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The influence of quenching by open reaction centres on the Photosystem II fluorescence emission spectrum

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Steady-state room-temperature emission spectra have been determined for spinach and barley (both wild-type and the chlorina mutant lacking Chl *b* and LHCII) thylakoids, a spinach Photosystem II membrane preparation and *Chlorella* cells, under conditions in which the Photosystem II reaction centres were either open or closed. The data show that the fluorescence quenching efficiency of open reaction centres is greatest for those chlorophyll species emitting in the 680–690 nm wavelength range. The reaction centre quenching efficiency decreases at both shorter and longer wavelengths. Calculation of the fluorescence emission ratio spectra for membrane preparations containing LHCII (wild-type barley thylakoids and isolated LHCII) with respect to those without LHCII (chlorina barley mutant thylakoids) indicates that this complex has characteristic emissions around 650 nm (Chl *b*) and 680 nm. The Chl *b* emission is about 2% that of the main Chl *a* emission at 683 nm (open reaction centres). As the characteristic LHCII emission bands near 650 nm and 680 nm are almost absent in the ratio between LHCII emission spectrum and both spinach and wild-type barley thylakoid emission spectra at open reaction centres, it is concluded that most Photosystem II fluorescence is from LHCII when reaction centres are open. Thus, excitation energy does not seem to be preferentially localised in the core antenna complexes prior to fluorescence emission. The data are interpreted in terms of a model in which energy transfer from antenna to reaction centres is basically diffusion limited.

Introduction

It is generally accepted that the antenna of green plant photosystems is made up of 200–300 chlorophyll molecules which transfer excitation energy to RCs, where primary charge separation occurs. Strong evidence exists indicating that, in addition to Chl *b*, which absorbs maximally near 648 nm, at least five different Chl *a* Q_y spectral forms exist in the wavelength range 660–700 nm [1–4]. These are associated with a number of polypeptides, thus comprising the chlorophyll-protein complexes of both antenna [5] and RCs [6]. Assuming a dipole–dipole R^{-6} excitation energy transfer mechanism [7], the average chlorophyll–chlorophyll transfer time is thought to be of the order of 1 ps [8]. The average trapping (charge separation) time for PS II has been shown to be around 300–500 ps [9]. This clearly demonstrates that for PS II there are a large number of transfer steps before trapping by RCs occurs.

In an attempt to describe the energy-transfer relationship between PS II antenna and RCs, several different models have been proposed. Thus, Duysens [10] and Holzwarth and co-workers [11] favour a situation in which there is an extremely fast excited state equilibration amongst the matrix chlorophylls (including RC chlorophyll) and in which energy is transferred into and out of the reaction special pair just as between any other chlorophyll matrix elements. Such a situation would arise if the RC trapping constant were small with respect to the excited state transfer out of the RC

Abbreviations: BBY, Berthold, Babcock, Yocum [16]; Chl, chlorophyll; DCMU, 3-(3,4-dichlorophenyl)-1,1-dimethylurea; F_m, fluorescence yield with reaction centres closed; F_o, fluorescence yield with reaction centres open; F_i, fluorescence at the plateau level of the fast induction phase; LHCII, light harvesting chlorophyll *a*/*b* protein complex II; PS I, Photosystem I; PS II, Photosystem II; RC, reaction centre; Tricine, *N*-tris(hydroxymethyl)methylglycine.

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special pair. This situation is often referred to as the 'trap limited' model.

On the other hand it is often assumed that trapping at RCs is fast with respect to excited state transfer out of the special pair. In this case trapping should occur with a high probability upon excitation of the RC special pair. This model, developed substantially by Butler and co-workers [12,13], is commonly referred to as the 'diffusion limited' case.

General expressions have been developed to describe both these extreme situations [14]. It is interesting to note that the energy-transfer characteristics in the antenna matrix (hopping rate) influence the overall trapping efficiency only in the diffusion-limited case and not in the trap-limited one.

Evidence has recently been published, based on the measurement of the fluorescence quenching efficiency of open RCs, suggesting that the PS II trapping efficiency is a function of the excitation wavelength in the range 645–700 nm [15]. The trapping efficiency was reported to be greatest around 683 nm and to decline at both longer and shorter wavelengths. Such evidence is supportive of the diffusion-limited model for PS II. In the present study we have sought to extend these observations by measuring fluorescence quenching due to open RCs as a function of emission wavelength from 650 nm to 710 nm in a variety of plant materials. The data are interpreted in terms of a substantially diffusion-limited PS II model in which the external antenna makes a major contribution to PS II fluorescence, particularly when RCs are open.

Materials and Methods

BBY-grana were prepared from freshly harvested spinach leaves by the method of Berthold et al. [16] but omitting the last Triton X-100 treatment [17]. This preparation contains both LHCII and the PS II core pigment-protein complexes and is free of PS I complexes as demonstrated by mild SDS-gel electrophoresis (unpublished observation). The chlorophyll recovery yield was typically about 30%. Final resuspension was in 30 mM Tricine (pH 8)/10 mM NaCl/5 mM $MgCl_2$/0.2 M sucrose.

LHCII, the principal PS II antenna complex, was prepared from spinach leaves according to Ryrie et al. [18]. Final resuspension was in a medium containing 0.05 M sucrose and 5 mM Tricine (pH 8). The chlorophyll *a*/*b* ratio was 1.1.

Thylakoids were prepared from freshly harvested leaves of spinach or barley (both the chlorina mutant lacking LHCII and its wild type) as previously described [4]. The final resuspension was in 30 mM Tricine (pH 8)/10 mM NaCl/5 mM $MgCl_2$.

Chlorella vulgaris (SAG 30.80) cells were grown in a medium similar to that described by Hase and Morimura [19] with the following additions: EDTA (disodium salt) 37 mg/l, $Co(NO_3)_2 \cdot 6H_2O$ 0.04 mg/l; the final pH was 5.6. Growth conditions were as described by Hase and Morimura [19] in the 'Preculture and separation etc.' section and yielded a uniform population of mature, non-dividing cells.

The F_o and F_m fluorescence levels were measured as previously described [20]. F_m was determined in the presence of 25 μM DCMU and 2 mM hydroxylamine, while F_o was measured in the same sample but in their absence.

Fluorescence emission spectra were measured with an EG&G OMA III (model 1460) multichannel spectrometer. The resolution of this apparatus was about 0.5 nm/pixel. Excitation light was provided by a Heath monochromator (excitation wavelength either 480 nm or 440 nm; FWHM 1.2 nm) combined with two Corning 4-96 filters. The fluorescence emission was filtered across a Schott OG 530 filter. This arrangement ensured that stray, scattered light was not significant at wavelengths above 650 nm, as judged by using the fluorescence quencher, dibromothymoquinone (280 μM). Total counts accumulated in the peak channel of each spectrum were: 10^4 for closed trap spectra and $3 \cdot 10^3$ for LHCII spectra. Unless otherwise stated in figure legends, 60 emission spectra were averaged and smoothed according to Savitzsky-Golay [21]. Fluorescence was maintained near the F_o level by means of a weak excitation beam and with continual sample stirring in which only a small part of the sample was illuminated at any one time. Thylakoids were illuminated in the presence of 0.1 mM methylviologen. Under these conditions the fluorescence level attained for *Chlorella* cells and for the thylakoids was that of the plateau (F_i) of the fast induction phase. Emission spectra with closed RCs (F_m) were measured in the same sample after addition of 25 μM DCMU and 2 mM hydroxylamine.

Results

We have measured the fluorescence induction parameters (F_o, F_i, F_m) in both spinach thylakoids and the spinach BBY-grana preparation at several different emission wavelengths (Table I). The data indicate that both the F_m/F_o and F_m/F_i ratios are greater in the 680–690 nm region than around 650 nm. This seems to suggest that those chlorophyll molecules emitting in the 680–690 nm region are more strongly quenched by open RCs than those emitting at shorter wavelengths.

In order to investigate this phenomenon further, we have measured the chlorophyll fluorescence emission spectra of a variety of plant materials under conditions in which PS II RCs are either mostly open or closed (see Materials and Methods). Fig. 1 shows the ratio of these spectra for spinach thylakoids and the spinach

TABLE I

Ratios of the fluorescence induction parameters at different emission wavelengths in spinach thylakoids and BBY-grana

The excitation wavelength was 450 nm (Oriel 450 nm interference filter, FWHM 8 nm, plus Corning filter 4-96). The emission wavelengths were obtained with Baltzers interference filters (FWHM 8 nm) in combination with a Schott OG 530 filter. The small residual signal due to stray light (less than 1%) was determined in the presence of the chlorophyll fluorescence quencher dibromothymoquinone (280 μM) and was subtracted from the fluorescence measurements.

		Emission wavelength (nm)		
		650	681	691
Spinach thylakoids	F_m/F_o	4.78	5.02	5.08
	F_m/F_i	3.27	3.38	3.41
BBY-grana	F_m/F_o	3.44	–	3.60
	F_m/F_i	2.34	–	2.40

BBY-grana preparation. In both cases the ratio spectra have a distinct minimum value between 680 and 690 nm, indicating that the fluorescence quenching by open RCs is greatest for chlorophylls emitting at these wavelengths. In both cases these minima are red-shifted with respect to the fluorescence emission peak (near 683 nm, data not shown). In the case of the BBY-grana the shift is about 7 nm, while for the thylakoids it is around 2 nm. This difference between the two preparations may well be due to the presence of a minor, long-wavelength emission associated with PS I in the thylakoids but not in the BBY-grana (see Discussion for further details). On the short-wavelength side of the minima both spectra clearly show some internal structure. Thus, the emission ratio decline is rather steep in the 650–665 nm interval and somewhat less steep in the 665–685 nm interval.

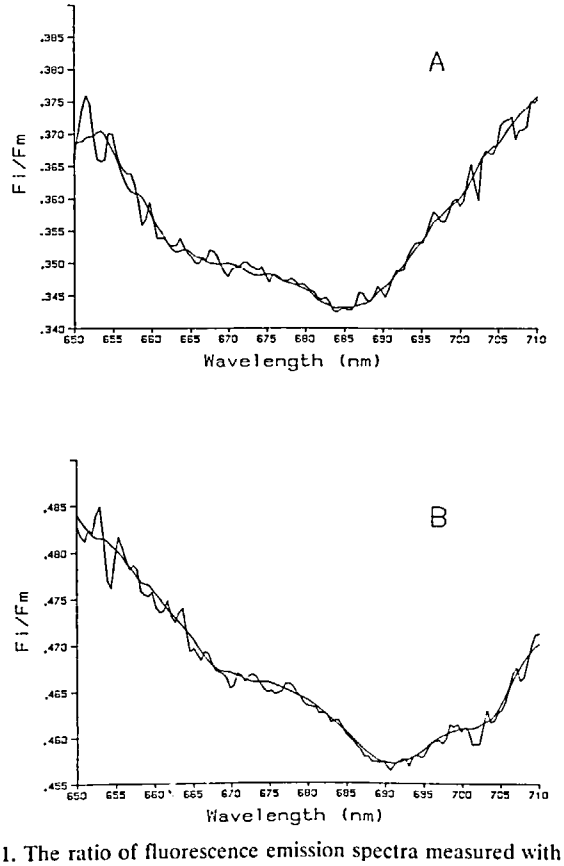

Fig. 1. The ratio of fluorescence emission spectra measured with PS II reaction centres open to that measured with closed RCs in spinach. (A) Thylakoids (45 spectra) and (B) the BBY-grana preparation (37 spectra). Both smoothed and nonsmoothed ratio spectra are presented. The excitation wavelength was 480 nm. For further details see Materials and Methods.

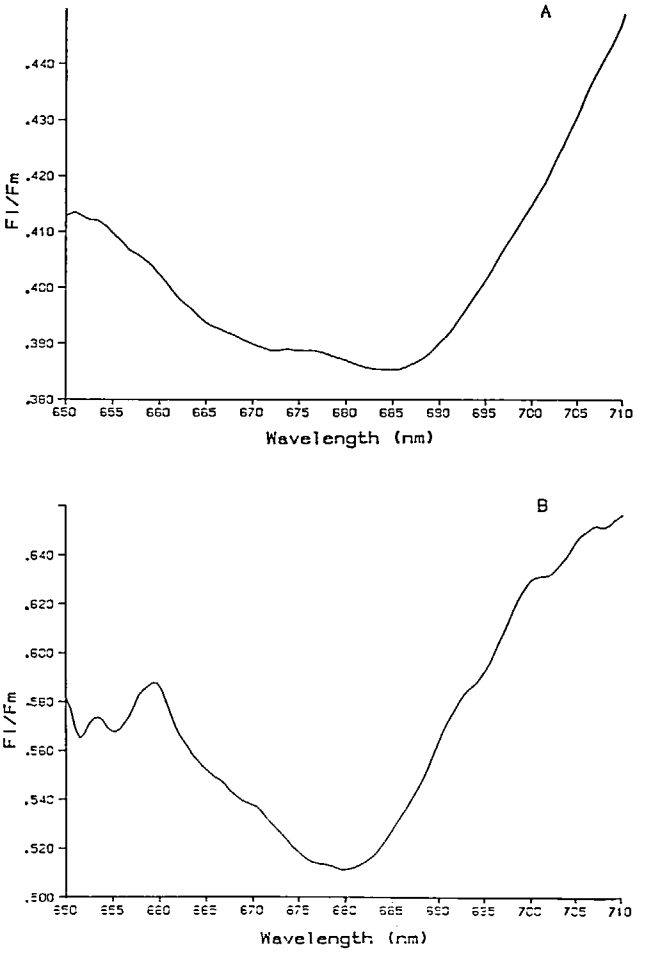

Fig. 2. The ratio of fluorescence emission spectra measured with PS II reaction centres open to that measured with closed RCs in barley thylakoids. (A) Wild-type; (B) chlorina mutant. The excitation wavelength was 440 nm. For further details see Materials and Methods. Signal-to-noise ratios are similar to those presented in Fig. 1.

In Fig. 2 the open-trap to closed-trap emission ratio spectra for thylakoids from both wild-type and the chlorina barley mutant lacking Chl *b* and LHCII are presented. The wild-type spectrum shows the characteristic dip described above for spinach, with a minimum value at approx. 685 nm. On the other hand, while the emission ratio spectrum of the mutant has a

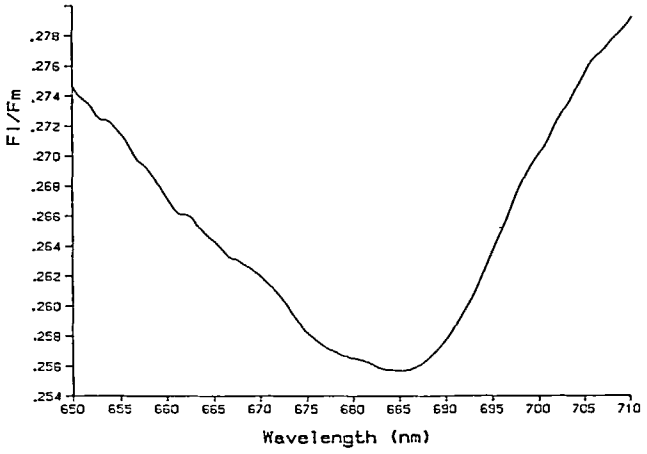

Fig. 3. The ratio of fluorescence emission spectra measured with PS II reaction centres open to that measured with closed RCs in *Chlorella* cells (48 spectra). The excitation wavelength was 480 nm. For further details see Materials and Methods. Signal-to-noise ratios are similar to those presented in Fig. 1.

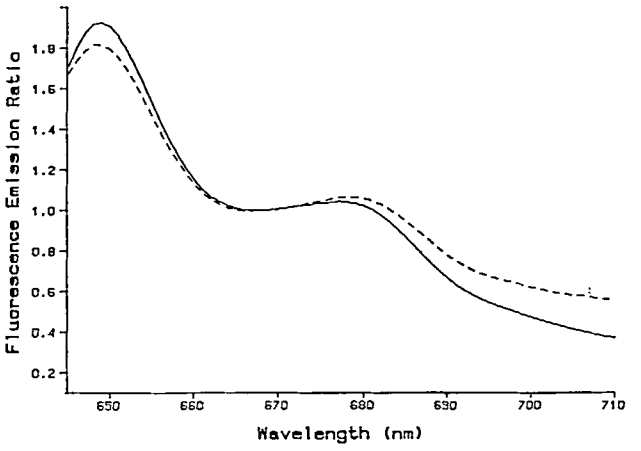

Fig. 4. The fluorescence emission ratio spectra for: (a) wild-type barley thylakoids with open RCs (broken line) and (b) isolated LHCII (full line), both with respect to the chlorina barley mutant with open RCs. The excitation wavelength was 440 nm. For further details see Materials and Methods.

similar overall structure, there are some differences in detail. Thus, the minimum at around 680 nm is slightly blue-shifted with respect to the fluorescence emission peak. This feature may be associated with a greater PS I emission in the mutant with respect to the wild type (see Discussion). It has been shown that the chlorina barley mutant has a much higher PS I/PS II antenna chlorophyll ratio than the wild type [22]. In addition, the steep decline between 650 and 665 nm is greatly reduced, with no apparent decrease in the 650–660 nm region.

In Fig. 3 the open-trap to closed-trap emission ratio spectrum for mature, non-dividing *Chlorella* cells is presented. This shows the characteristic dip, with a pronounced minimum around 685 nm. Similar data have been obtained for dividing *Chlorella* cells (data not presented).

To facilitate interpretation of the preceding open-trap to closed-trap ratio spectra we have sought to understand whether there is a major contribution to PS II fluorescence by the principal (external) antenna complex, LHCII, at room temperature. To this end we have calculated the ratio between the emission spectrum at open RCs in wild-type barley thylakoids with respect to that in the chlorina mutant (Fig. 4). The ratio spectrum decreases steeply between 650 and 665 nm and has a low peak near 680 nm. These data seem to indicate an enriched LHCII emission around 650 nm and 680 nm with respect to the total PS II emission. This conclusion is supported by the data presented in Fig. 4, where it is shown that the emission ratio of isolated LHCII (spinach) with respect to the open trap emission of the chlorina barley mutant is very similar to the above-mentioned wild-type to chlorina mutant emission ratio spectrum. Further support comes from the data presented in Fig. 5, where is can be seen that the ratio spectra for LHCII emission with respect to that of both wild-type barley and spinach thylakoids (open RCs) are substantially constant in the range 650–680 nm. These data seem to indicate that the external antenna complex, LHCII, makes a major contribution to the room-temperature fluorescence of PS II when RCs are open.

It should be pointed out that when the LHCII emission spectrum is compared with that of both spinach and barley thylakoids at the F_m level (closed reaction centres), small differences are observed with respect to those reported in Fig. 5 for the F_i level (unpublished data). These differences are due to the variations in the F_i/F_m ratio spectra reported in Figs. 1 and 2.

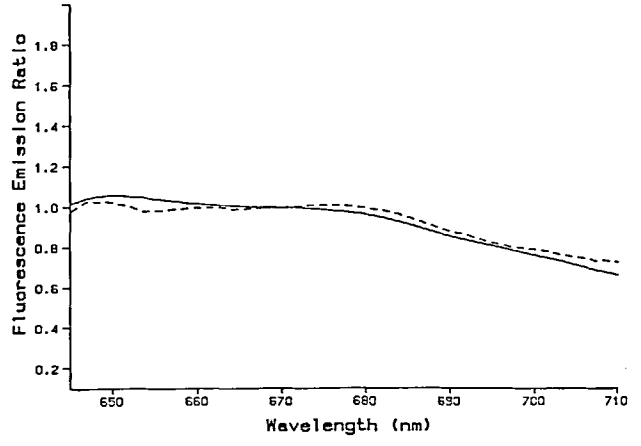

Fig. 5. The fluorescence emission ratio spectra for LHCII with respect to thylakoids with open RCs. Full line: wild-type barley thylakoids; broken line: spinach thylakoids. The excitation wavelength was 440 nm and 480 nm, respectively. For further details see Materials and Methods.

Discussion

In the present paper it is demonstrated that the efficiency of fluorescence quenching by open RCs increases progressively with increasing emission wavelength in the range 650 nm to 680–690 nm in a variety of plant materials. At wavelengths above 680–690 nm this parameter rapidly decreases. In a previous communication from this laboratory [15] it was shown that the maximal fluorescence quenching by RCs in the BBY-grana preparation and in thylakoids of the chlorina barley mutant lacking Chl *b* and LHCII was at excitation wavelengths near 683 nm and 675 nm, respectively. In this paper the RC quenching of fluorescence emission has been found to be maximal for these two preparations at around 690 nm and 680 nm, i.e., red shifted by about 7–5 nm with respect to the excitation RC-quenching peak. As this value is very similar to the Stokes shift for PS II fluorescence in thylakoids, it is concluded that there is excellent agreement between the two experimental approaches. Thus, it would seem reasonable that those chlorophyll species which emit fluorescence in the 680–690 nm range transfer energy to PS II RCs with greater efficiency than those chlorophyll forms emitting at both longer and shorter wavelengths. By comparison with our earlier fluorescence excitation study [15], we suggest that in spinach PS II this high transfer efficiency to RCs is most closely associated with the 684 nm Chl *a* absorption spectra form.

In order to be sure of this interpretation it is necessary to demonstrate that the structure of the open-trap to closed-trap emission ratio spectra is not caused by some kind of heterogeneous fluorescence emission not associated with the single PS II antenna-RC units. Several kinds of such emission heterogeneity may be considered.

(1) PS I–PS II. While almost all the room temperature emission from thylakoids is PS II in origin, a weak PS I fluorescence, peaking between 710 and 720 nm, exists [23]. Such an emission is expected to increase very little upon closure of PS II RCs and would thus have the effect of apparently increasing the open-trap to closed-trap emission ratio. This may therefore explain part of the steep rise in the emission ratio at wavelengths greater than 680–690 nm in the thylakoid and *Chlorella* spectra. However, it is not expected to be the case in the BBY-grana preparation, in which PS I is almost absent [22,15].

(2) PS II antenna heterogeneity. It is well known that green plant thylakoids are thought to contain two PS II populations, differing in their antenna properties [24–26]. Thus, PS II-β units seem to contain low amounts of LHCII, while this complex is the major antenna complex in PS II-α units. It seems unlikely that this factor contributes significantly to our results, as the characteristic open trap to closed trap fluorescence ratio dip is observed in both the BBY-grana preparation and the chlorina barley mutant thylakoids, systems considered to contain respectively only PS II-α units and PS II-β units [22].

(3) Antenna complexes detached from PS II units. It is possible to envisage a small population of fluorescing chlorophyll-protein complexes not assembled into the PS II antenna matrix. Such complexes would lead to an apparent increase of the open-trap to closed-trap emission ratio. It is, however, rather difficult to imagine that such a situation might give rise to the characteristic fluorescence ratio dip in a variety of membrane systems including thylakoids of the chlorina barley mutant which does not contain the major antenna complex, LHCII. In addition, we have noticed with *Chlorella* cells that the dip in the emission ratio spectrum is not significantly different in dividing cells, when the concentration of the hypothetical non-inserted antenna complexes might be expected to be greater, than in mature, non-dividing cells.

By comparing the open-trap emission spectrum of the chlorina mutant thylakoids with that of either wild-type barley or isolated LHCII, it has been possible to identify two spectral zones in which there is a preferential LHCII emission with respect to that of the total PS II chlorophyll matrix. Thus, LHCII fluorescence, both in vivo and in vitro, is relatively greater around 650 and 680 nm. The '650 emission' is particularly interesting, as it would seem likely that this is associated with Chl *b*. The emission ratio spectrum peak for this component is near 649 nm (Fig. 4), which is only slightly red-shifted with respect to the Chl *b* absorption band in vivo, obtained by absorption spectrum deconvolution [4]. However, the wild-type barley minus chlorina mutant fluorescence difference spectrum peaks at 654 nm (unpublished data), i.e., red-shifted by about 6 nm with respect to the Chl *b* absorption. The stokes shift calculated from the 648 nm absorption band using the Stepanov [27] relation (temperature 300 K) is about 4 nm.

It should be pointed out that fluorescence emission at 650 nm is rather weak, being only about 5% that of the peak value (near 683 nm) in normal Chl-*b*-containing thylakoids and in isolated LHCII. In the chlorina barley mutant lacking Chl *b*, emission at 650 nm is about 3% of the peak value. Thus, we conclude that, around 650 nm, the Chl *b* emission is roughly equivalent to 2% of the Chl *a* peak value. If one considers a Boltzmann-weighted population of excited states for Chl *b* (648 nm) and Chl *a* (676 nm) in which thermal equilibrium is attained prior to fluorescence emission, a value of approx. 1:20 is obtained. Taking into account the lower fluorescence yield of Chl *b* with respect to Chl *a* [28] and their relative concentrations in PS II antenna (about 0.5), a Chl *b* fluorescence emis-

sion of around 1% that of Chl *a* is expected. Thus, the measured value of approx. 2% at the emission peaks does not seem at all unreasonable.

As mentioned above, the fluorescence emission associated with Chl *b* has a maximum ratio spectrum value near 649 nm and declines rapidly with increasing wavelength up to 665 nm. It is therefore reasonable to suggest that the rapid decline in the open-trap to closed-trap emission ratio spectra between 650 nm and 665 nm for spinach BBY-grana, spinach and wild-type barley thylakoids and possibly also *Chlorella* cells is associated mainly with Chl *b*. This suggestion is strengthened by the absence of any such decline in the open-trap to closed-trap ratio spectrum of the chlorina barley mutant. These data therefore suggest that Chl *b* is the least efficient of the various chlorophyll forms in transfering energy to PS II RCs. The fact that the open-trap to closed-trap ratio spectra decline continuously also at wavelengths greater than 660 nm suggests that the energy transfer efficiency to RCs is a function of the absorption and emission properties of the chlorophyll spectral forms. This situation is most clearly exemplified in the chlorina barley mutant thylakoids, where Chl *b* is absent. Thus we conclude, consistent with our earlier suggestion [15], that energy transfer to RCs is essentially determined by the transfer microparameters of the donor-acceptor antenna chlorophylls.

By comparing the emission spectra of both spinach and wild-type barley thylakoids (PS II RCs open) with that of LHCII (Fig. 5), it is reasonable to conclude that it is in fact LHCII which is the major emitting complex in the 650–680 nm interval. As LHCII is generally considered to be the main external antenna complex of PS II [5] this observation suggests that excitation energy is not concentrated in the core complexes prior to fluorescence emission when RCs are open. Such a conclusion is therefore not in close agreement with the so-called 'funnel' concept of antenna organisation [29–31] in which the short wavelength chlorophyll forms are considered to be more distant from RCs than the longer wavelength forms. This organisation is proposed to 'direct' excitation energy flow preferentially towards the RCs.

As discussed above, it is demonstrated here that the PS II fluorescence emission when RCs are open is spectrally different from the fluorescence emission when RCs are closed. This suggests that the open-trap steady-state fluorescence emission does not represent an equilibrium distribution of excited states within the antenna matrix. Such a situation is not in agreement with a PS II transfer model which is substantially 'trap-limited', as this would give rise to fluorescence emission spectra which are independent of the redox state of the RCs. Thus, the present data favour the 'diffusion-limited' PS II energy-transfer model.

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