sphaeroides* R26 or 241 as well as for isolated reaction centers of the later species. For untreated reaction centers or chromatophores, the kinetics of the absorbance changes measured at wavelengths slightly above 801 nm are about three times faster than the ones measured below 801 nm. These results are taken as evidence of two conformational states for the reaction centers. Each of these two states has a characteristic decay rate, 11.5 and 36.5 ms respectively, in the case of R26 reaction centers, and a slightly different light-induced spectrum. These two states are in the ratio 60/40, the more abundant being the faster one. They are not influenced by the pH, the presence of the secondary acceptor or the removal of the accessory bacteriochlorophyll molecule of the non photoactive branch. These two conformational states are still present in LM particles, although the kinetics of their charge recombination process are slowed down by a factor of 2.5. Preillumination of reaction centers at room temperature, which lengthens the charges recombination process at low temperature (Kleinfeld, D., Okamura, M.Y. and Feher, G. (1984) Biochemistry 23, 5780–5786) affects only the kinetics of the slow decaying state. This light pretreatment leads also to a ratio of 0.5 between the fast and the slow decaying states, compared to 1.5 for dark-adapted reaction centers. The non-exponential decay of the $P^+Q_A^-$ state measured in the long-wavelength band at low temperature is explained by the superposition of the decays of these two conformational states of the reaction center.

Introduction

The primary photochemical reaction in photosynthesis occurs in a pigment-protein complex called reaction center. In purple photosynthetic bacteria, this reaction center contains four molecules of bacteriochlorophyll, two molecules of bacteriopheophytin, one or two molecules of quinones and one non-heme iron (Fe^{2+}) (see for a review Ref. 1). Upon light absorption, a bacteriochlorophyll dimer (P) is raised to an excited state (P*). In a few picoseconds, electron transfer from this excited state to one of the BPheo molecules generates a transient radical-pair state (P^+BPheo^-) [2–4]. An electron then migrates

Abbreviations: RC, reaction center; TMPD, *N,N,N',N'*-tetramethyl-*p*-phenylenediamine dihydrochloride; BChl, bacteriochlorophyll; BPheo, bacteriopheophytin.

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from the reduced BPheo to one of the quinone molecule (Q_A) in about 200 ps [5–8]. This process is remarkable by, at least, two of its properties. Firstly, the rate of the back reaction of each step is several orders of magnitude slower than the forward reaction, resulting in a quantum yield equal to 1 [9]. Secondly, the reactions exhibit no or only a weak temperature dependence with an unusual negative activation energy [10,11]. This is, for example, the case for the recombination of charges between the photooxidized primary donor (P^+) and the reduced acceptor (Q_A^-). This process is characterized by a very slow life time of $t_{1/2} = 6 \cdot 10^{-2}$ s at room temperature [12] compared to a value of $t_{1/2} = 200 \cdot 10^{-12}$ s for the forward reaction [5–8]. The rate of the back reaction is independent of temperature in the range 1.5–80 K [13], and decreases by a factor of 1.5–5, depending on the species, when increasing the temperature from 100 to 300 K [14]. This process was assigned to an activationless electron transfer [15,16]. Another peculiarity of the charge recombination process is that its kinetics observed at cryogenic temperature depend strongly on the light pretreatment received by the reaction centers at room temperature and during the cooling process [17]. For dark-adapted reaction centers, isolated from *Rhodobacter sphaeroides* strain R26, the value of the half-time of the charge recombination is equal to 17 ms, while it increases to a value of 83 ms for reaction centers cooled under continuous illumination [17]. In both cases, the recombination kinetics observed show deviations from a pure exponential time dependence, deviations more pronounced in the case of reaction centers cooled in the light [17]. Since these kinetics depend strongly on the three-dimensional configuration of the reactants, Kleinfeld et al. [17] have interpreted the non-exponential character of the decay kinetic of state $P^+Q_A^-$ in terms of a distribution of the donor–acceptor electron-transfer distances. According to these authors [17], the light pretreatment induces structural changes which lead to a broadening and a shift to larger distances of this distribution.

In the present paper, we have studied the low-temperature charge recombination of state $P^+Q_A^-$ for isolated reaction centers and intact chromatophores membranes of the species *Rb. sphaeroides* and *Rhodospirillum rubrum*. We observed that the kinetics of the back reaction depend strongly on the wavelength of the analysing beam in the spectral region where the absorbance changes are changing sign. We take these results as evidence for two conformational states of the reaction center.

Materials and Methods

Rb. sphaeroides strain R26 and 2-4-1 and *R. rubrum* strain G9 were grown in the light in degassed Hutner medium. Chromatophores and reaction centers were prepared as previously reported [18]. 2-4-1 reaction centers were prepared according to the procedure of Jolchine and Reiss-Husson [19]. R26 LM particles were obtained following the procedure of Debus et al. [20]. To remove one of the accessory BChl molecules R26 RC's were treated with sodium borohydride as described in Refs. 21 and 22, followed by a purification step on a DEAE cellulose column.

Absorption changes were measured with a home-built apparatus. The analysis beam provided by a 800 W quartz halogen lamp coupled to a monochromator (Jobin-Yvon, HRS2) was detected by a photodiode. The slit bandwidths were set at 0.2 nm. The output of the photodiode was amplified and numerized to allow signal averaging and storage. Actinic light was provided by a pulsed YAG laser (Quantel 481 A) coupled to a dye laser (Quantel TDL III). The characteristics of the light pulse were, around 600 nm, 10 ns duration and 50 mJ. Low-temperature measurements were achieved with an helium gas flow cryostat (Meric). Chromatophores or reaction centers suspensions in 10 mM Tris HCl (pH 8) were mixed with two volumes of glycerol to obtain clear samples at low temperature.

Decay curves were analyzed with the VOYONS program, a general software for spectroscopic data analysis [23]. Experimental decays were fitted by a sum of exponentials, with the simplex method, until the differences between experimental and computed points were purely random. The standard deviation of these differences is about 0.5%.

Results

Fig. 1 shows the light-induced difference spectrum related to the $P^+Q_A^-$ state, observed in the

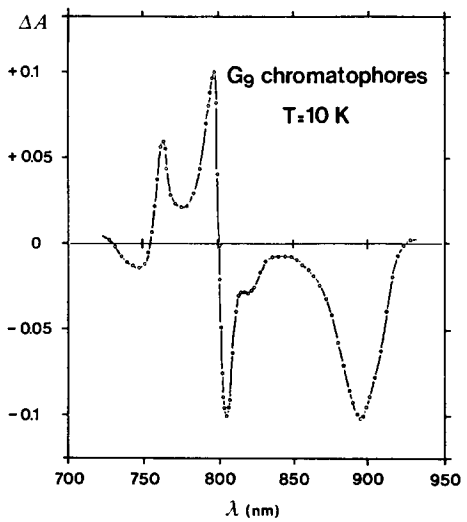

Fig. 1. Light-induced difference spectrum measured at 10 K for a suspension of chromatophores (0.5 absorbance at 800 nm) prepared from *R. rubrum* strain G9. The absorbance changes were measured 3 ms after the laser actinic flash.

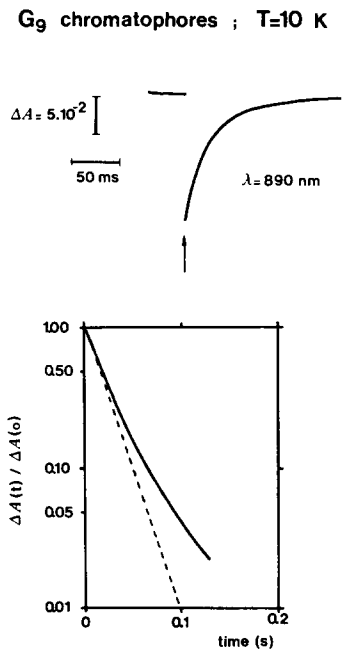

Fig. 2. Upper part: Kinetics of the flash-induced absorbance changes occurring in the long-wavelength band of the primary electron donor at 890 nm. Temperature: 10 K. Chromatophores from *R. rubrum* strain G9, in presence of 5 mM orthophenanthroline. Lower part: Semi-log plot of the kinetics depicted in the upper part.

near-infra-red region, 3 ms after the actinic laser flash for a suspension of *R. rubrum* G9 chromatophores. The temperature of the experiment was 10 K. This difference spectrum exhibits usual features associated to state $P^+Q_A^-$ at low temperature for isolated reaction centers: red bandshift of the BPheo transitions in the 730–780 nm region, positive and negative changes related to BChl molecules around 800 nm and bleaching of the long wavelength (890 nm) band of the primary donor. The decay of state $P^+Q_A^-$, i.e., the charge recombination kinetics, can be monitored at 890 nm in the long-wavelength band of the primary donor. Such kinetics is depicted in the upper part of Fig. 2. The semi-log plot (Fig. 2, lower part) demonstrated the deviation from a pure exponential time dependence as previously reported [17].

The apparent half-time $t_{1/2} = 15$ ms is comparable to what has been previously measured for isolated G9 reaction centers or related species [13]. Careful examination of the absorption changes occurring around the crossing points of the light-induced difference spectrum (approx. 801 and approx. 757 nm, Fig. 1) reveals important differences in their kinetics. This is particularly clear at 800.6 nm and 801.4 nm as shown in the upper part of fig. 3 where a factor of about 3 can be measured between the half-times of the two decays. The half-times measured at 800.6 nm and 801.4 nm are equal to 10 and 30 ms, respectively. A striking indication of the complexity of the kinetics of charges recombination is given in Fig. 3 where, at 801.2 nm, the absorbance change is negative in the first milliseconds and subsequently positive. For comparison kinetics observed at 890 nm ($t_{1/2} = 15$ ms) is also plotted in Fig. 3. Similar observations can be made with *Rb. sphaeroides* R26 or 2-4-1 chromatophores and for temperature comprised between 10 K and 80 K (data not shown).

The complexity of the kinetics of the light-induced absorbance changes around 801 nm or 757 nm is not related to electron transfer from the primary acceptor (Q_A) to the secondary (Q_B), since this transfer is strongly inhibited at cryogenic temperature. Moreover, the experiments of Fig. 3 were performed in the presence of 5 mM orthophenanthroline, a compound which blocks

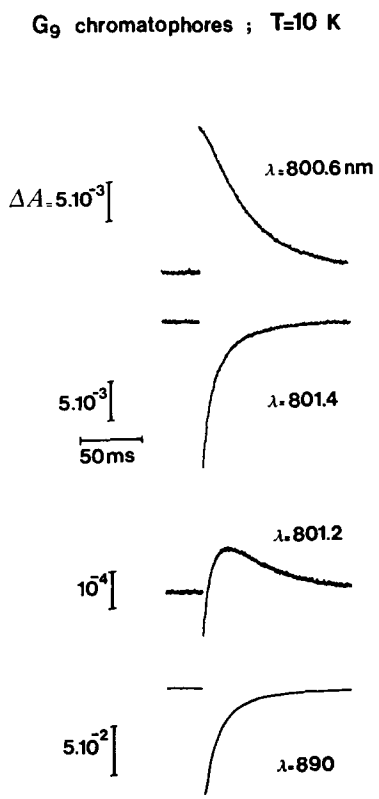

Fig. 3. Comparison of the kinetics occurring around the crossing-point of the light-induced difference spectrum (approx. 801 nm) and in the long-wavelength band (890 nm). Same conditions as Fig. 2. The half-times are: 30 ms at 800.6 nm; 10 ms at 801.4 nm; 15 ms at 890 nm.

such electron transfer [24,25]. In fact, addition of this chemical suppresses a slow decaying component of small amplitude (less than 5% of the total absorbance changes occurring at 890 nm), suggesting that some electron transfer between Q_A and Q_B occurs even at low temperature (unpublished results).

We also checked the effect of the redox state of the secondary acceptor Q_B on the kinetics of charge recombination and on the complex wavelength dependence kinetics observed around 801 nm. To that end, dark-adapted G9 chromatophores were preilluminated at room temperature by one saturating flash in the presence of 10 mM reduced TMPD as electron donor and rapidly frozen at low temperature. In such conditions the state P $Q_A Q_B^-$ is trapped and stabilized [26]. Subsequent flash excitation at low temperature will induce the state $P^+Q_A^-Q_B^-$. We found no effect of the negatively charged secondary acceptor (Q_B^-) on the kinetics of back reaction between P^+ and Q_A^- measured at 890 nm or around 801 nm compared to control experiments where the sample has not experienced a flash preillumination at room temperature (data not shown).

A similar wavelength dependence of the kinetics of charge recombination around 801 nm is also observed for isolated reaction of *Rb. sphaeroides* R26 (not shown) and 2-4-1 reaction centers (Fig. 4). There are some slight differences between the various biological preparations concerning the wavelength of the crossing points and the half-times of the kinetics of the light-induced absorbance changes. The results are summarized in Table I. No important difference could be de-

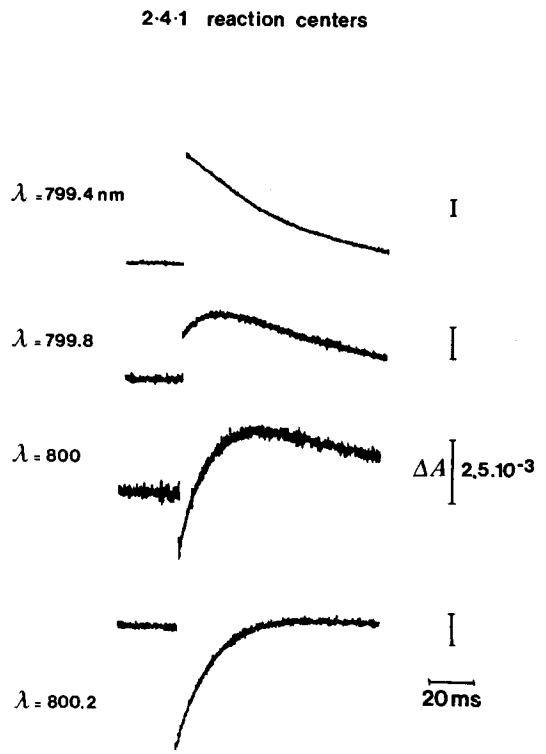

Fig. 4. Kinetics of the flash-induced absorbance changes observed around the crossing point (approx. 801 nm) of the light-induced difference spectrum of a suspension of *Rb. sphaeroides* 2-4-1 purified reaction centers. Temperature: 10 K, optical density at 800 nm, 0.4; the half-times were 32.5 ms at 799.4 nm; 9 ms at 800.2 nm and 17.5 ms at 890 nm.

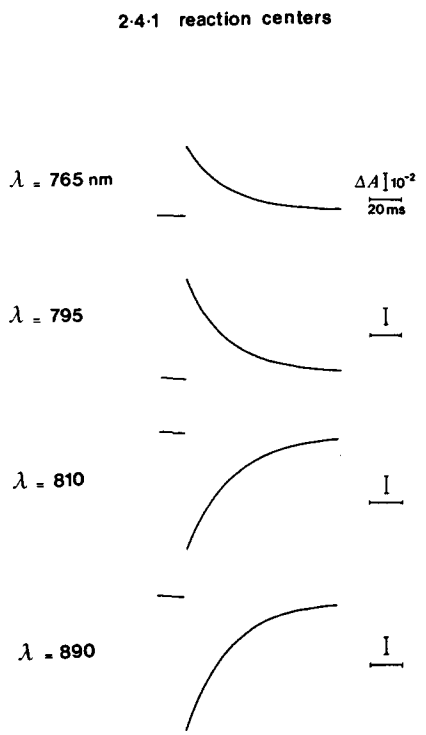

Fig. 5. Same conditions as Fig. 4 but for the main absorbance changes of the light-induced difference spectrum: 765, 795, 810 and 890 nm. The half-times are equal to 17.5 ms for the different wavelengths.

tected when comparing the kinetics of the main absorbance changes at 765, 795 and 810 nm to those of the long wavelength bleaching at 890 nm in both chromatophores (not shown) or isolated reaction centers (Fig. 5). However, significative differences are observed for the half-times measured at wavelengths between 795 nm and 810 nm (Table II). These observations further confirm that electron transfer between primary and secondary acceptors is not responsible of the complex kinetics we observed, since the RC's used in the experiment of Fig. 4 contains only one quinone molecule as shown by the half-time (80 ms) measured at room temperature for the kinetics of the charge recombination.

The simplest interpretation we can propose to explain the above findings is to suppose the presence of two populations of reaction centers. Each of those two populations possesses its own rate of charge recombination and, moreover, slightly distinct light-induced difference spectra. In that case, at any given wavelength the total absorption change will be the algebraic sum of the contribution of each population of reaction centers. If at one particular wavelength the contribution of one population is equal to zero (crossing point for its light-induced difference spectrum), only the kinetics of the other population shall be observed. For G9 chromatophores, this is approximatively the case at 800.6 and 801.4 nm (Fig. 3, upper part). Between these two wavelengths the total absorption change is the sum of two changes with opposite sign leading to the complex shape of Fig. 3. In the case of isolated 2-4-1 reaction centers, the decay of state $P^+Q_A^-$ for each of the two populations can be monitored at 799.4 and 800.2 nm, respectively (Fig. 4). The very restricted wavelength range (Figs. 3 and 4) where the two kinetic components of the decay of state $P^+Q_A^-$ can be distinguished may explain why this phenomenon has not been reported previously. Kirmaier et al. have reported different kinetic components as a function of probe wavelength for the formation of state $P^+Q_A^-$ in both *Rb. sphaeroides* [11] and *Chloroflexus aurantiacus* [27] reaction centers.

TABLE I

SUMMARY OF THE HALF-TIMES AND WAVELENGTH OCCURRENCE OF THE FAST AND SLOW PHASES OF THE BACK REACTION AT 10 K

Preparation	Fast phase		Slow phase		$t_{1/2}$ at 890 nm (ms)
	λ (nm)	$t_{1/2}$ (ms)	λ (nm)	$t_{1/2}$ (ms)	
G9 chromatophores	801.4	10	800.6	30	15
2-4-1 reaction centers	800.2	9	799.4	32.5	17.5
R26 reaction centers	801.5	11.5	800.4	36.5	18

TABLE II

SUMMARY OF DECAY KINETICS OF STATE $P^+Q_A^-$ IN *RHODOBACTER SPHAEROIDES* 2-4-1 REACTION CENTERS AT 10 K

λ (nm) of observation	765	794	796	798	799.4	800.2	801	802	806	810	890
$t_{1/2}$ (ms)	17.5	17.5	19	20	32.5	9	15.5	16.5	17.5	17.5	17.5

What are the possible origin of these two populations of reaction centers? Since they are present in the intact membrane, we can exclude their artefactual formation during the RC's purification and therefore be sure that they represent an intrinsic property of native RC's. They probably do not correspond to two distinct types of reactions center for two reasons. Firstly, there is no relation between the two populations we have evidenced kinetically at low temperature and the two types of reaction center which can be separated during DEAE column chromatography and which differ in their isoelectric points [20], since we observed that both types of RC exhibit the complex kinetics depicted in Fig. 4 (data not shown). Secondly, at room temperature the kinetics of the back reaction can be well fitted with a single exponential component [17]. This suggests no heterogeneity between RC's at least in their kinetic behaviour.

We therefore prefer a working hypothesis where a single type of reaction centers exists at low temperature under two distinct states.

These two states could preexist before the charge separation or be the consequence of nuclear relaxations induced by the photochemistry. The first hypothesis is similar to the one proposed by Kleinfeld et al. [17], who postulated that reaction center exist at low temperature under different conformational states. The second hypothesis is derived from the proposal of Kirmaier et al. [11,27] that readjustments of the pigments and/or the protein can follow the charge separation process (see also Refs. 28 and 29). We could also consider a third hypothesis, where the electron occupies two distinct sites in the reaction centers or follows two different routes during the back reaction.

To obtain more information on the factors which possibly govern these two states, we have performed several experiments with reaction centers subjected to different modifications or treatments. In order to determine the effect of protonation we first checked the influence of pH on both the main kinetics at 890 nm and the complex behaviour in the 801 nm region. For both isolated reaction centers and chromatophores, the overall kinetics of the decay of state $P^+Q_A^-$ is pH independent at low temperature in the range 6–9 in agreement with a previous report [17]. No pH effect could be observed on the kinetics occurring in the 801 nm region (data not shown). This excludes the possibility that the two populations of reaction center are interconvertible with changing pH as it is the case for state $Q_A^-Fe^{2+}$ of Photosystem II reaction centers [30].

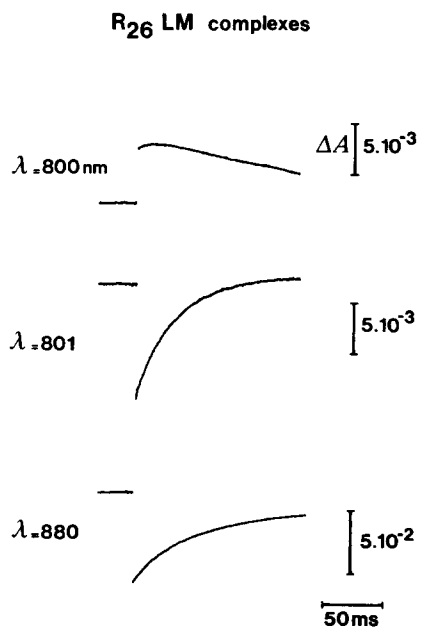

Fig. 6. Kinetics of flash-induced absorbance changes occurring at 800 nm, 801 nm and in the long-wavelength band (880 nm) for LM complexes of *Rb. sphaeroides* R26. The half-times are equal to 90 ms at 800 nm, 26 ms at 801 nm and 47 ms at 880 nm.

Removal of the H polypeptide slows down by a factor of 2.5 the half-time of the kinetics of charges recombination at 10 K (Fig. 6, lower trace) as already reported [20]. The complex behaviour around the crossing point is, however, still observed (Fig. 6, upper and middle traces). Removal of the accessory BChl molecule (BC_{MA}) not involved in the photoactive branch of the reaction center by borohydride treatment affects [21,22] neither the rate of the back reaction measured at 890 nm nor the complex behaviour in the 801 nm region (data not shown).

Illumination of reaction centers containing only one quinone molecule during the cooling process has been reported by Kleinfeld et al. [17] to slow down by a factor of 4–5 the recombination half-time measured at cryogenic temperature. We confirmed a slowing down of the kinetics of back reaction measured at low temperature when the reaction centers have been preilluminated at room temperature. However, in our case, the half-time of the kinetics measured in the long-wavelengths band (890 nm) was only multiplied by a factor of 3 in the average (Fig. 7, right trace). This difference between our experiments and those of Kleinfeld et al. [17] may be not significant, since we noticed, as these authors, variations in half-time between samples. Analysis of the light-absorption changes in the spectral region where the RC's populations can be distinguished kinetically shows that the light pretreatment lengthens by a factor of 3.5 the already slow component measured at 800.5 nm, while the kinetics of the fast component (801.5 nm) was unaffected (Fig. 7). Note, however, the smaller amplitude of this fast component in the preilluminated sample compared to the one observed for dark-adapted RC's.

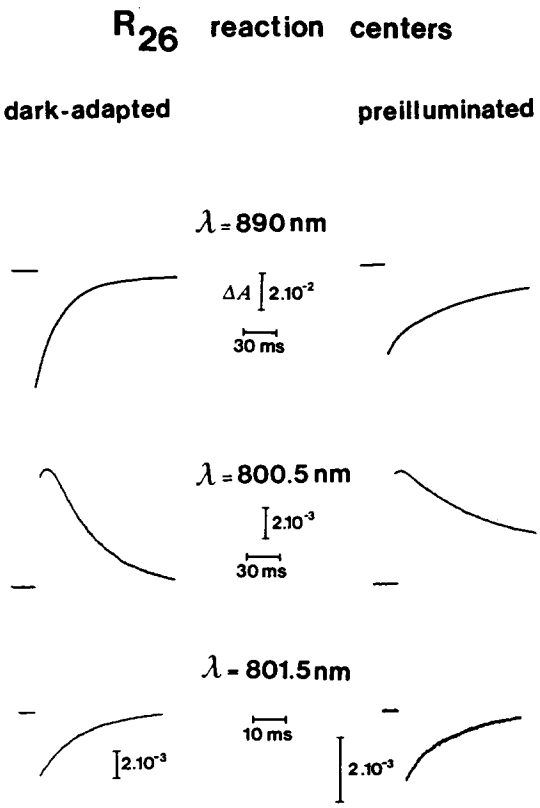

Fig. 7. Comparison of the flash-induced absorbance changes observed around 801 nm and in the long-wavelength region (890 nm) for R26 purified reaction centers. Left traces: dark-adapted reaction centers. The half-times are equal to 18 ms at 890 nm, 30 ms at 800.5 nm and 11 ms at 801.5 nm. Right traces: reaction centers preilluminated at room temperature. The half-times are equal to 51 ms at 890 nm, 105 ms at 800.5 nm and 11 ms at 801.5 nm. The temperature of the experiment was 10 K.

Discussion

The carefully spectral analysis we have performed on the light-induced absorbance changes occurring at low temperature in the near infra-red region demonstrates the complexity of the kinetics of the charge recombination process. We interpret this complexity as reflecting the presence of two states for the reaction centers at low temperature. As already stated it seems unlikely that these two states of the reaction center reflect two distinct populations of reaction centers. We can also exclude the possibility that the two difference decays of state $P^+Q_A^-$ correspond to charge recombinations though the two branches of the reaction center, since the removal of the BC_{MA} molecule affects neither the recombination times nor the ratio of the two decaying components. We therefore prefer a hypothesis when the reaction centers can exist under two different conformational states. Following the proposal of Kleinfeld et al. [17], the reaction centers exist under different conformational states. The equilibrium rate between these states at room temperature is much faster than the rate of the charge recombination. This leads to a single exponential component for the

back-reaction kinetics at room temperature. On the other hand, these different conformational states are frozen in at low temperature. Each state decays with its own time characteristic. In contrast to the proposal of Kleinfeld et al. [17] wo supposed a distribution of different structural states, we interpret our results as evidence of only two discrete states for the reaction centers. These two conformational states could also result from movements of pigments and/or protein after the charge separation [11,27–29].

The presence of two conformational states of reaction centers exhibiting different rates of charge recombination may give a straightforward explanation of the non-exponential time dependence of the decay of state $P^+Q_A^-$ observed at 890 nm (Fig. 2, lower part) [17], since for that analysing wavelength both states contribute to the light-induced absorbance change. To check that possibility we have first analysed the charge recombination kinetics of each population monitored at 800.4 and 801.5 nm in the case of R26 isolated reaction centers. Both light-induced absorbance changes can be well fitted with a single exponential (Fig. 8A and B) if one excepts a fast component of very small amplitude (1%) of the total absorbance changes, which is probably a laser artefact. The relative error, defined as the ratio between the root mean square deviation divided by the maximum signal amplitude, is equal to 5/1000 at 801.5 nm and 4/1000 at 800.4 nm. The values of the half-time of the two decays are 11.5 and 36.5 ms at 801.5 and 800.4 nm, respectively. Similar good fittings could also be obtained for the kinetics observed for G9 chromatophores or 2-4-1 RC's (results not shown). We have then decomposed the absorbance changes occurring in the long-wavelength region by imposing the number of exponential components (2) and the values (11.5 and 36.5 ms) for the half-times as determined from the previous analysis of the kinetics observed at 800.4 and 801.5 nm. A very good fit could be obtained under these conditions for the kinetics observed at 890 nm (Fig. 9). The standard deviation between the experimental curve and the computed one is equal to 2/1000. The ratio of the amplitudes of the fast and slow components was in the average equal to 1.5 (60/40).

The situation is more complex for LM particles. In addition to the two exponential components observed around 800 and 801 nm (Fig. 6) a third component has to be added in the decomposition to obtain a good fit of the kinetics of the long-wavelength bleaching. The half-time of this component is 600 ms and its amplitude 12% of the total absorption changes. The two other components are still in the ratio 1.5.

For reaction centers preilluminated at room temperature a good decomposition of the main absorbance changes occurring at 890 nm with two exponential components whose characteristic times have been deduced from the kinetics observed at 800.5 and 801.5 nm, i.e., 11 and 105 ms, respec-

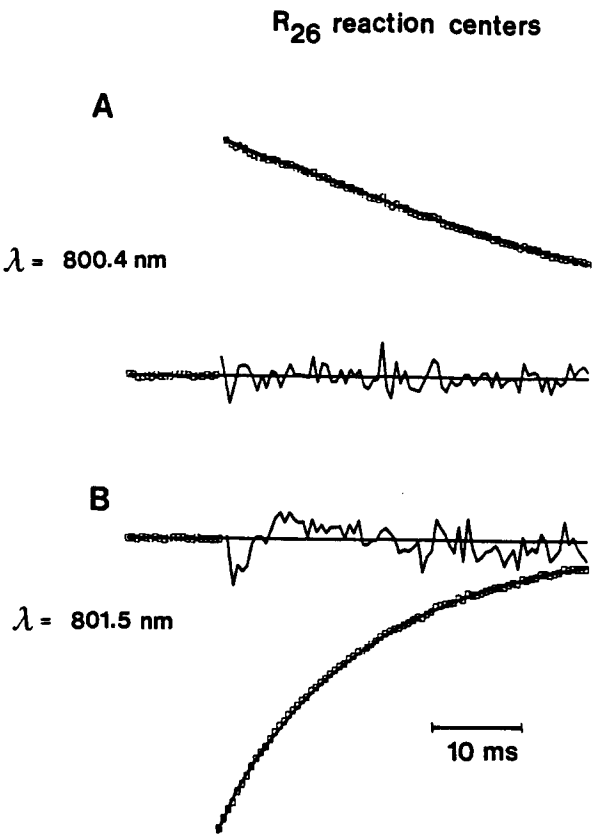

Fig. 8. Fitting of computed exponentials (full line) and experimental decay curves (square) measured respectively at 800.4 (A) and 801.5 nm (B) for a suspension of R26 reaction centers. The half-times of the two exponential components are equal to 36.5 ms (800.4 nm) and 11.5 ms (801.5 nm). The difference between the computed and experimental curves, enlarged by a factor of 10, represents a standard deviation of 4/1000 (curve A) and 5/1000 (curve B).

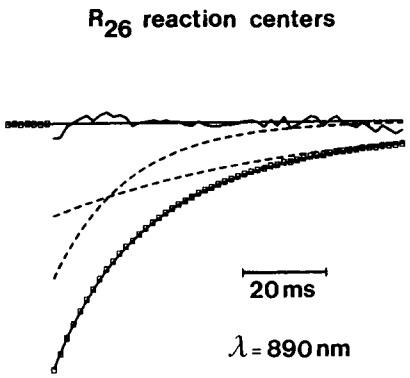

Fig. 9. Decomposition of the absorbance changes occurring in the long-wavelength band (890 nm) of a suspension of R26 reaction centers. The experimental curve (squares) is compared to the computed sum (full line) of the two exponentials (broken line) fitting the absorbance changes observed at 800.4 and 801.5 nm (see Fig. 8). The relative weights are 62 and 38 for the fast and slow components, respectively. The standard deviation is equal to 2/1000. The difference between computed and experimental curves is enlarged by a factor 10.

tively, is also obtained (not shown). In this case, however, the ratio between the fast and the slow components is equal to 0.5 instead of 1.5 for dark-adapted reaction centers (Fig. 8).

We therefore conclude that the non-exponential character of the relaxing process of state $P^+Q_A^-$, measured in the long wavelength band is due to the presence of two conformational states decaying with different rates.

These two conformational states are not affected by pH. They do not depend on the redox state of the secondary acceptor or even on its presence in the reaction center. Important modifications of the reaction center like the removal of the H polypeptide affects the two conformational states in a similar manner by lengthening their decays by a factor of 2.5. On the other hand, preillumination at room temperature of reaction centers slows down by a factor of 3.5 the kinetics of the slow decaying state, while those of the fast decaying state is not affected. The ratio between the fast and slow decaying states is 0.5 for RC preilluminated at room temperature, instead of 1.5 for dark-adapted reaction centers. This suggests that room-temperature preillumination leads to a preferential structural state as already proposed in Ref. 17.

By assuming that the different half-times depend only on the donor acceptor distances we can calculate with $t(r) = t(\bar{r}) \cdot \exp(r - \bar{r})/r_0$ (where r_0 is a scaling factor of 1 Ångstrøm according to Hopfield [15] and Jortner [16]), that the two conformational states of dark-adapted reaction centers differ by 5% in that distance. For reaction centers cooled under illumination this difference reaches 12%.

Further work will be needed to specify the physiological significance of these two states of the reaction center, their structural characteristic, their temperature dependence and their occurrence in other species.

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