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Rapid report

Electron transfer in the heliobacterial reaction center: evidence against a quinone-type electron acceptor functioning analogous to A_1 in photosystem I

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Abstract

Membrane fragments from *Heliobacillus mobilis* were characterized using time resolved optical spectroscopy and photovoltage measurements in order to detect a possible participation of menaquinone (MQ), functioning analogous to the phylloquinone A_1 in photosystem I, as intermediate in electron transfer from the primary acceptor A_0 to the iron–sulfur cluster F_X in the photosynthetic reaction center. The spectroscopic data obtained exclude that electron transfer from a semiquinone anion MQ^- to F_X occurred in the time window from 2 ns to 4 μs, where it would be expected in analogy to photosystem I. In the case of a prereduction of F_X, only the primary pair $P798^+A_0^-$ was formed. The photovoltage data yielded a single kinetic phase with a time constant of 700 ps for the transmembrane electron transfer beyond A_0; the relative amplitude of this phase suggests that it reflects electron transfer from A_0^- to F_X. © 1998 Published by Elsevier Science B.V.

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Heliobacteria are recently discovered strictly anaerobic, anoxygenic photosynthetic bacteria (reviewed in Ref. [1]) which are characterized by the presence of bacteriochlorophyll (BChl) *g* as the major pigment [2]. Their photosynthetic reaction center (RC) is located in the cytoplasmic membrane, and is believed to be organized as a single homodimeric pigment–protein complex [3] in which two identical protein subunits harbor the light-harvesting antenna BChl *g* molecules and the RC cofactors (reviewed in Ref. [4]). Heliobacteria perform predominantly cyclic electron transport [5] using menaquinone (MQ)—the sole quinone found in heliobacteria—for their quinone pool [6].

A number of studies have shown that the heliobacterial RCs resemble those of green sulfur bacteria and photosystem I (PS I) from higher plants, and the similarity between their electron acceptor chains has become obvious [7,8]. Like PS I and green sulfur bacteria, heliobacteria use iron sulfur clusters as terminal electron acceptors and a monomeric chlorophyll (Chl) molecule as primary electron acceptor A_0 (reviewed in Refs. [4,9]).

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Electron transfer processes in the RC of PS I have been studied in some detail, and the path of electron transfer is reasonably well established. Three [4Fe–4S] clusters, F_X, F_A and F_B, serve as terminal acceptors. Initial charge separation occurs within a few picoseconds from the primary donor P700, a Chl *a* dimer, to A_0, a monomeric Chl *a*. From A_0 the electron is then transferred in about 30 ps to the secondary acceptor A_1, a phylloquinone, and subsequently in 15 to 200 ns to F_X before proceeding to F_A/F_B (see Ref. [10] for a recent review).

In the RC of heliobacteria, the primary electron donor (P798) has been proposed to be a dimer of BChl *g* [11] or its epimer Bchl *g′* [12], absorbing at 798 nm [13,11]. The primary acceptor A_0 in heliobacteria has been identified as a monomeric 8′-hydroxy-chlorophyll *a* molecule [14]. The reoxidation of A_0^- during forward electron transfer occurs in about 600 ps [15–17]. Optical studies [18] as well as primary structure analyses [8,3] suggest a [4Fe–4S] cluster analogous to F_X in heliobacteria, although to date no corresponding EPR signal has been observed. Nitschke et al. [19] reported EPR signals analogous to F_A and F_B in *Heliobacterium (Hb.) chlorum*. Despite the obvious similarities between PS I and the heliobacterial RC with respect to the pathways of electron transfer and the cofactors involved, the question of a possible involvement of a quinone acceptor in heliobacteria, analogous to A_1 in PS I, has remained very controversial during the years. A similar controversy exists for green sulfur bacteria (see Refs. [20,21] for recent reviews) which are not subject of this report.

In heliobacteria, Brok et al. [22] suggested the involvement of a semiquinone as electron acceptor based upon the properties of a high *g*-value EPR radical signal photoaccumulated during freezing. The existence of an A_1 analogue was also suggested on the basis of charge recombination kinetics at low temperatures which appeared to be faster than the recombination of $P798^+F_X^-$ but slower than that of $P798^+A_0^-$ [19,23]. A role of an A_1 analogue in the heliobacterial RC at room temperature was supported by Trost et al. [8] based upon a two-electron fit to the redox titration of long-lived (0.6 ms) photobleaching of P798, possibly indicating double reduction of a quinone.

On the other hand, extraction of menaquinone—the only quinone present—from heliobacterial membranes did not result in significant changes in the yield of stable charge separation, suggesting that menaquinone is not an essential participant in the electron acceptor chain [24]. In addition, picosecond transient absorption difference spectroscopy on *Heliobacillus (Hc.) mobilis* indicated the only charge-separated state observed after $P798^+A_0^-$ to be $P798^+F_X^-$ [25]. This study was restricted, however, to wavelengths above 400 nm and times below 1 ns.

In an attempt to address the existing controversy, and to detect a possible participation of menaquinone as an intermediate in electron transfer from the primary acceptor A_0 to the iron–sulfur cluster F_X, we performed transient absorption measurements on a timescale of 2 ns to 4 μs at wavelengths between 360 and 450 nm. This is the first room temperature study trying to resolve a possible A_1 contribution in a heliobacterial species on the time scale and in the spectral region where the function of A_1 can be detected in PS I. Complementary data were obtained by photovoltage measurements following the transmembrane electron transfer with a time resolution of about 50 ps. Preliminary results of this work have been presented [26,27].

Growth of *Hc. mobilis* cultures (obtained from the DSM, Braunschweig, Germany) and the isolation of periplasmic membrane fragments followed the protocol described in Ref. [28]. The membranes were resuspended in 50 mM degassed MOPS (3-[*N*-Morpholino]propane-sulfonic acid) buffer, pH 7.0. Sample manipulations were carried out under argon.

Flash absorption spectroscopy (see Ref. [29] for a brief description of principles and applications in photosynthesis research) in the near UV and blue spectral region was performed with a set-up similar to the one described in Ref. [30]. The sample was excited repetitively (1 Hz) with laser pulses of either 7 ns pulse duration and about 1 mJ/cm^2 at 700 nm from a dye laser (ND6000 from Continuum), or with pulses of 300 ps pulse duration and about 1 mJ/cm^2 at 532 nm from a frequency doubled Nd/YAG laser (YG501-10 from Quantel). The measuring light was provided by the relatively flat top of a 50 μs Xe flash. Adequate combinations of interference and colored glass filters placed in the measuring beam before the sample and in front of the detector (FND100Q from EG and G) yielded a spectral bandwidth of 2 to

8 nm. The detector output to 50 Ω was amplified (30 dB, 500 Hz–1.7 GHz) and recorded by a digitizing oscilloscope (DSA 602A with amplifier plug-in 11A52 from Tektronix).

The kinetics of $P798^+$ in double flash experiments were monitored at 865 nm using a cw laser diode (TXSK 2503 from Telefunken) as measuring light source. The sample was excited from one side by an about 90% saturating Xe flash of 3 μs (FWHM) duration, and from the other side by a laser flash of 300 ps duration and about 10 mJ/cm^2 at 532 nm (see above). The detection system was the same as described above, except for measurements on a millisecond time scale where the detector was terminated with 10 kΩ and coupled directly to an 11A33 amplifier plug-in, yielding a time resolution of about 5 μs.

Picosecond photovoltage measurements were carried out in a microcoaxial measuring cell. Details of the technique and the data analysis can be found elsewhere [31]. Membrane fragments were oriented by applying a short electric pulse (400 V/cm, ~30 ms) to the electrodes. The excitation flash was obtained from a frequency-doubled Nd-YAG laser (PY61 from Continuum, pulse duration 20 ps). The electric signals were amplified by a broad band amplifier (Nuclétude, bandwidth 6 GHz), and digitized with a 7 GHz oscilloscope (Intertechnique IN7000). Time resolution and apparatus response function were determined by measuring the ultrafast charge displacement in oriented purple membranes from *Halobacterium halobium*. Kinetic analysis was based on iterative convolution of a multiexponential charge separation current according to a sequential reaction scheme with the apparatus response function.

All measurements were performed at room temperature.

In order to detect a possible contribution of menaquinone as A_1 analogue, we recorded flash-induced absorption changes in membranes from *Hc. mobilis* between 360 and 450 nm in steps of 10 nm. This spectral region was chosen because reduction of menaquinone to its semiquinone anion (MQ^-) is accompanied by a broad absorption increase around 400 nm with a differential extinction coefficient $\Delta\varepsilon$ of about 11 $mM^{-1}\ cm^{-1}$ [32–34], and reduction of F_X is accompanied by a broad bleaching around 430 nm with $\Delta\varepsilon$ of 10–11 $mM^{-1}\ cm^{-1}$ [18]. Hence, should electron transfer occur from MQ^- to F_X, bleachings with the same kinetics as this transfer should be observed around 400 nm (due to reoxidation of MQ^-) and around 430 nm (due to reduction of F_X). This kinetics should follow an initial absorbance change reflecting the first two steps of charge separation which occur on a sub-nanosecond time scale (see above).

Fig. 1 shows typical traces measured with a time-resolution of about 10 ns. Depending on the wavelength, we observed a step-like rise or decrease of absorbance. The absorbance level reached after about 10 ns remained essentially stable on the depicted time scale (1.6 μs), and even on an extended time scale of 4 μs (not shown). Based on the current knowledge of electron transfer in heliobacteria and in line with published spectra [18,7], we attribute the relatively

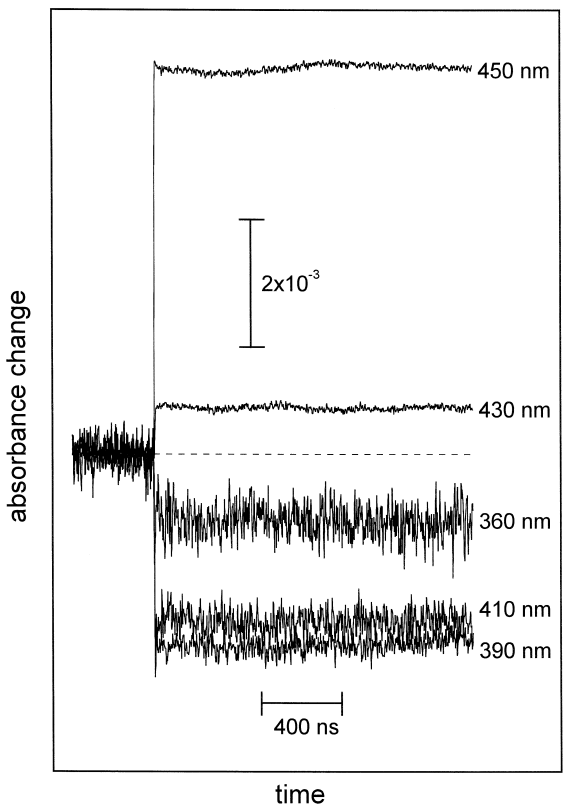

Fig. 1. Flash-induced absorbance changes at the indicated wavelengths for membrane fragments from *Hc. mobilis*. The sample contained 20 μM phenazine methosulfate and 20 mM sodium ascorbate, and had an optical density of 1 at 788 nm along the 2 mm optical path for the measuring light. Excitation: 7 ns laser pulses at 700 nm; electronic bandwidth of the detection system: 500 Hz–100 MHz. Each trace is the average of 200 transients.

long-lived absorbance changes to a charge separated state $P798^+F^-$, where F is one of the three iron–sulfur clusters, most likely F_X (see below). A bleaching phase as expected for electron transfer from MQ^- to F_X could not be detected in the time window from 10 ns to 4 μs. This holds also true for the wavelengths 370, 380, 400, 420 and 440 nm (data not shown).

From published spectra [18] we estimate the amplitude of the long-lived $P798^+F_X^-$ signal at 450 nm to correspond to $\Delta\varepsilon \approx 32–35\ mM^{-1}\ cm^{-1}$. The bleaching due to electron transfer from MQ^- to F_X should be in the order of at least 30% of this amplitude at some wavelengths in the 360 to 450 nm range, or even larger if the bleachings due to the reoxidation of MQ^- and the reduction of F_X overlap (see above). Hence we should have been able to detect electron transfer from MQ^- to F_X if it occurred in the time window from 10 ns to 4 μs.

We considered the possibility that this electron transfer might be considerably faster than the corresponding transfer from A_1^- to F_X in PS I ($t_{1/2} = 15–200$ ns). By using shorter excitation pulses (300 ps FWHM) and a larger bandwidth of the digitizer, we achieved a time resolution of about 2 ns. Even with this time resolution we were not able to observe a significant bleaching phase between 360 and 450 nm at times later than 2 ns after the excitation flash. Fig. 2 shows traces at three wavelengths out of the ten measured; the amplitude of the absorbance change at 450 nm (not shown) was 5.8×10^{-3}, thus electron transfer from MQ^- to F_X should have resulted in a bleaching phase with an amplitude of at least 1.7×10^{-3} (see above). At 430 nm the initial bleaching was followed by an absorbance increase with an apparent half time of about 1 ns. The sign of this phase is opposite to what one expects for electron transfer from MQ^- to F_X, and may reflect the response of the detection system to the decay of singlet excited antenna pigments. In summary, our results exclude that electron transfer from MQ^- to F_X occurred in the time window from 2 ns to 4 μs.

As an independent approach to check for the existence of an A_1 analogue in heliobacteria, we performed the following double flash experiment: membranes from *Hc. mobilis* containing an efficient artificial electron donor (200 μM neutral red, reduced by dithionite) were exposed to two nearly saturating excitation flashes spaced by 4.3 ms, and the kinetics

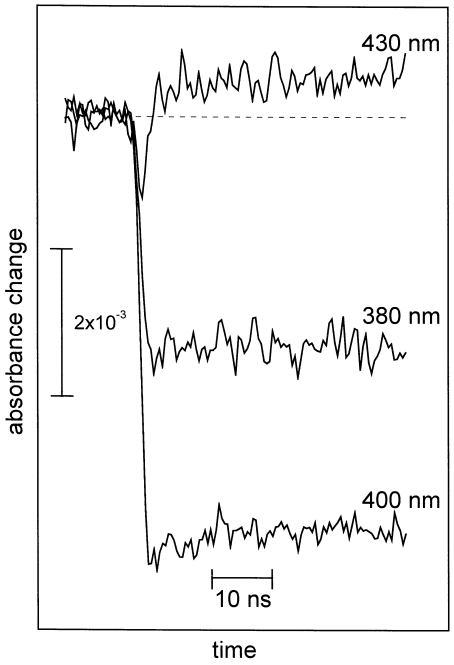

Fig. 2. Absorbance changes measured as in Fig. 1 but with improved time resolution (excitation: 300 ps laser pulses at 532 nm; electronic bandwidth: 500 Hz–200 MHz).

of $P798^+$ was followed by monitoring the absorbance changes at 865 nm. The first flash created the state $P798^+F_X^-$ (F_A and F_B were most likely absent or not functional in our preparation; see below). $P798^+$ was rereduced by neutral red with $t_{1/2} \approx 700$ μs, so that just before the second flash, only about 2% of P798 was still oxidized (Fig. 3). As no artificial electron acceptor was present, F_X should have remained in the reduced state in the majority of the centers. Hence the second flash was expected to create a radical pair composed of $P798^+$ and the reduced form of the acceptor preceding F_X. $P798^+$ should then decay with the kinetics characteristic for charge recombination in this pair. In fact, about 80% of the absorption change on the second flash decayed with $t_{1/2} \approx 15$ ns (inset of Fig. 3), the well known kinetics of charge recombination in the primary pair $P798^+A_0^-$ [35,36]. About 6% decayed with $t_{1/2} \approx 20$ μs, and the remainder with $t_{1/2} \approx 700$ μs as on the first flash. The last phase most likely arose from centers which had not been hit by the first flash (about 10%), and from centers where charge recombination in the pair $P798^+F_X^-$ (intrinsic half time about 10 ms, see below) had occurred between the two flashes in compe-

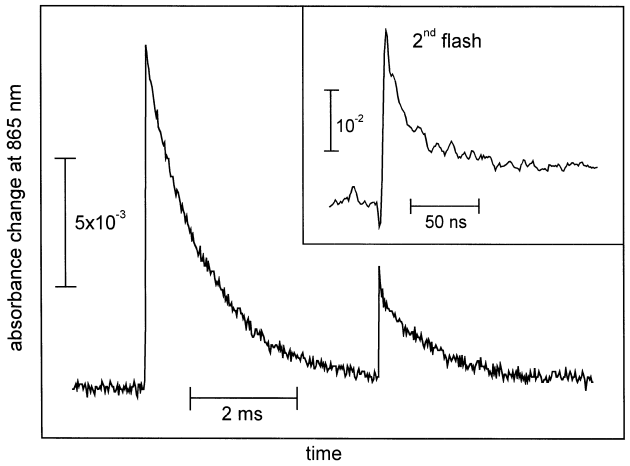

Fig. 3. Absorbance changes at 865 nm for membrane fragments from *Hc. mobilis*, induced by a 3 μs Xe flash and a 300 ps laser flash at 532 nm fired at an interval of 4.3 ms. The sample contained 200 μM neutral red and 20 mM sodium dithionite, and had an optical density of 14 at 788 nm along the 10 mm optical path for the measuring light. Electronic bandwidths: DC—70 kHz for the main trace and 500 Hz–100 MHz for the inset. Averages of 3 transients for the main trace and 28 transients for the inset. Distance between the flash groups: 10 s.

tition with electron donation by neutral red. The small 20 μs phase presumably reflects the decay of the P798 triplet state formed via charge recombination in the primary pair [35,36]. Thus our data demonstrate that in the case of a prereduction of F_X, only the primary pair $P798^+A_0^-$ is formed, providing additional evidence against a functional intermediate between A_0 and F_X in heliobacteria. It could be argued that reduction of F_X might prohibit electron transfer from A_0^- to MQ. This seems, however, unlikely because the driving force of this transfer is expected to be rather large (in the order of 300 meV for the analogous transfer in PS I [10]).

The occurrence of primary pair recombination upon the second flash indicates that the membrane fragments used in the present study contain only one functional electron acceptor beyond A_0. This acceptor is most likely F_X since in the absence of an efficient artificial electron donor, we observed rereduction of a large fraction of $P798^+$ with $t_{1/2} \approx 10$ ms (not shown), a value which is close to the 14 ms attributed to charge recombination in the pair $P798^+F_X^-$ in *Hc. mobilis* membranes devoid of F_A and F_B [18]. Thus it appears that F_A and F_B are easily lost or inactivated during preparation of heliobacterial membranes. Loss of F_A and F_B may similarly explain the prominent 8 ms charge recombination observed by Kleinherenbrink and Amesz in membranes from *Hb. chlorum* [37].

The fast photoelectric response induced by a picosecond flash in oriented membranes of *Hc. mobilis* is shown in Fig. 4. On a timescale of up to 6.5 ns two different kinetic phases can clearly be distinguished. An initial fast rise ($\tau < 50$ ps) is followed by a slower rising phase, characterized by a time constant of about 700 ps and a relative amplitude of $38 \pm 6\%$.

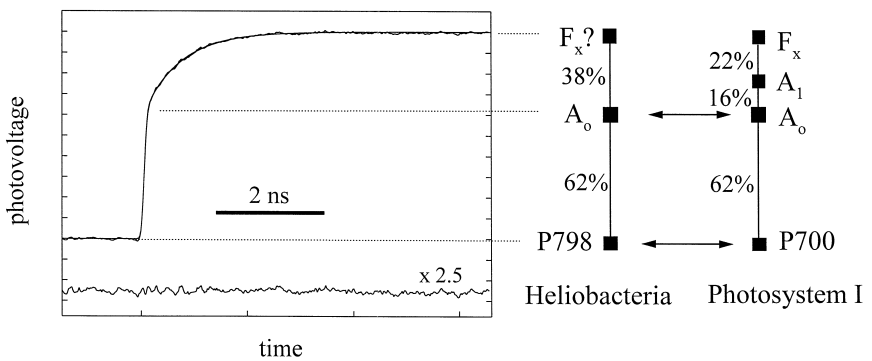

Fig. 4. Left: Photoelectric response induced by a picosecond flash in oriented membrane fragments from *Hc. mobilis*. The sample contained 10 mM sodium ascorbate and 100 μM phenazine methosulfate. A best fit of the rise kinetics with two exponential phases is shown superimposed on the experimental trace, and the residual plot is shown below. Fit parameters: time constants $\tau_1 = 30$ ps, $\tau_2 = 690$ ps; electrogenicity ratio $a_2{:}a_1 = 0.62$. An exponential decay due to the discharge of the capacitative measuring cell ($\tau_d = 1.73$ ns) has been deconvoluted for clarity. Right: Comparison of (dielectrically weighted) transmembrane distances between cofactors in the RC of heliobacteria and PS I. See text for further details.

We attribute the first, faster phase to trapping of the excitation energy by formation of the primary pair $P798^+A_0^-$. The second, slower phase is attributed to charge stabilization by electron transfer from A_0^- to a further electron acceptor. The electrically observed kinetics of 700 ps compares well to the kinetics of recovery of the bleaching at 670 nm observed in optical measurements [15,16] which was attributed to reoxidation of the chlorophyll acceptor A_0^- due to forward electron transfer. We could not detect a third electrogenic phase in the time window up to 6.5 ns.

Photovoltage measurements cannot give direct information on the chemical nature of the secondary acceptor to which the electron is transferred with a time constant of 700 ps. However, the amplitudes of the electrogenic phases provide important information about the relative transmembrane distances between the cofactors involved. In PS I from *Synechocystis*, the electron transfer steps $A_0 \rightarrow A_1$ and $A_1 \rightarrow F_X$ are electrogenic, with relative contributions of 16% and 22%, respectively, to the transfer P700 $\rightarrow F_X$ [38,39], suggesting a transmembrane position of A_1 roughly midway between A_0 and F_X. The relative electrogenic amplitude of the 700 ps phase in heliobacteria matches the sum of the two phases attributed to the transfer $A_0 \rightarrow A_1 \rightarrow F_X$ in PS I. Assuming a similar dielectric environment, this finding together with the absence of a third electrogenic phase suggests that during the 700 ps reaction in heliobacteria the electron arrives on F_X. It is of note, however, that in the case of MQ and F_X lying in the same membrane plane, electron transfer between them would not be electrogenic, thus not detectable by photovoltage measurements.

The absence of a bleaching phase between 360 and 450 nm in the time window from 2 ns to 4 μs in *Hc. mobilis* (Figs. 1 and 2) excludes that electron transfer from MQ^- to F_X occurred with similar kinetics as the transfer from A_1^- to F_X in PS I. Should electron transfer from MQ^- to F_X occur outside of this time window and not be electrogenic (see above), one would still expect to observe the pair $P798^+MQ^-$ in the above double flash experiment (Fig. 3). This was not the case, strongly supporting the notion of electron transfer from A_0^- to F_X in *Hc. mobilis* to be direct, with a time constant of about 700 ps. Data on purple bacterial RCs indicate that the edge to edge distance between the cofactors should be shorter than about 10 Å to realize an intraprotein electron transfer in 700 ps [40]. The edge to edge distance between A_0 and F_X in PS I as estimated from the X-ray structure [41] is about 14 Å (see also Ref. [10]). This suggests that A_0 and F_X in heliobacteria are closer than in PS I. Alternatively, the 700 ps transfer may proceed via the superexchange mechanism (see Ref. [42] for a brief description) with e.g. an aromatic amino acid as virtual intermediate.

Our data are consistent with findings mentioned before [24,25] but provide in addition for the first time conclusive spectroscopic evidence that menaquinone is not involved in forward electron transfer in the RC of *Hc. mobilis* in a way similar to the phylloquinone A_1 in PS I. Most of the previous work favoring a function of menaquinone as A_1 analogue in heliobacteria has presented only indirect evidence [8,19,22]. The only direct spectroscopic data in favor of a menaquinone intermediate in the heliobacterial RC is a low temperature photoaccumulated EPR spectrum considered to be consistent with a semiquinone radical [22]. Whatever the chemical nature of the observed species, it may be on a side path rather than being a constituent of the normal forward electron transfer chain, and may be reduced with significant quantum yield only at low temperature (a possibility mentioned already in Ref. [22]).

One could speculate that the absence of an A_1 analogue in heliobacteria might be related to their strict anaerobicity. In PS I in oxygenic systems phylloquinone A_1 might have a protective function by accelerating the reoxidation of A_0^-, thus decreasing the probability of primary pair recombination and concomitant formation of the P700 triplet state, which could react with oxygen forming harmful singlet oxygen. In the strictly anaerobic heliobacteria such a 'quinone protector' might be redundant.

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