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The emission spectra of thermoluminescence from the photosynthetic apparatus

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Emission spectra of two thermoluminescence (TL) components emitted from the photosynthetic apparatus were determined by the use of an imaging photon detector system. The following results were obtained: (i) The TL B-band emitted from whole thylakoids and PS II core complex showed the same emission spectrum peaking at about 690 nm (uncorrected for wavelength dependence of the photocathode) with a broad tailing in far-red region. This spectrum agrees with the reported emission spectrum of delayed luminescence, consistent with the view that the TL B-band arises from thermally stimulated recombination of charge pairs in PS II. (ii) The emission spectrum of TL Z-band was variant, depending on the PS I/PS II ratio in the sample. This was because the Z-band consists of two spectral components emitting at about 740 nm and 690 nm (also uncorrected for the wavelength dependence of the photocathode); the former originates exclusively from PS I and the latter from PS II. (iii) Light-harvesting chlorophyll protein complexes (LHCI and LHCII) emit the respective spectral components of Z-band more strongly than do their respective core complexes, indicating that reaction center photochemistry is not involved in energy storage for the Z-band. (iv) A methanol extract of thylakoids emitted only a weak Z-band at 690 nm, but the formation of a chlorophyll aggregate markedly enhanced the Z-band intensity, concomitant with a shift in emission maximum to 740 nm. The mechanism of energy storage for Z-band is discussed in relation to the local chlorophyll concentration.

Introduction

Plant thermoluminescence (TL), the phenomenon whereby preilluminated and frozen thylakoids emit light when heated, was discovered by Arnold and Sherwood in 1957 [1]. Subsequent studies have indicated that seven or more TL components are emitted at different temperatures under different excitation conditions [2]. Of these TL components, the B-band emitting at around 30 °C has been best characterized: it arises from recombination of charge pairs, $S_2Q_B^-$ and $S_3Q_B^-$, generated by Photosystem (PS) II photoreaction (Ref. 3; cf. theory, Ref. 4). Note that the positive charges, S_2 and S_3, are two- and three-step oxidized intermediate states of water oxidation enzyme system and the negative charge, Q_B^-, is a one-electron reduced form of the secondary quinone acceptor of PS II. Similar identification of charge pairs responsible for several other TL components has been reported [2,5]. Based on these assignments, several groups have been vigorously progressing characterization of the linear four-step reaction involved in the oxygen-evolving system using TL measurement as a probe for PS II photoreactions [5].

The Z-band emitted at around −160 °C, however, can be observed with boiled leaves or isolated chlorophyll [6], so it has been inferred that this band is not related to photosynthetic charge separation or electron transfer. The Z-band was reported to have an emission peak at around 740 nm [6,7], and its origin was suggested to be the triplet state of chlorophyll [6]. These results were, however, obtained by measuring the luminescence from intact leaves and inevitably strong self-absorption, so that the determined emission spec-

Abbreviations: PS, Phtosystem; FL, fluorescence; TL, thermoluminescence; DL, delayed luminescence; DCMU, 3-(3,4-dichlorophenyl)-1,1-dimethylurea; Mes, 2-(*N*-morpholino)ethanesulfonic acid.

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trum contains ambiguities, particularly in the 670–690 nm region. As to the other TL components, no emission spectra have ever been reported. A more reliable emission spectrum has been for delayed luminescence (DL), which is also an emission due to charge recombination in PS II [8,9], and the spectrum is essentially the same as that of fluorescence [10]. Although TL glow curves provide unique information about the activation free energy of various types of charge recombination, they do not contain any information concerning the pigment species emitting light: from the glow curves alone, it is impossible to tell whether a certain TL band consists of a single spectral component or several spectral components having the same emission (recombination) temperature. Thus, determination of the emission spectra of TL bands is useful for a better understanding of the TL phenomenon as well as for its better application as a probe for PS II photoreactions.

The difficulty in determining the emission spectrum of TL is due mainly to its very weak intensity. The yield of TL emission has not yet been determined, but it is expected to be weaker than that of DL, which is about 1/100 of fluorescence [11]. In this study, two TL components, the B-band and the Z-band, were detected by an imaging photon detector system. By integrating the resulting two-dimensional image data with respect to wavelength, the emission spectra of the two TL bands have been unambiguously determined for (to our knowledge) the first time with various preparations of PS I and PS II.

Materials and Methods

Spinach thylakoids were prepared as described in Ref.12. Crude PS I and PS II fragments were prepared by French pressure cell disruption according to Sane et al. [13]. LHC I, and PS I core complex devoid of LHC I were prepared according to Haworth et al. [14]. LHC II, and PS II core complex devoid of LHC II were prepared by the method of Burke et al. [15] and Enami et al. [16], respectively. Cyanobacterial PS II particles of *Synechococcus vulcanus* Copeland were prepared by the method of Koike et al. [17]. The chlorophyll solution was prepared by extracting spinach thylakoids with methanol after one wash with hexane to remove quinones and some carotenoids.

The samples were suspended in 25% (v/v) glycerol, 10 mM $MgCl_2$ and 50 mM Mes-NaOH (pH 6.5) at 0.2–0.5 mg Chl/ml. The TL glow curves were measured as described previously [12]. The excitation conditions for B-band measurements were as follows. Samples were exposed to continuous light for several tens of seconds, dark-adapted at room temperature for several minutes, and then stored in darkness in at ice temperature. An aliquot of the relaxed sample was excited by two flashes, followed by quick cooling in liq. N_2. Durations of the continuous illumination and the subsequent dark-adaptation were typically 45 s and 7 min, respectively, for spinach preparations. For the *Synechococcus* PS II particles, dark-adaptation of 45 min was employed. These protocols maximized the TL B-band intensity after two flash excitation by modulating the initial ratio of $S_2 + S_3/S_0 + S_1$ states and Q_B^-/Q_B [18].

For spectrum measurements, the light emission from a sample spread on a 2×2 cm^2 filter paper was focused onto the slit of a polychromator (low dispersion monochromator), and the dispersed spectrum image was recorded with an imaging photon detector system (Surface Science Instruments, Model 2601A). In this system, photoelectrons emitted from the red-sensitive photocathode (MA3) were multiplied by five layers of microchannel plate electron multipliers, and multiplied electron pulses hitting the resistive anode encoder were registered into a position computer. The photon counts at each position (1024×1024 points) were stored and processed with a personal computer. The resulting two-dimensional image data were then integrated towards the y-axis (i.e., slit height direction) with respect to the x-axis (i.e., wavelength) to obtain an emission spectrum. The wavelength was calibrated by using the band maxima of an He/Ne laser and several interference filters as wavelength standards. The obtained emission spectra were directly presented without correction for wavelength dependence of the photocathode. By the use of a mechanical shutter placed before the polychromator, a particular TL component was selectively measured: between 0°C to 57°C for B-band and −180°C to −120°C for Z-band measurements. For Z-band measurement, sample warning was started 90 s after excitation to avoid the influence of phosphorescence. The measurements were repeated 5 to 24 times and the accumulated data were presented. In case of fluorescence measurements, the sample was illuminated through an optical guide with a light beam from a tungsten lamp passing through a Corning CS 4-96 filter, a 487 nm interference filter and a 15 cm layer of water. The photocathode of the imaging photon detector was protected with a red dichroic filter (Nihon Shinku Co., Japan). To measure the emission from the isolated chlorophyll, the methanol/water solution of the thylakoid extract was placed in a thin flat container (1 mm in depth) fixed to the sample heating unit for TL measurement.

Results

Spectra a, b and c in Fig. 1 show the emission spectra of TL B-band emitted from spinach thylakoids, spinach PS II core complex and *Synechococcus* PS II particles, respectively. Upon warming the illuminated and then frozen samples, typical glow curves were observed, each exhibiting B-band emission at around 30°C (Fig. 2A).

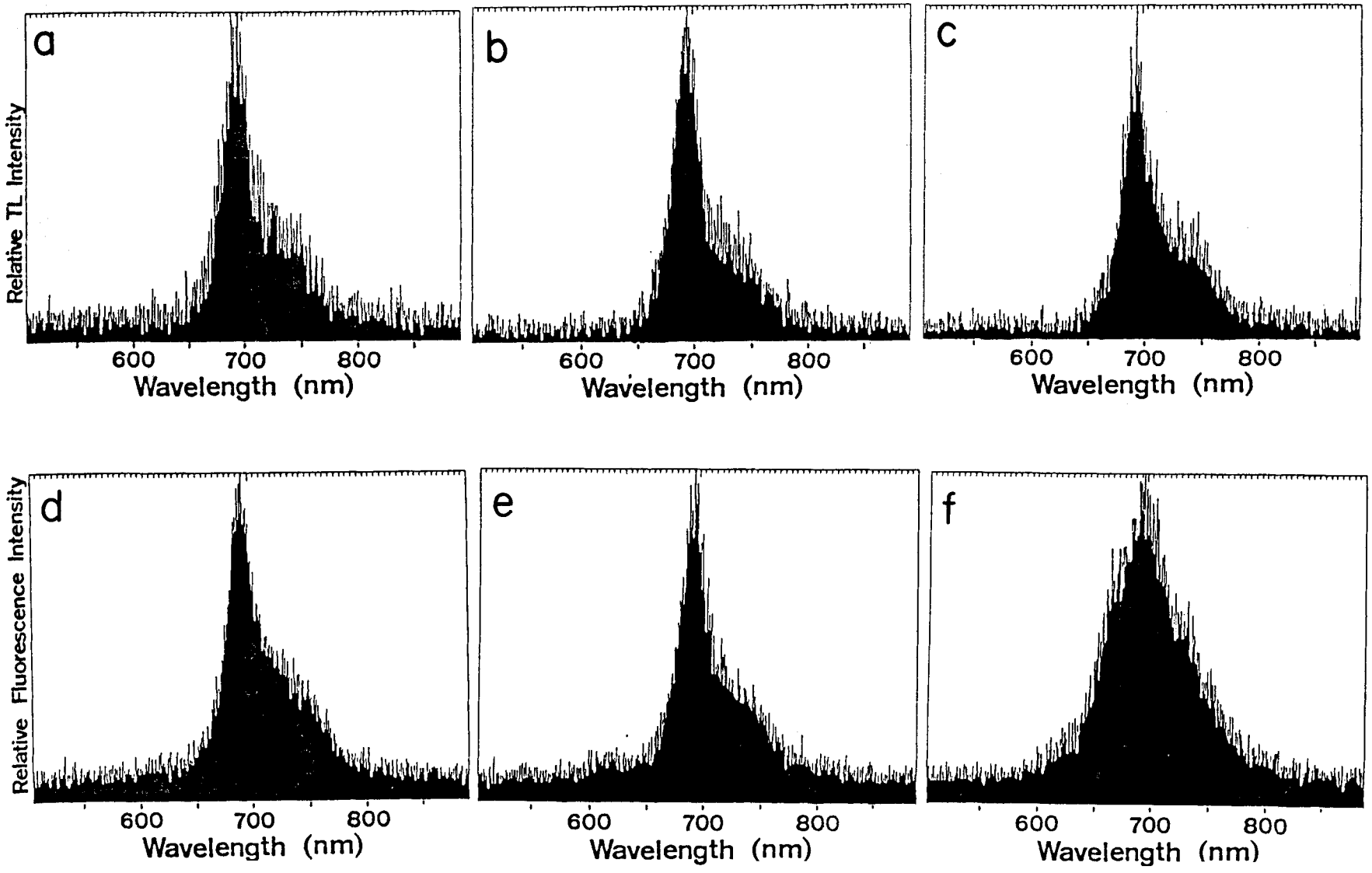


Fig. 1. Emission spectra of thermoluminescence B-band (a, b, c) and fluorescence (d, e, f) from spinach thylakoids (a, d), purified spinach PS II core complex (b, e) and PS-II-enriched particles of *Synechococcus* (c, f), respectively. Thermoluminescence photons were collected during heating from 0 °C to 57 °C, and fluorescence was measured at room temperature. Chl concentrations were 0.2 mg/ml for all samples.

By use of a mechanical shutter, the photon emission during warming from 0 °C to 57 °C was collected. The resulted TL B-band emission spectra from the three samples similarly exhibited a sharp maximum at 690 nm accompanied by an appreciable shoulder in far-red region (Fig. 1a, b, c). These TL spectra are identical or highly similar to the FL spectra of the three respective samples (Fig. 1d, e, f) measured by the same instrumental arrangement under very weak blue excitation (487 ± 13 nm), except for the appreciable fluorescence emission from phycobilins in cyanobacterial PS II (Fig. 1f).

From comparison between these TL and FL emission spectra, the following points are made. (i) The TL spectrum from thylakoids (a) is identical to that from PS II core complex (b) which is completely devoid of PS I. This can be interpreted that PS I is not involving in TL B-band emission and that the features at around 740 nm is not due to the contribution by PS I, but is attributable to the second vibrational band of the same chlorophyll species that emit at 690 nm. (ii) The TL emission spectra of thylakoids and PS II core complex (a, b) are identical to the FL emission spectra (d, e) of respective samples. This implies that the TL B-band arises from the chlorophyll species that are responsible for emission of DL. Note that DL has been revealed to show the same emission spectrum as FL (see, for example, Ref. 10). (iii) The TL emission spectrum from the cyanobacterial PS-II-enriched preparation (c) differs appreciably from its corresponding FL emission spectrum (f), but is identical to the TL spectrum of spinach thylakoids or PS II core complex (a, b). This implies that phycobilins remaining in the cyanobacterial preparation are not involved in TL emission, i.e., migration of the energy released by charge recombination is restricted within the chlorophyll molecules located in the vicinity of PS II reaction center. All these characteristics are consistent with the view that the TL B-band arises from recombination of $S_2Q_B^-$ or $S_3Q_B^-$ charge pairs in the PS II reaction center [2–5].

Spectra a, b, c in Fig. 3 show the emission spectra of the TL Z-band emitted from thylakoids and two membrane fragments enriched in PS I and PS II, respectively. Samples were illuminated with continuous white light for 5 min at 77 K, followed by relaxation in darkness at 77 K to produce phosphorescence decay. In contrast to the B-band, the Z-band required 5 min continuous illumination for full charging. Upon warming, typical glow curves exhibiting the Z-band at around −160 °C were obtained (Fig. 2B). Photons emitted during warming from −180 to −120 °C were collected. The emission spectrum of the TL Z-band thus obtained for thylakoids showed a major maximum at 740 nm with a very faint shoulder at around 690 nm (a). Comparison of this TL emission spectrum with the 77 K FL spectrum (d) of the same sample reveals that the emission intensity at 690 nm is significantly lower in the TL spectrum than in the FL spectrum. This suggests that the Z-band emission is mostly contributed by PS I, and contribution by PS II is negligibly small. When thylakoids were fractionated into PS-I-enriched and PS

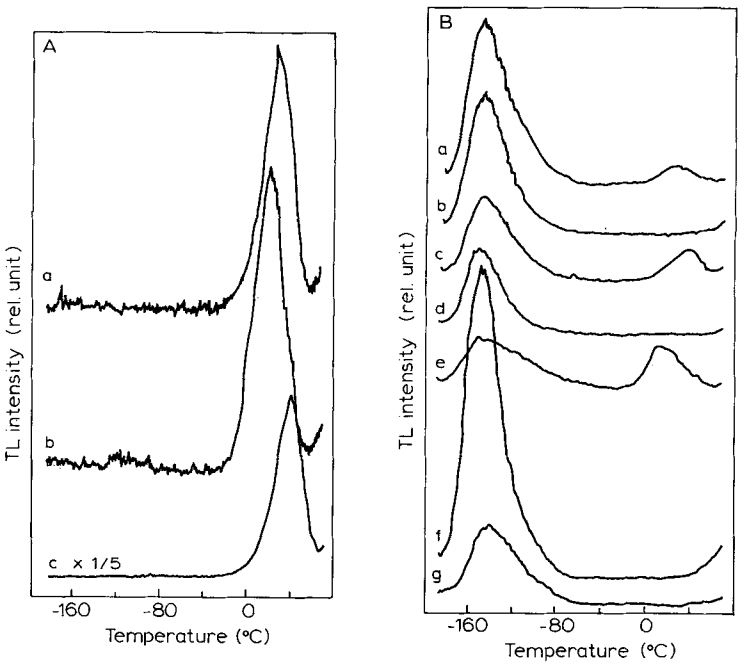


Fig. 2. Glow curves of thermoluminescence B-band (panel A) and Z-band (panel B). Panel A: spinach thylakoids, a; purified spinach PS II core complex, b; PS-II-enriched particles of *Synechococcus*, c. Panel B: spinach thylakoids, a; French-press-prepared spinach PS I, b; French-press-prepared spinach PS II, c; purified spinach PS I core complex, d; purified spinach PS II core complex, e; purified spinach LHC I, f; purified spinach LHC II, g. Chl concentrations were 0.3 mg/ml for all samples.

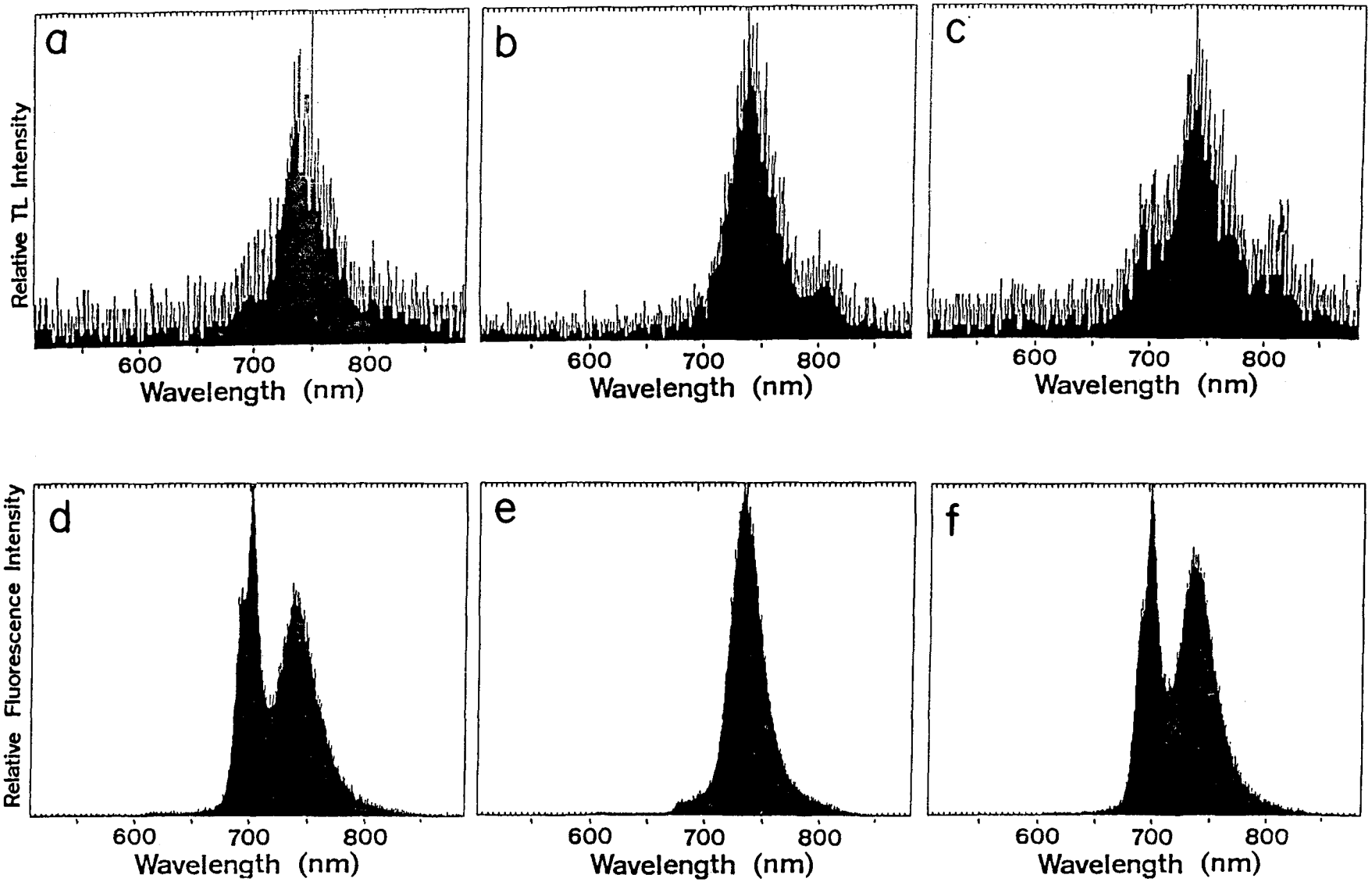


Fig. 3. Emission spectra of thermoluminescence Z-band (a, b, c) and low-temperature fluorescence (d, e, f) from spinach thylakoids (a, d), French-press-prepared PS I (b, e) and French-press-prepared PS II (c, f), respectively. Thermoluminescence photons were collected during heating from $-180\,^{\circ}C$ to $-120\,^{\circ}C$, and fluorescence was measured at 77 K. Chl concentrations were 0.3 mg/ml for all samples.

II-enriched membrane fragments, the PS I fraction showed a Z-band emission spectrum having a single peak at 740 nm with no shoulder at around 690 nm (b). The Z-band spectrum from the PS II fraction also showed a major peak at 740 nm, but the peak was accompanied by a small but significant shoulder at around 690 nm (c). Since mechanical disruption by a French pressure cell [13], rather than detergent solubilization, was employed in separating the two photosystems to avoid artifacts due to detergent-solubilized free chlorophyll, fractionation cannot be complete, and particularly, the PS II fraction contained significant amount of contaminating PS I, as evidenced by the FL spectrum (f). However, the appreciable enhancement of the 690 nm shoulder emission in the TL spectrum of the PS-II-enriched fraction strongly suggests that not only PS I but also PS II is capable of emitting the Z-band, even though its contribution is largely masked by the much stronger emission at 740 nm by the contaminating PS I.

The above view was further examined in experiments in which we split both PS I and PS II into respective reaction center core complexes and light-harvesting chlorophyll protein complexes. Spectra a and e in Fig. 4 show the Z-band emission spectra from the PS I core and the PS II core, respectively. The spectra indicate that the PS I core emits the Z-band exclusively at 740 nm, whereas the PS II core emits mostly at 690 nm but a broad tailing in the far-red region accompanies this. Since the two Z-band components were emitted from PS I core and PS II core complexes at two different respective wavelengths, one may consider that energy storage for Z-band involves photochemical reactions in respective reaction centers. However, this is not the case. The Z-band from PS I core complex was found to be insensitive to the presence of ferricyanide in the medium (data not shown). This precludes the involvement of the reaction center in charging the Z-band. This view was confirmed by the observation that isolated LHC I and LHC II, the light-harvesting chlorophyll protein complexes of PS I and PS II, respectively, are capable of emitting the Z-band. LHC I emits strongly at 740 nm with a very faint shoulder at 700 nm due to free chlorophyll solubilized by detergent (b), while LHC II emits at 685 nm with a broad tailing in the far-red region (f). Again, these TL emission spectra are in good agreement with their corresponding FL spectra at 77 K (d, h). It is thus concluded that: (i) The TL Z-band consists of two spectral components emitting at two different wavelengths, 740 nm and 690 nm. (ii) The former originates from PS I and the latter from PS II. (iii) In both PS I and PS II, the two components arise mostly from light-harvesting chlorophyll proteins and partly from reaction center chlorophyll proteins as well.

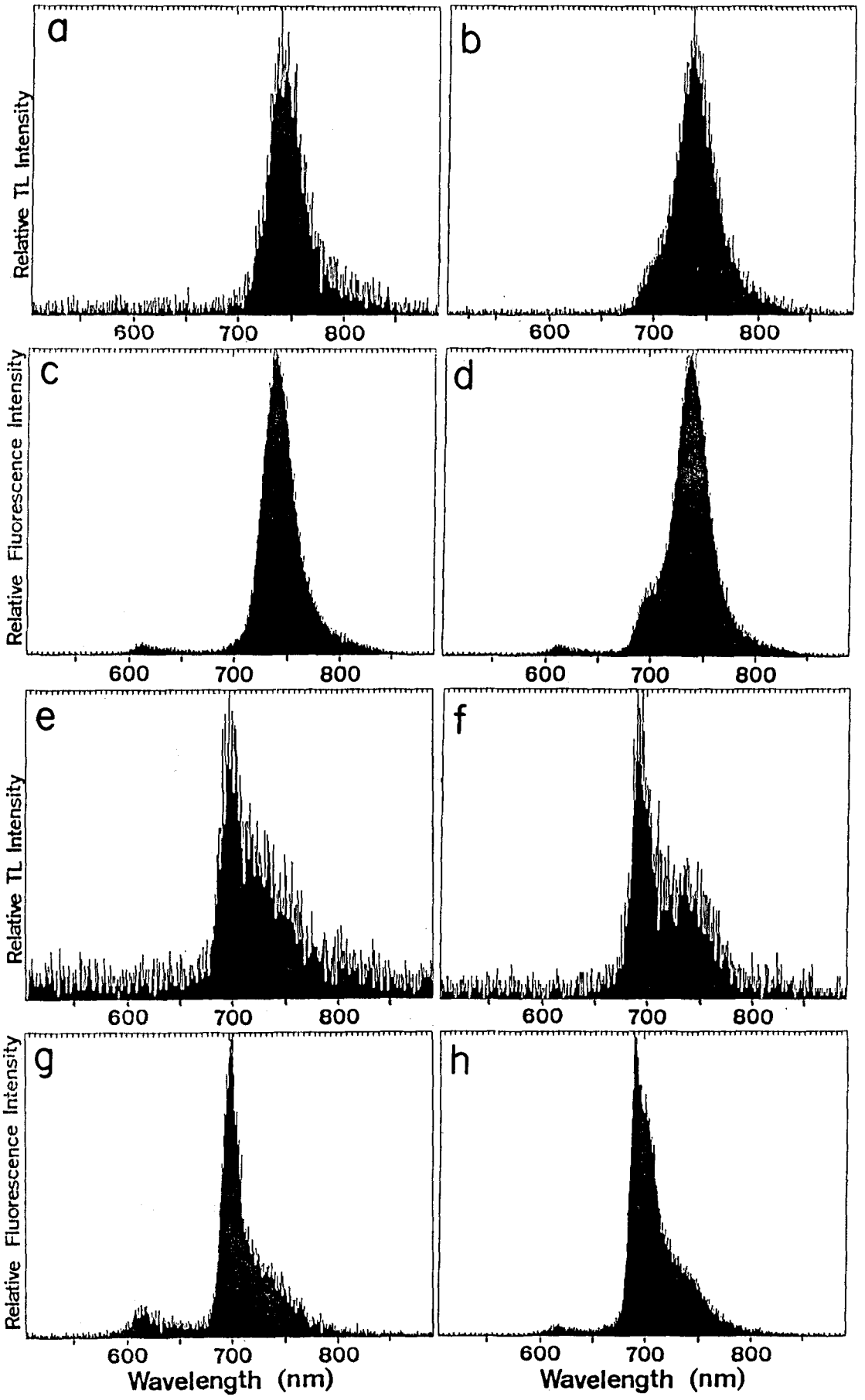
It is notable in this relation that the emission intensity of the 740 nm component from LHC I is higher than that of the 690 nm component from LHC II by a factor of 4 (see Fig. 2B). Similarly, the emission intensity from the PS I core complex is twice as high as that from the PS II core complex. These differences in Z-band-emitting capability are consistent with the very small contribution of the 690 nm component as compared with that of the 740 nm component in the emission spectrum of the TL Z-band from whole thylakoids, and seem to be related to the difference in local chlorophyll concentration between these preparations, as will be discussed later.

The above results, that the Z-band is emitted from LHC I and LHC II, the pigment proteins devoid of the reaction center, are consistent with the report [6] that boiled leaves emit the Z-band. These results suggest the possibility that the Z-band can also be emitted from purified chlorophyll. Fig. 5 shows the Z-band emission from a methanol extract of hexane-washed spinach thylakoids. The extract was capable of emitting the Z-band only very weakly, but when supplemented with water, the Z-band increased in strength, and the emission intensity markedly increased with increasing water concentration. Notably, considerable aggregation of pigments accompanies the increase in water concentration. The emission spectrum of the Z-band emitted from the extract containing no water showed a vague maximum at around 690 nm, whereas the band emitted from the extract containing 50% water showed a clearer maximum at 740 nm, as shown by Fig. 5b and c, respectively. The former peak position coincides with that of the Z-band from LHC II and the PS II core, and the latter with that of the Z-band from LHC I and the PS I core.

Discussion

In this study we determined the emission spectra of two TL components from the photosynthetic apparatus, the B-band and the Z-band, emitted at around $+30\,^{\circ}C$ and $-160\,^{\circ}C$, respectively. Of the two TL components, the B-band is known to originate from charge recombination between S_2/S_3 and Q_B^- [3,4]. Our results unambiguously showed that the emission spectrum of the B-band is identical with that of FL at room temperature. Since DL has been reported to show the same emission spectrum as FL [10], it is likely that the

Fig. 4. Emission spectra of thermoluminescence Z-band (a, b, e, f) and low temperature fluorescence (c, d, g, h) from purified PS I core complex (a, c), purified LHC I (b, d), purified PS II core complex (e, g) and purified LHC II (f, h), respectively. Other conditions are the same as in Fig. 3.


a
b
c
d
e
f
g
h
Relative TL Intensity
Relative Fluorescence Intensity
Relative TL Intensity
Relative Fluorescence Intensity
600
700
800
Wavelength (nm)

chlorophyll species responsible for the emission may be the same between DL and TL. This is consistent with the present view that both TL and DL arise from charge recombination in PS II reaction center as a reversal of photosynthetic charge separation [4,8]. Although we could not determine the emission spectra of all the TL components in this study, we may expect that those TL components originating from charge recombination in PS II (Q-band, A-band and possibly Z_v-band arising from $S_2Q_A^-$, $S_3Q_A^-$ and $Y_z^+Q_A^-$ charge pairs, respectively [2,5]) will probably show the same emission spectra as the B-band.

The emission spectrum of the TL B-band from thylakoids shows a sharp peak at 685–695 nm with an appreciable tailing toward far-red region (Fig. 1). This spectrum almost completely agrees with the DL emission spectrum spectrum of thylakoids recently reported by Hideg et al. [19]. As we can preclude the contribution by PS I based on the origin of this TL component, there remain two interpretations for the tailing in far-red region: (i) a second vibrational band of 690 nm emitting chlorophyll species; or (ii) an artifact due to self-absorption. Although self-absorption is minimized in all of our spectral measurements, it is technically difficult to certify its complete absence. However, we note in this context that almost the same emission spectrum having an appreciable shoulder could be observed for the purified PS II core complex (Fig. 1b). This implies that, in the measurement of thylakoid spectrum, self-absorption by large abundance of PS I chlorophyll was well mini-

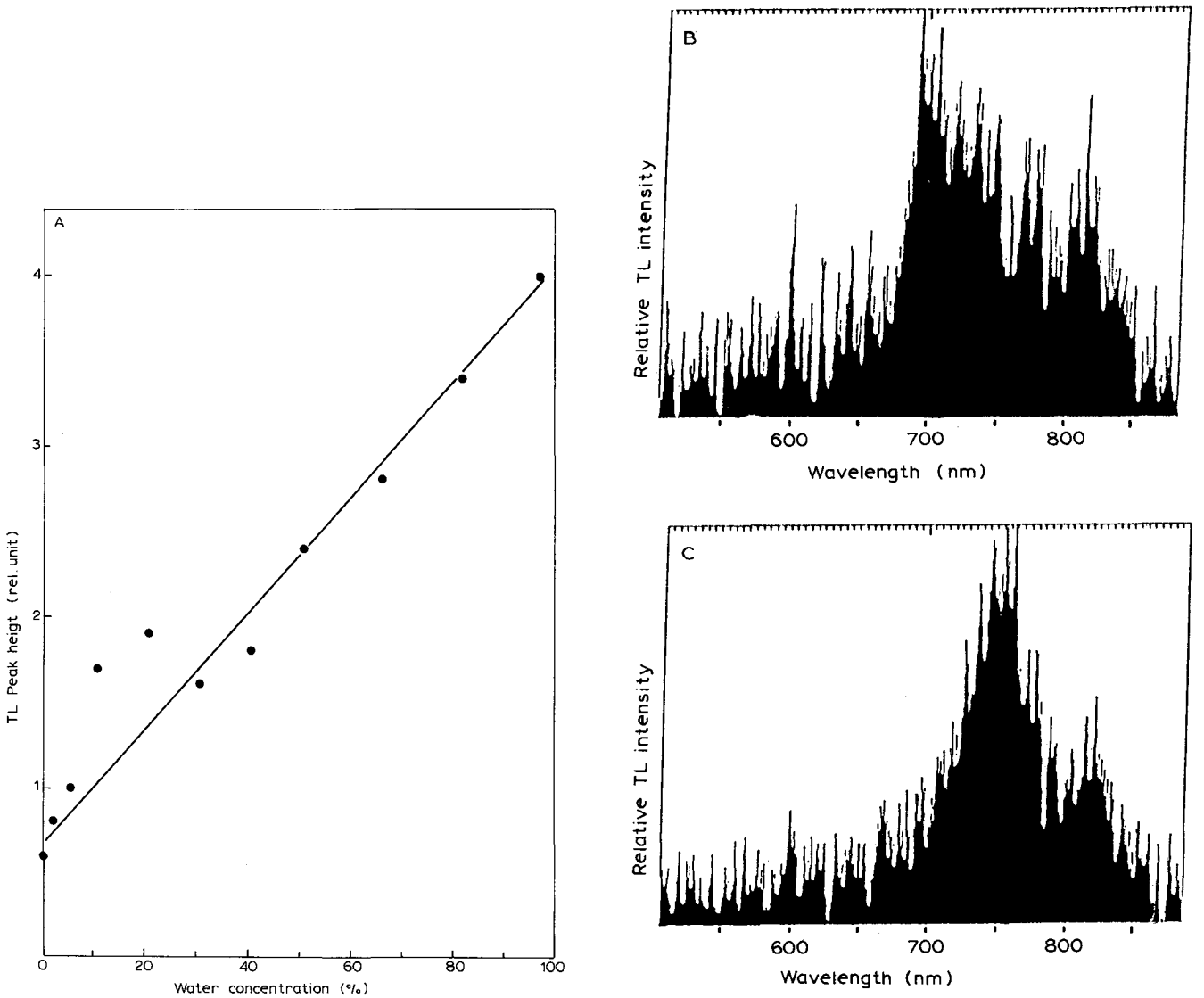


Fig. 5. (a) Enhancement of thermoluminescence Z-band intensity from free Chl by inclusion of water in methanol solution. A portion of concentrated chlorophyll dissolved in methanol was diluted with various methanol/water solutions to yield a constant Chl concentration (0.2 mg Chl/ml) in methanol/water with different water content, and relative Z-band intensities were determined from the glow curves as shown in Fig. 2. (b) and (c) are the emission spectra obtained with 0% and 50% water, respectively. Both spectra were processed by five-point smoothing.

mized under our experimental conditions. From these considerations, we tentatively conclude that the far-red tail is not an artifact due to self-absorption, but is an intrinsic characteristic, the second vibrational band of the chlorophyll species emitting the major band. However, a contribution due to self-absorption cannot be definitely excluded.

The Z-band emission spectrum was revealed to consists of two spectral components, the long-wavelength (740 nm) and the short-wavelength (690 nm) components. It was also clearly demonstrated that the former and latter components are emitted exclusively from PS I and PS II, respectively. Since the Z-band emission from both photosystems was completely insensitive to DCMU and ferricyanide (data not shown), and since purified LHC I and LHC II were found to be capable of emitting the Z-band at higher yields (Fig. 4b, f), we can exclude the possibility that the energy storage for this TL component involves primary charge separation in the reaction center and subsequent electron transfer. A possible mechanism for energy storage without involving reaction center photochemistry will be stabilization of the triplet state at low temperature. This idea, however, is unlikely, since the half decay time of the energy storage state for Z-band was as long as 30 min at 77 K (data not shown), which is markedly longer than the half-life of chlorophyll triplet state at 77 K (about 2 ms).

We cannot propose at present any plausible mechanism as to the energy storage for the Z-band, but we may point out that the local chlorophyll concentration as a factor might be related to the energy storage mechanism. In a PS I core complex, 110–120 chlorophyll molecules bind to a pair of 82–83 kDa polypeptides (*psa*A and *psa*B products) [20]. This can be expressed as 0.73 Chl/kDa protein. Similarly, the local chlorophyll concentrations in the PS II core complex, LHC I and LHC II, are estimated to be 0.13, 1.0 and 0.46 mol Chl/kDa protein, respectively, indicating that the Chl concentrations in the PS I core and LHC I are higher than in the PS II core and LHC II by factors of 5.6 and 2.1, respectively. When we compare the Z-band heights emitted from these preparations as shown in Fig. 2B, PS I core complex and LHC I emit 1.7- and 3.9-times more strongly than PS II core complex and LHC II, respectively, suggesting a tendency that a high Chl concentration gives rise to a high capability of Z-band emission. Although this correlation is not always valid between the PS I core complex and LHC II, we speculate that at high local concentrations, there will occur an interaction between two or more chlorophyll molecules, which makes these chlorophyll molecules capable of storing a part of absorbed light energy as a form of charge transfer complex.

The above speculation may be supported by the experiments of Fig. 5, which revealed that aggregate formation by the addition of water markedly enhanced the capability of Z-band emission by the chlorophyll dissolved in methanol. Interestingly, this enhancement in Z-band-emitting capability is accompanied by a shift in emission maximum. As indicated by the two spectra Fig. 5b and c, the low-intensity Z-band from free chlorophyll dissolved in methanol shows a maximum emission at 690 nm having broad tailing in the far-red region, whereas the high-intensity Z-band from aggregated chlorophyll in the methanol/water mixture (50%, v/v) shows a maximum at 740 nm: chlorophyll in pure methanol emits an LHC-II-type Z-band, whereas aggregated chlorophyll in methanol/water emits an LHC-I-type Z-band. It might be considered that this spectral shift is due to self-absorption, since the high local concentration of chlorophyll in the aggregate causes strong self-absorption and, as a result, the original peak at 690 nm diminishes with an apparent enhancement of the 740 nm band. According to this explanation, however, a stronger emission is expected for the 690 nm band from methanol-dissolved chlorophyll but not for the 740 nm band from aggregated chlorophyll. This is clearly not the case. Moreover, the same relationship should be expected between the two spectral components of Z-bands from PS I and PS II. This is also contradictory to our experimental evidence that PS I (LHC I) emits at 740 nm much more strongly than does PS II (LHC II) at 690 nm. These contradictions clearly deny the idea of self-absorption as the cause for the spectral shift accompanying the formation of chlorophyll aggregate, or for the difference in emission peak between PS I (LHC I) and PS II (LHC II).

In spite of the good correlation between local chlorophyll concentration and Z-band-emitting capability, the details of the energy-storage mechanism for the Z-band are not clear. However, the work of Yang and Brody [21] may be related to this problem: injection of illuminated carotenoid to aggregated chlorophyll results in DL emission. Their results suggest that carotenoids are responsible for energy storage and that aggregated chlorophyll plays a role in converting the stored energy into light. This idea may not be compatible with our above speculation that aggregated chlorophyll functions in energy storage for the Z-band. However, in our present experiments, the methanol solution of chlorophyll was prepared from the thylakoids briefly washed with hexane to remove quinones and some carotenoids, so that there remains a possibility that the extract contains some residual carotenoids, probably xanthophylls, which might have acted in energy storage. It has been well known since 1968 (see Arnold and Azzi [22]) that blue light is more effective in charging the Z-band. Our preliminary experiments in LHC I confirm this. The role of carotenoids or some quinones in energy storage for Z-band emission is the subject of further study.

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