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# CHARACTERISTICS OF THE PHOTOSYSTEM II REACTION CENTRE

## II. ELECTRON DONORS

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**The properties of Photosystem II electron donation were investigated by EPR spectrometry at cryogenic temperatures. Using preparations from mutants which lacked Photosystem I, the main electron donor through the Photosystem II reaction centre to the quinone-iron acceptor was shown to be the component termed Signal II. A radical of 10 G line width observed as an electron donor at cryogenic temperatures under some conditions probably arises through modification of the normal pathway of electron donation. High-potential cytochrome *b*-559 was not observed on the main pathway of electron donation. Two types of PS II centres with identical EPR components but different electron-transport kinetics were identified, together with anomalies between preparations in the amount of Signal II compared to the quinone-iron acceptor. Results of experiments using cells from mutants of *Scenedesmus obliquus* confirm the involvement of the Signal II component, manganese and high-potential cytochrome *b*-559 in the physiological process leading to oxygen evolution.**

### Introduction

The primary photochemistry of the PS II reaction centre involves the transfer of an electron from the primary chlorophyll donor, P-680, to the primary acceptor, a quinone-iron complex, QFe. The pathway of electron donation from water to $P\text{-}680^+$ is poorly understood, despite much research, because the pathway is complex [1]. Of the EPR-detectable components arising from this pathway, Signal II, an organic radical near *g* 2.00, has received most attention. Signal II was first observed by Commoner et al [2] and is light induced, with a number of components observed at room temperature, each characterised by their rates of decay [3–8]. All of these components appear to have high redox potentials and identical EPR spectra. Two of these, termed Signal $II_{vf}$ and Signal $II_f$, both rapid transient signals at room temperature, have been used as probes of the electron donation to P-680 and of reactions involving electron acceptor components [9–11]. The study of the *g* 2.00 radical region by electron spin echo [12] also detected multiple signals arising from the Signal II component and it was observed that reversible and irreversible changes occurred upon illumination. The structure of the component producing Signal II is unknown, but evidence has been presented suggesting the involvement of a radical of plastoquinone [12–15].

In a previous paper [16], we reported the detection of a light-induced EPR Signal II at cryogenic temperatures, termed Signal $II_{lt}$. The appearance of this signal upon illumination at temperatures as low as 5 K indicated that the components giving

Abbreviation: PS, photosystem.

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rise to this signal occupied a position close to the reaction centre. Further work using chloroplasts from a mutant of barley lacking PS I [17] and oxygen-evolving particles from *Phormidium laminosum* [18] suggested that Signal $II_{lt}$ was a good marker for PS II activity at cryogenic temperatures.

Two further signals tentatively identified as PS II donors were a 10 G, *g* 2.00 radical [17] and a *g* 3 cytochrome signal attributed to native or modified high-potential cytochrome *b*-559 [18–20], both of which were photo-oxidised at cryogenic temperatures. The relationships between these components and Signal II were not fully understood.

In this study we examined the properties of PS II donation at cryogenic temperatures using a variety of preparations, including mutants where PS I was absent. These preparations allowed direct direction of both acceptor and donor components

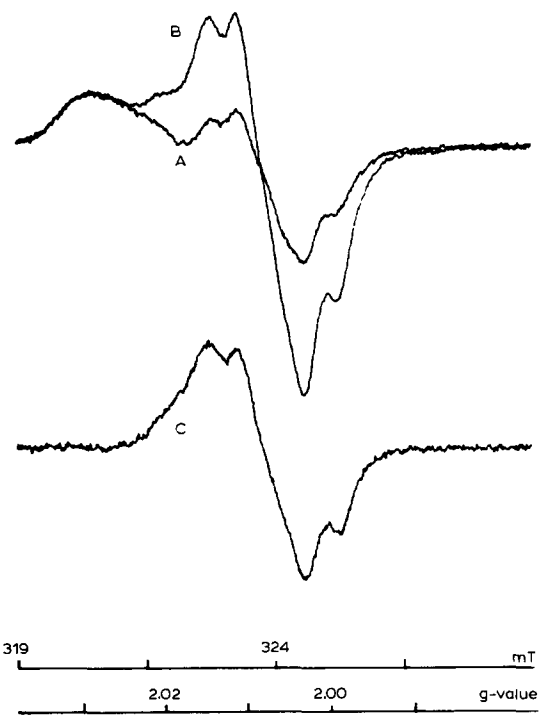


Fig. 1. Signal II photoinduced at 13 K in freshly prepared membranes of *C. reinhardii* mutant F54-14. (A) Inner line: sample of membranes frozen after dark adaptation for 15 min; (B) outer line: sample after subsequent illumination at 13 K; (C) difference spectrum (B–A) showing light-induced signal with Signal II characteristics. EPR conditions: temperature 13 K; modulation amplitude 0.2 mT; microwave power 1 mW.

which demonstrated that normal electron donation to the quinone-iron acceptor produces Signal $II_{lt}$ and suggested that the 10 G radical probably arises through abnormal electron transport due to membrane damage.

## Materials and Methods

Barley (*Hordeum vulgare*) viridis zb63 chloroplasts and *Chlamydomonas reinhardii* F54-14 mutant membranes, detergent and DEAE-treated particles were prepared as described in Refs. (21 and 22). EPR spectrometry, *g* value calculation and illumination were performed as detailed in Refs. 16 and 21. A Chromatix tunable dye laser was used for the experiment shown in Fig. 3.

*Scenedesmus obliquus* strain $D_3$ and the mutants LF1 and LF2 were grown as described in Refs. 23 and 24. Illumination of the growing cultures was as described in the text. Samples of these mutants were prepared by washing the cell pellet obtained after centrifugation at 5000 × *g* for 5 min in growth medium. After further centrifugation the cells were resuspended in a minimum volume of growth medium before transfer to EPR tubes as 0.3-ml samples. Oxygen evolution was measured with a standard oxygen electrode using ferricyanide as artificial electron acceptor.

## Results

### *Signal $II_{lt}$*

In membranes of the *C. reinhardii* mutant F54-14 and chloroplasts of barley zb63, the lack of changes near *g* 2.00 associated with P-700 of PS I allowed a clear study of the changes occurring in PS II. In freshly prepared membranes of mutant F54-14 or in zb63 chloroplasts, the light-induced changes observed at low temperature in this region were mainly restricted to those involving the EPR species termed Signal II. Fig. 1 demonstrates the effect of illumination on a frozen sample of freshly prepared F54-14 membranes. The membranes were frozen after a period of dark adaptation and show a feature around 320 mT of the possible high-potential iron protein component shown in Ref. 21. Before illumination, Fig. 1A (inner line), some Signal II was present together with a narrower unknown component. After illumination, in

this case at 13 K, Fig. 1B (outer line), a large proportion of the signal produced by illumination was also Signal II, i.e., Signal $II_{lt}$ [16]. Fig. 1C shows the difference spectrum revealing the line shape of the signal produced by the 13 K illumination period. Subsequent illumination of this sample at 200 K produced a further increase in signal size, again mostly composed of Signal II. The appearance of the *g* 1.82 signal of the semiquinone-iron acceptor followed this pattern with reduction occurring only in a minority of centres below 100 K and illumination at 200 K or above being required to develop the full semiquinone-iron signal size [21]. This heterogeneity among PS II centres is discussed further below.

In previous papers [16,18,25], we had suggested that the appearance of Signal II represented a donation of electrons through the reaction centre

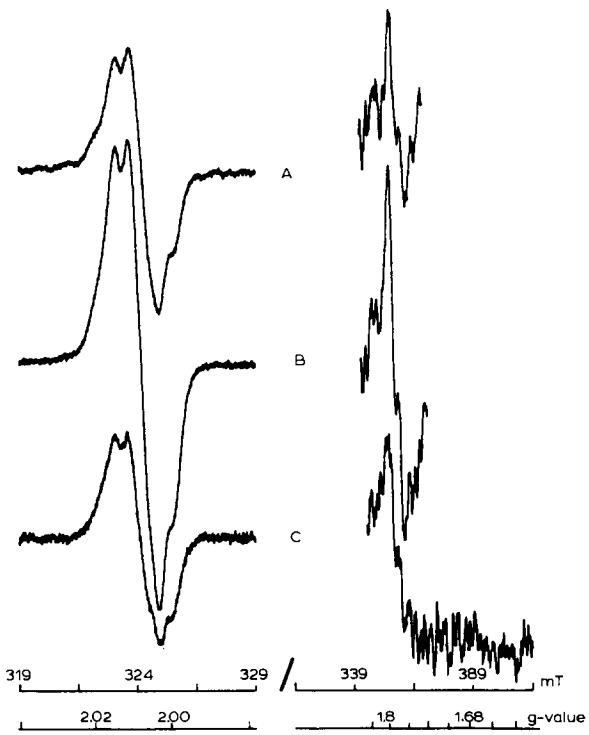


Fig. 2. EPR spectra showing donor and acceptor components at 4.5 K in a sample of intact *C. reinhardii* F54-14 particles frozen under illumination and stored at 77 K in the dark. (A) Before illumination; (B) after illumination at 4.5 K; (C) light-induced spectrum (B–A) showing electron donor (Signal II) and electron acceptor ($Q^-Fe$) spectra. EPR conditions: temperature 4.5 K; microwave power, Signal II 25 μW, $Q^-Fe$ 25 mW; modulation amplitude, Signal II 0.1 mT, $Q^-Fe$ 1 mT.

to the quinone-iron acceptor, and this is demonstrated by Fig. 2. The main type of sample we used to study Signal $II_{lt}$ in unfractionated membranes stored at 77 K or detergent-treated membranes was one where the preparation was frozen under illumination and kept at 77 K in the dark before use. Although this type of sample preparation was developed to eliminate the effects of PS I in our previous experiments [16,18], it was used here to give clear resolution of Signal II and the semiquinone-iron signal in all samples. This consistency was not found in dark-adapted samples as will be shown later. Fig. 2 shows the spectra obtained from a sample of F54-14 detergent particles following this treatment. The figure shows spectra in the *g* 1.82 region representing the semiquinone-iron acceptor, $Q^-Fe$ and in the *g* 2.00 organic radical region for Signal II. Fig. 2A shows the spectra in these regions of a sample after 1 week of dark storage at 77 K following freezing under illumination. Mostly Signal II was observed near *g* 2.00 with a corresponding *g* 1.82 signal indicating the presence of some reduced acceptor, $Q^-Fe$. After illumination at 4.5 K, during which a rapid rise in both signals occurred, the spectra in Fig. 2B were observed. The *g* 1.82 signal of $Q^-Fe$ was now at maximum size and illumination at higher temperatures produced no change in Signal II or $Q^-Fe$. The difference spectra showing the light-induced signals, Signal II and $Q^-Fe$, are shown in Fig. 2C. Measurements on this and other membrane, intact or DEAE particle samples frozen under illumination indicated that the ratios of Signal II to $Q^-Fe$ corrected for baseline changes were about equal in each of the dark, illuminated and difference spectra. This indicated that in this type of sample, all of the positive charge left by electron donation to $Q^-Fe$ could be represented by Signal II. The initial treatment of freezing under illumination produced spectra equivalent to those seen in Fig. 2B, with fully reduced $Q^-Fe$ and oxidised Signal II, the subsequent storage in the dark at 77 K allowing only a partical recombination resulting in spectra as in Fig. 2A. This recombination appeared biphasic as after the initial illumination, storage for a few hours produced significant recombination, but further storage time resulted in only a very slow decrease in the observed signal sizes.

The results show that there appears to be heterogeneity in PS II centres which allows a proportion, about one third, to have efficient electron donation at 100 K or below, and the rest to fuction only above 200 K. Storage at 77 K in the dark presumably favours the recombination of $Q^-Fe$ with the Signal II species donating below 100 K, accounting for the partial recombination behaviour in samples frozen under illumination.

Closer examination of the electron donation at various temperatures indicated that the characteristics of the Signal II donor at temperatures above 200 K closely resembled that of the donor termed D1 studied using optical absorbance measurements by Mathis and co-workers [26–28]. The donation by D1 became slower than the back-reaction from $Q^-Fe$ to P-680$^+$ at about 200 K, and therefore could no longer compete. A similar process can now be directly detected with part of the Signal II component.

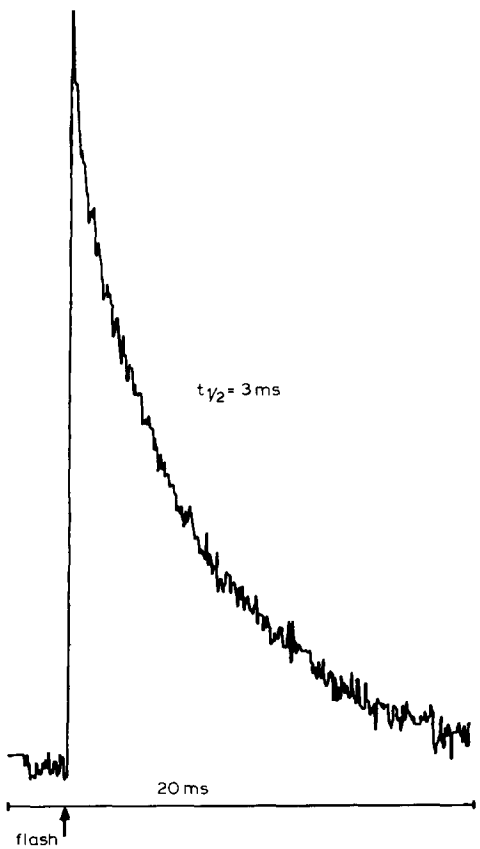


Fig. 3. Kinetics of the back reaction between P-680$^+$ and $Q^-Fe$, monitored by flash illumination of sample at $g$ 2.006. DEAE particles (1 mg/ml) from *C. reinhardii* mutant F54-14 frozen in the dark and an average of the response to 4096, 800-ns laser flashes at 660 nm peak wavelength taken. Spectra were recorded via a Datalab signal averager and transient recorder. EPR conditions: temperature 15 K; microwave power 50 $\mu$W.

*Primary chlorophyll donor P-680*

Apart from the slowly decaying signals mentioned above, rapidly reversible signals were observed near $g$ 2.00 upon laser flash illumination of PS II preparations. Samples frozen under illumination did not show this phenomenon because as discussed earlier the secondary donor, Signal II, was oxidised and acceptor, QFe, reduced, leaving only very rapidly reacting radical pairs such as P-680$^+$-pheophytin which recombined faster than the EPR spectrometer resolution time (50 $\mu$s). However, in samples frozen in the dark, either untreated or reduced with ascorbate prior to freezing, a reversible signal was measured (Fig. 3) on laser flash illumination below 200 K. With secondary donation only available in some centres, the rest were free for the back reaction between P-680$^+$ and $Q^-Fe$ to occur. The decay half-time at $t_{1/2} = 3$ ms was similar to results obtained by other techniques for this back reaction [1,27]. Pre-illumination above 200 K removed the reversible signal as QFe was reduced in all centres due to improved electron donation to the reaction centre. In Fig. 3, the signal monitored near $g$ 2.00 was P-680$^+$ as the $Q^-Fe$ EPR signal was observed at $g$ 1.82 in this sample. The reciprocal experiment of monitoring the decay of the $g$ 1.82 signal was unsuccessful due to the poor signal-to-noise ratio of this signal in kinetic experiments.

*Other g 2.00 radicals*

Effects due to modification of membranes or chloroplasts caused by ageing, freeze-thawing or detergent treatments were seen on PS II donors in samples dark adapted before freezing. The treatments resulted in the loss of the ability to induce Signal $II_{lt}$ by illumination at cryogenic temperatures as occurs in fresh membranes (Fig. 1). Illumination at cryogenic temperatures did give rise to a change near $g$ 2.00, but the new signal had a $g$ value ($g$ 2.003), line width (10–11 G) and line shape characteristics completely different from

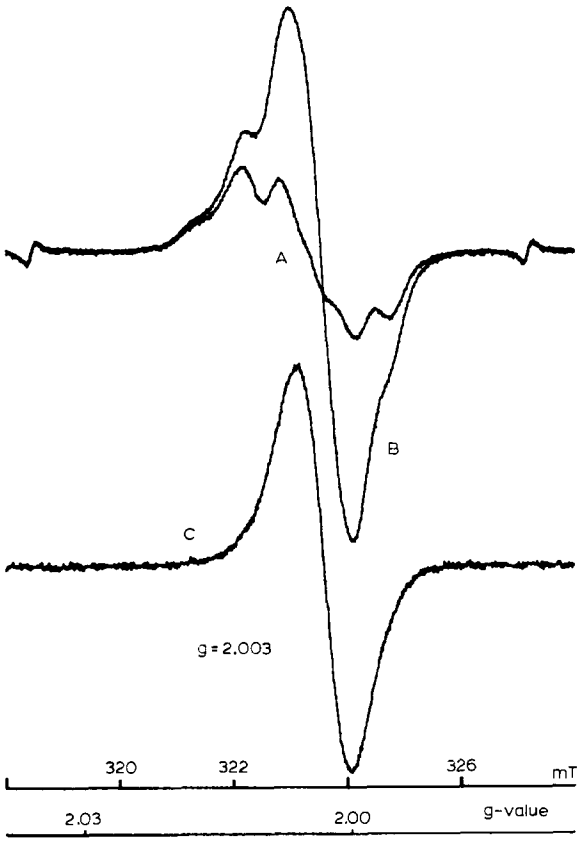


Fig. 4. EPR spectra showing the light-induced radical near $g$ 2 in *C. reinhardii* F54-14 DEAE particles at 77 K. (A) Sample frozen after dark adaptation for 15 min; (B) sample after illumination at 77 K; (C) difference spectrum showing line shape of the 10 G radical. EPR conditions: temperature 77 K; modulation amplitude 0.2 mT; microwave power 50 $\mu$W. The signals at the extreme right and left of the scan are from the $g$ value marker.

those of Signal II. This behaviour of dark-adapted samples also contrasts with the results seen in Fig. 2 where normal Signal $II_{lt}$ formation was observed in samples from these preparations which had been frozen under illumination.

Fig. 4 shows the formation of the 10 G radical at 77 K in F54-14 DEAE-treated particles. During purification of these particles, loss of a part of the Signal II content occurred together with the complete loss of non-reaction centre components as mentioned in Ref. 21. In Fig. 4A (inner line), Signal II was still present in the dark-adapted sample, but after illumination, Fig. 4B (outer line) the line shape changed due to the production of a 10–11 G wide radical revealed by the difference spectrum, Fig. 4C. This signal decayed very slowly during storage at 77 K in the dark at a rate similar to that of Signal II in fresh membranes. The appearance of the $g$ 2.00 radical was correlated with the appearance of the signal of the semiquinone-iron, $Q^-Fe$ at $g$ 1.82. Illumination above 200 K resulted in a further transfer of electrons increasing both the 10 G radical at $g$ 2.003 and the $g$ 1.82 signal. Illumination at 200 K resulted in full reduction of QFe in all stages of purification until the DEAE step where particle modification resulted in the ability to photoreduce only about two thirds of QFe in frozen samples. As with the Signal II donation in freshly isolated membranes, no significant differences were observed in the line shape or $g$ value of the donating species in a particular sample between 4 and 230 K. Therefore, the heterogeneity of electron-transport rates in PS II centres applies to both secondary donors, Signal II and the 10 G radical.

*Cytochrome radicals*

In membranes of *C. reinhardii* F54-14, both the $g$ 6 and $g$ 3 regions contained radical signals attributed to cytochromes, but as shown in Ref. 21 detergent followed by DEAE treatment removed the $g$ 6 radical. This was correlated with the loss of a cytochrome $b_6$-$f$ complex and subsequently the $g$ 6 radical was assigned to cytochrome $b$-563 [21,29,30].

In membranes of F54-14, room-temperature illumination produced changes in line shape of the $g$ 6 radical. These must result from PS II electron transport and may arise from cytochrome $b$-563, which has been suggested by Joliot and Joliot [31] to be linked to PS II electron acceptors in some centres .

The cytochrome $b$-559 present in F54-14 particles and represented by either low or high-spin signals [19,20,29] was not photo-oxidised at cryogenic temperatures, unlike that in barley zb63 chloroplasts [32] or *P. laminosum* oxygen-evolving particles [18]. The consequences of this difference on the low-temperature EPR photochemistry, apart from a lack of change near $g$ 3, were seen in the $g$ 2.00 region. In zb63 chloroplasts [17,32] an experiment, where two periods of illumination at 77 K were given with an intervening period of dark

storage at 77 K, revealed an increased Signal II production during the second illumination period and it was proposed this was due to the irreversible donation by cytochrome *b*-559 during the first illumination being replaced by Signal II donation on subsequent illuminations. This proposal is supported by the absence of this effect following sequential illumination of F54-14 preparations at 77 K, as predicted by the absence of cytochrome *b*-559 donation at cryogenic temperatures.

*Signal II in whole cells*

Further information on Signal II formation was gained using two mutants of *S. obliquus*, LF1 and LF2, shown by Bishop and co-workers [23,24] to have lesions involving small molecular weight changes in only one 34000 molecular weight membrane protein which resulted in the inactivation of the oxygen-evolving system and decrease in the redox potential of high-potential cytochrome *b*-559. The mutant designated LF2 was, however, temperature sensitive and was normal when grown at 18°C, losing oxygen evolution gradually above this temperature until 34°C where it was completely inactive. LF1 was completely devoid of oxygen evolution at all temperatures. Addition of 1 mM manganese to LF2 cultures was found to overcome the temperature-sensitive block, and to restore oxygen evolution at 34°C [23,24]. Fig. 5 shows the results of a series of experiments looking at Signal II in the wild-type, LF1 and LF2 mutants of *S. obliquus*. The spectra shown are from packed whole cells, in each case grown mixotrophically, i.e., under illumination with glucose and yeast extract supplements. These conditions were used to avoid the possible decay of Signal II in the dark due to lack of PS II turnover, and avoided changes caused by cell disruption. The illumination during growth does, however, lead to some 'leakiness' in the lesion as indicated in Ref. 23. Despite this and the presence of both PS I components and manganese, changes in Signal II were observed. The results in Fig. 5 (left) show clearly the presence of Signal II in dark-adapted samples of LF2 at 18°C with much lower amounts at 34°C. The LF1 mutant also has a very low Signal II content. Freezing samples under illumination, Fig. 5 (right), resulted in the generation of P-700$^+$ by PS I turnover, but the Signal II as seen from the shoulder arrowed, again shows that only wild-type and strain LF2 grown at 18°C had significant amounts of Signal II in the light. Fig. 5 (right) C and D shows the degree of leakiness of the mutation mentioned above when cultures were grown under illumination.

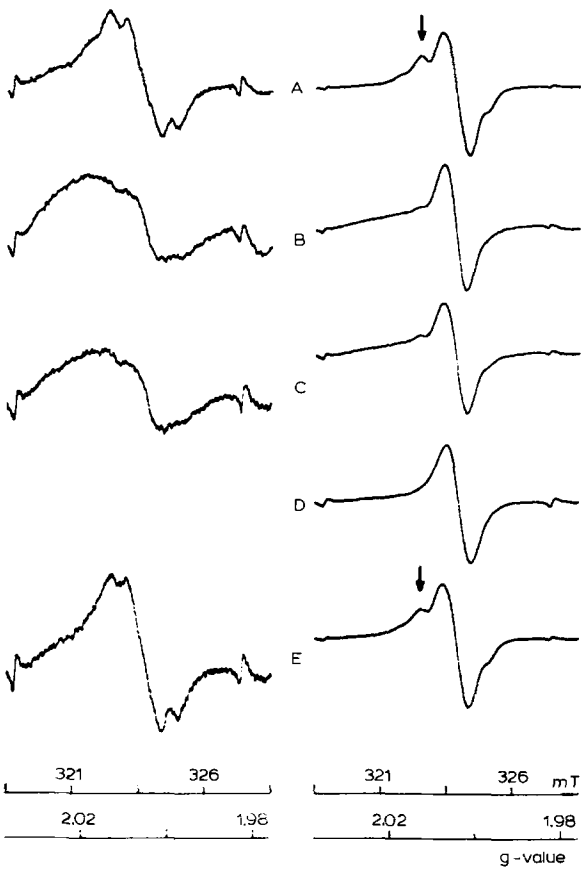


Fig. 5. Signal II content of mutants of *S. obliquus* as shown by *g* 2.0 region spectra of fresh cells either frozen in the dark (left) or under illumination (right). (A) Wild type grown at 34°C, oxygen evolution and Signal II normal; (B) LF1 grown at 34°C, no oxygen evolution; (C) LF2 grown at 34°C, no oxygen evolution; (D) LF2 grown at 34°C in the dark, no oxygen evolution; (E) LF2 grown at 18°C, oxygen evolution and Signal II normal. EPR conditions: temperature 13 K; modulation width 0.2 mT; microwave power 100 μW, *g* value marker peaks are seen on the extreme right and left.

Experiments were then performed to show the effect of growing the LF2 mutant at 34°C, with a supplement of 1 mM manganese chloride during the last 16 h before harvesting. Fig. 6A shows the spectrum of the light-induced signal of the LF2 mutant grown at 34°C without manganese supplement. Fig. 6B shows the same spectrum of the

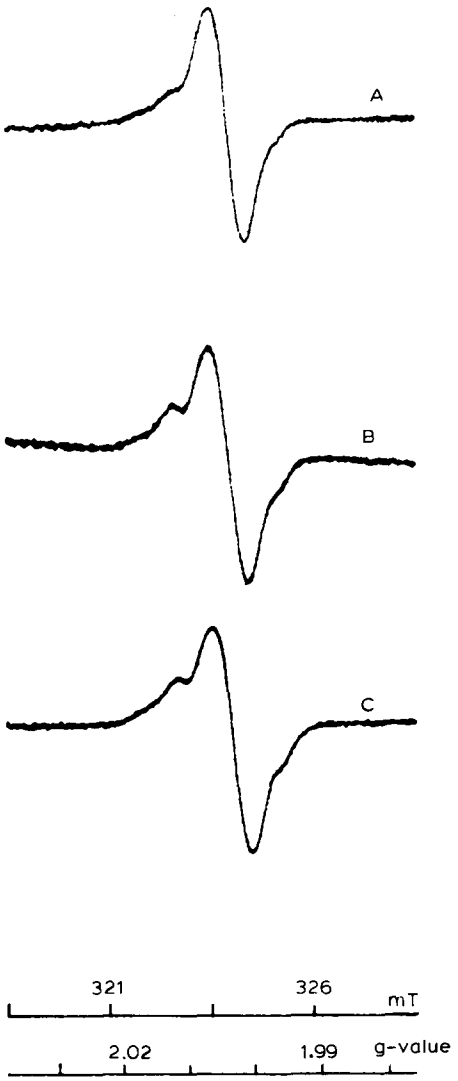


Fig. 6. Effect of manganese supplement in the LF2 *Scenedesmus* mutant. Spectra shown are formed by subtraction of the spectrum of dark-adapted samples and samples frozen under illumination. This removes the distortion caused by manganese signals and shows increased Signal II content in B and C. (A) LF2 grown at 34°C, no oxygen evolution; (B) LF2 grown at 34°C plus 1 mM manganese chloride, oxygen evolution present; (C) LF2 grown at 18°C, oxygen evolution present. EPR conditions: temperature 13 K; modulation width 0.2 mT; microwave power 100 $\mu$W, *g* value marker peaks are seen on the extreme right and left.

mutant grown with the manganese supplement and clearly demonstrates the increased Signal II content. The spectra are presented as difference spectra because the supplemental manganese caused distortion of this region of the EPR spectrum, and therefore dark-adapted samples were used to record baseline spectra, then subtraction of these from the spectra of samples frozen under illumination removed the effects of the manganese signals and dark-stable Signal II. Fig. 6C shows for comparison the spectrum of the LF2 mutant grown at 18°C, where the PS II activity was normal. The result demonstrates that manganese restores the ability of the PS II of LF2 to generate Signal II. Further experiments showed that restoration of oxygen evolution proceeded in parallel with the appearance of the Signal II in this mutant. An increase in the redox potential of high-potential cytochrome *b*-559 accompanying these changes was also found previously [23,24], these results therefore reaffirming the link also noted in previous papers between oxygen evolution, manganese, Signal II and the redox potential of high-potential cytochrome *b*-559 [7,18,23,33–35].

## Discussion

The evidence from a variety of preparations shows that Signal II represents one of the physiological secondary electron donors of PS II. Signal II is thought to arise from an immobilised plastoquinone radical or a derivative of plastoquinone [12–15]. This is supported by the solvent extraction experiments of Kohl and co-workers [13] who proposed that Signal II was a plastochromenoxyl radical. Signal II also has a *g* value similar to that of semiquinones and Hales and Gupta [15] have recently used model semiquinone systems and theoretical parameters to simulate the line shape of Signal II. They conclude that Signal II probably arises from a plastosemiquinone, not protonated but perturbed by a metal cation, possibly $Mg^{2+}$ or $Ca^{2+}$.

Using the model of Hales and Gupta [15] that Signal II arises from a plastosemiquinone stabilised by two positively charged ligands, the EPR effects shown in detergent particles can be accounted for by changes in the quinone complex. The high-potential semiquinone, Signal II, will be more easily affected by redox reagents, and pH conditions could allow replacement of the ligands by, for example, protons. Displacement of protons and ligands to regenerate Signal II cannot occur

once a sample is frozen and would prevent generation of Signal II by illumination at cryogenic temperatures, which we have shown occurs in all but freshly isolated membrane preparations. The 10 G radical formed in place of Signal II does not have a *g* value similar to semiquinone radicals or characteristics of a preceding component on the electron donor chain, so we propose that a high-potential intermediate oxidises adjacent components not usually involved in the donor chain, such as chlorophyll or carotenoid, which could then account for the spectrum obtained. A period of room-temperature illumination in these detergent particles, as shown in Fig. 2. restores the Signal II, probably because exchange of ligands and restoration of the Signal II precursor can occur. Once frozen, in this illuminated sample, no ligand exchange occurs and the Signal II can undergo recombination with acceptor components during storage at 77 K and be reoxidised on subsequent illumination at cryogenic temperatures.

The review of PS II electron donors by Bouges-Bouquet [1] suggested a scheme which involved two donors to P-680 termed $Z_1$ and $Z_2$. The properties of $Z_2$ were correlated with donor D, Signal $II_{vf}$ and Signal $II_f$, whilst the donor $Z_1$, either in series or parallel with $Z_2$, accounted for the fast 30 ns reduction of P-680. The electron donation by Signal II appears too slow to account for $Z_1$ although different kinetic pools exist and there may be a species donating in a time EPR cannot resolve. The properties of Signal $II_{lt}$, although possessing some characteristics of electron donation at cryogenic temperatures attributed to $Z_1$, appear more likely to be due to Signal $II_{vf}$, i.e., $Z_2$, trapped by the low-temperature conditions. This leaves a fast donor functioning between Signal II and P-680 to be positively identified.

The role of high-potential cytochrome *b*-559 as an alternative secondary donor at cryogenic temperatures has been discussed elsewhere [18]. The role of this cytochrome in physiological electron transport is unclear, but may be as part of the link involving other Signal II components between donors and acceptors that allows deactivation of the oxygen-evolving system. The positively charged haem iron may also be used to stabilise anionic species in the same way suggested for other quinone-cytochrome complexes [36]. The link between Signal II, cytochrome *b*-559 and manganese mentioned previously implies a role for manganese in the electron donor chain in PS II. In an earlier paper [18], we suggested that there may be a manganese association with the component responsible for Signal II, but Hales and Gupta [15] suggest that the metal ion involved is probably not a transition metal ion. Therefore, although the properties of both manganese and high-potential cytochrome *b*-559 could be used in the electron donor chain, their exact role has still to be found.

The reasons for the heterogeneity of electron donation shown earlier, or for the multiple species of Signal II needed to account for its kinetic properties, remain to be understood. As with quinone acceptor components, there may be roles for species to operate between semiquinone and quinol, whilst others operate between quinone and quinol via the semiquinone. The different species of Signal II could result from identical components but situated in different environments and therefore having different p*K* values, redox potentials and kinetic properties. These further experiments to understand physiological roles are now possible using the EPR signals of both donor and acceptor components shown in this and accompanying paper.

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