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Evidence for long-range excitation energy migration in macroaggregates of the chlorophyll *a/b* light-harvesting antenna complexes

Virginijus Barzda [a,b], Győző Garab [a,*], Vidas Gulbinas [b], Leonas Valkunas [b]

[a] *Institute of Plant Biology, Biological Research Center, Hungarian Academy of Sciences, P.O. Box 521, H-6701 Szeged, Hungary*
[b] *Institute of Physics, Vilnius, Lithuania*

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Abstract

We investigated the picosecond transient absorbance kinetics under singlet-singlet annihilation conditions and the steady-state spectroscopic features, absorbance, circular dichroism and low-temperature fluorescence spectra, in large, three-dimensional, stacked lamellar aggregates of the purified light-harvesting chlorophyll *a/b* complexes (LHCII) and its form of small aggregates. Our data strongly suggest that the macroorganizational parameters significantly influence the spectroscopic properties and strongly affect the energy migration pathways in the aggregates. In small aggregates ($d \approx 100$ nm) of LHCII trimers the excitation energy migration could be characterized with a percolation type of excitation migration in a small cluster of chromophores. In contrast, in chirally organized macroaggregates ($d \approx 2$–4 μm), the annihilation kinetics were consistent with a model predicted for (infinitely) large three-dimensional aggregates, showing that LHCII macroaggregates can constitute a structural basis for long-range migration of the excitation energy.

Keywords: Circular dichroism; Energy migration; Light-harvesting chlorophyll *a/b* complex (LHCII); Psi-type aggregate; Singlet-singlet annihilation; Ultrafast absorbance transient

1. Introduction

The role of the light-harvesting antenna in photosynthesis is to absorb the solar energy and transfer the excitation energy to the reaction center where charge separation takes place. Excitation energy which does not reach the reaction center is emitted as fluorescence or dissipated to heat. High efficiency of photochemical utilization of light energy in vivo requires a fast rate of energy transfer. This can be satisfied with a high degree of molecular organization of the antenna [1]. The molecular organization of the antenna system may influence both the energy migration and the dissipative processes. In this respect, it is of special interest to characterize the macroorganization of the antenna system and to determine the effective distances of the excitation energy migration.

Thylakoid membranes contain particles of 100–180 Å in diameter, which probably include one reaction center complex which is surrounded by light harvesting antenna complexes [2]. Based mainly on circular dichroism (CD) spectroscopic investigations it has been shown that Photosystem II (PS II) particles readily assemble into chiral macrodomains which give rise to psi-type CD bands (psi, polymerization or salt induced) [3–5]. (Psi-type aggregates are three-dimensional macroaggregates containing high density of interacting chromophores and possessing sizes commensurate with the wavelength of the measuring light [6].) The formation of chiral macrodomains in thylakoid membranes is facilitated by LHCII [4], probably via electrostatic adhesion of the peripheral complexes [7]. Isolated, purified LHCII also easily forms macroaggregates and liquid crystalline sheets [8]. Stacked lamellar aggregates of LHCII have been shown to exhibit psi-type CD features due to long-range chiral order [9].

Large 'functional' domains containing at least several hundred Chl molecules have been detected by singlet-triplet annihilation measurements both in aggregated isolated LHCII and in chloroplast thylakoid membranes [10]. Time-resolved picosecond absorption measurements also showed that in LHCII macroaggregates fast excitation

Abbreviations: Chl, chlorophyll; CD, circular dichroism; LHCII, light-harvesting chlorophyll *a/b* complex; PS II, Photosystem II; psi, polymerization or salt induced.

* Corresponding author. Fax: +36 62 433434; e-mail: GARAB@ROSI.SZBK.U-SZEGED.HU.

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energy transfer occurs over a large number of Chl molecules [11]. In thylakoid membranes, picosecond spectroscopic measurements showed that the excitation energy is rapidly transferred from a large pool of Chl molecules (probably LHCII) to smaller units [12]. Analysis of fluorescence induction kinetics revealed connectivity of PS II reaction centers [13,14]. Macroaggregated LHCII in thylakoid membranes has been hypothesized to be involved in the regulation of the dissipation of excess excitation energy [15]. However, our understanding of the significance of the macroorganization of LHCII in vivo and our knowledge concerning the molecular mechanism of the regulated dissipation of the excess excitation energy in the membranes are far from being complete.

In this work we performed systematic comparative measurements on aggregates of LHCII of stacked lamellar structure and their smaller aggregates. Energy migration pathways were characterized by ultrafast absorbance transients due to singlet-singlet annihilation processes. Spectroscopic properties, absorbance, CD and low-temperature fluorescence emission were also compared in the two types of LHCII aggregate. We show that in LHCII the spectroscopic features and the energy migration pathways depend significantly on the size of aggregates.

2. Materials and methods

LHCII was prepared from pea (*Pisum sativum*) leaves according to [16]. Essentially identical results were obtained with a preparation technique described in [17]. These techniques yield three-dimensional aggregates with quasi lamellar structure [18]. Samples were suspended in buffer containing 10 mM Tricine/KOH (pH = 7.5). Aggregation and disaggregation of LHCII was reversibly induced by the addition of $MgCl_2$ (0–5 mM) and Triton-X 100 (0–0.02%, v/v), respectively. The size of aggregates was determined by negative-staining electron microscopy in a Zeiss 902 electron microscope and by fluorescence microscopy in a Leitz Laborlux S microscope.

Absorption spectra were measured in a Shimadzu UV-3000 spectrophotometer. CD spectra were recorded in a Jobin-Yvon CD6 dichrograph, and represented in units of absorbance. Fluorescence measurements were performed in a setup described earlier [19].

Determination of exciton-exciton annihilation dynamics was carried out by means of pump-probe absorption spectroscopy. The picosecond spectrometer was based on a passively mode locked $KGd(WO_4)_2$ laser. The samples were excited at 590 nm with pulses of the second harmonics of the Raman scattered light from the main radiation of the laser. Raman scattering took place in the active $KGd(WO_4)_2$ rods of oscillator and amplifiers. Radiation of the optical parametric generator tunable between 400 nm and 1700 nm, or its second harmonics was used as a probe light. The response time of the instrument was 7 ps. The exciting pulse was focused to a 1 mm diameter spot. The diameter of the probe pulse was 0.3 mm and its energy density was by more than two orders of magnitude lower than that of the exciting pulse. The pulse repetition rate was 2 Hz, and thus accumulation of triplets was avoided.

3. Theory

Comparative analysis of the kinetics of singlet-singlet annihilation in macroaggregates and in small aggregates of LHCII was performed by the following kinetic equation:

$$\frac{dn}{dt} = -kn - \gamma(t)n^2, \tag{1}$$

where n is the number of excited Chl a molecules in the domain, k is the linear decay rate of the excitation due to the natural relaxation and trapping by various quenching centers, $\gamma(t)$ is the rate of the non-linear relaxation due to singlet-singlet annihilation.

The solution of Eq. 1 for the general case can be written as follows:

$$n(t) = \frac{n_0 \exp(-kt)}{1 + n_0 \int_0^t \gamma(t') \exp(-kt') dt'}, \tag{2}$$

where n_0 is the initial value of excitations at $t = 0$.

The general analysis shows that time-dependence of the nonlinear annihilation rate $\gamma(t) = \gamma_0/t^\alpha$ is due to the correlative effect of excitations [20] and/or to the dimensionality of the structure under consideration [21,22]. Also, by assuming the linear decay time, k^{-1}, to be slow (of the order of ns) compared to the time interval investigated, Eq. 2 can be written in the following analytical form:

$$n(t) = \frac{n_0}{1 + n_0 \gamma_0 \frac{1}{1-\alpha} t^{1-\alpha}}. \tag{3}$$

For three-dimensional, large (strictly speaking, of infinite size) systems, γ becomes time-independent (i.e., $\alpha \rightarrow 0$). In such a case, the kinetics determined by Eq. 2 can be written as:

$$n(t) = \frac{n_0 \exp(-kt)}{1 + n_0 \gamma k^{-1} [1 - \exp(-kt)]}. \tag{4}$$

The correlation between $n(t)$ and the experimentally determined quantity, the absorption changes, $\Delta A(t)$, is found as follows:

$$\Delta A(t) \sim (\sigma_1 - \sigma) \cdot n(t), \tag{5}$$

$$A \sim \sigma \cdot C.$$

where σ and σ_1 are molecular cross-sections of the ground state and the excited state absorptions, respectively, C is the concentration of Chl a molecules in the domain. The coefficient of the proportionality contains the size and

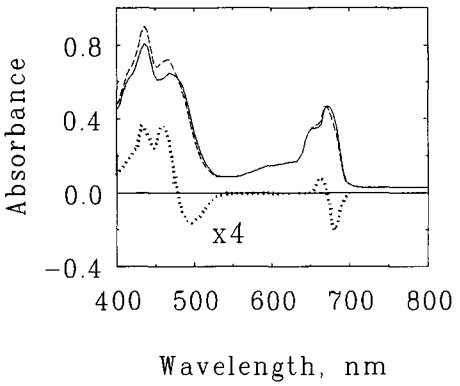

Fig. 1. Absorption spectra of macroaggregates (————) and small aggregates (– – –) of LHCII, and the difference spectrum (- - -). LHCII macroaggregates were suspended in 10 mM Tricine, disaggregation was achieved by addition of Triton X-100 (0.02% (v/v)); optical pathway, 1 cm; Chl($a+b$) content, 20 μg/ml.

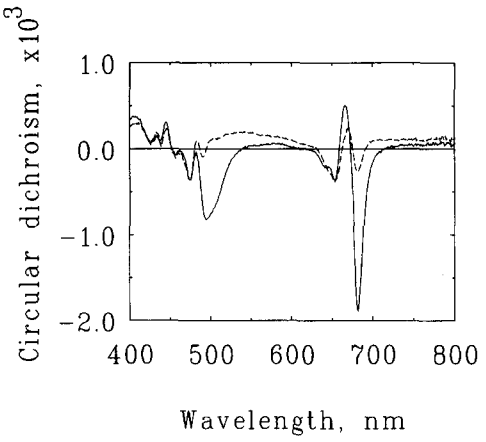

Fig. 2. CD spectra of macroaggregates (————) and small aggregates (– – –) of LHCII. (The same samples were used as in Fig. 1.)

the number of the domain in the path of the light beam. The excitation concentration kinetics, $n(t)$, can be determined by the temporal evolution of the optical absorption via the following relation:

$$n(t) = C\frac{\sigma}{(\sigma_1 - \sigma)}\frac{\Delta A}{A}. \quad (7)$$

4. Experimental results

Absorption spectra of LHCII upon the aggregation exhibit minor red shifts at 680 nm and 490 nm, and some decrease of absorbance in the Soret region could also be detected (Fig. 1). These changes can be explained by an increase in light scattering and by flattening effect [23] due to the presence of large aggregates. The size of the macroaggregates typically ranged between 2 and 4 μm, whereas the small aggregates of trimers were about 100 nm in diameter (data not shown). Under these conditions macroaggregation of LHCII does not lead to any major change in the absorption which would be expected if macroaggregation would bring about intense excitonic interactions between pigment molecules. Most of these interactions appear to be contained in trimers of LHCII [24] or in smaller aggregates of trimers [25] and macroaggregation of small aggregates of trimers does not induce major changes in the absorbance.

In contrast to the absorbance, formation of macroaggregates leads to the appearance of anomalous CD bands with extreme large amplitudes (Fig. 2). We have shown earlier that the increase of these CD bands at 493 nm and 682 nm is monotonous with the size of aggregates, if the average size ranges between about 100 nm and 4 μm [26]. As can be seen in Fig. 2, CD between 400 and 460 nm and 600 and 660 nm remains essentially unchanged, showing that the bands originating in short-range interactions are invariant on the size of the aggregate, whereas size-dependency is strong in psi-type bands associated with long-range chiral order in the macroaggregates [4,26].

Fluorescence quantum yield at 77 K in macroaggregates was typically lower by about 30% than in the smaller aggregates (Fig. 3) which exhibit no psi-type CD bands. Such changes in fluorescence yield compared with literature data [15,27], in which fluorescence quantum yield increased by more than 5–10-times due to disaggregation, is attributed to the fact that under our experimental conditions disintegration to trimers or monomers was not reached. Upon increasing the concentration of detergent a substantial increase in the fluorescence yield was observed, which, however, was accompanied with changes in the excitonic CD bands (data not shown cf. [28]).

Measurement of the singlet-singlet annihilation is a convenient tool to investigate the excitation energy migration pathways in the domain. The migration distance is determined either by the real size of the molecular array or by the excitation migration radius (if the size of the macromolecular structure is larger than that parameter).

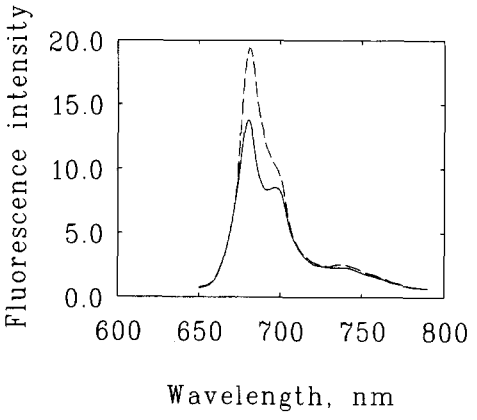

Fig. 3. 77 K fluorescence emission spectra of macroaggregates (————) and small aggregates (– – –) of LHCII. (The aliquots were taken from the samples used in absorbance and CD measurements.)

The bleaching kinetics of Chl *a* upon the excitation with the laser pulse can be monitored at 680 nm. (Similar kinetics were obtained at other wavelengths between 670–690 nm, whereas around 650 nm we found that the initially excited Chl *b* molecules transferred the excitation energy to Chl *a* within the limits of the time-resolution of our measurement, i.e., within less than ten ps (data not shown cf. [29]).)

As shown in Fig. 4A, with the pulse intensities used in our experiments, the small aggregates of LHCII trimers display nonlinear kinetics, i.e., the decay time depends on the intensity of the exciting pulse.

The kinetic traces obtained for small aggregates were fitted with Eq. 3, by varying the parameter α. The same α value was used for the traces obtained with different excitation intensities. This kinetic equation is valid for the case with slow linear decay time. (Since the linear relaxation time in small aggregates is in the order of ns, this condition is fulfilled.) The best fit of experimental data was obtained with $\alpha = 0.25 \pm 0.03$. This can be seen more clearly in Fig. 4B, in the linearized plot.

Qualitatively, this result means that the small aggregates are systems where the excitation correlative effects are essential, or they create fractal structures whose spectral dimension, d_s, is $d_s = 2(1 - \alpha) = 1.5$ [30]. This indicates that small aggregates of LHCII behave like energetically disordered structures where the excitations migrate through percolation type cluster [21] of the pigment molecules creating the local minima for excitations.

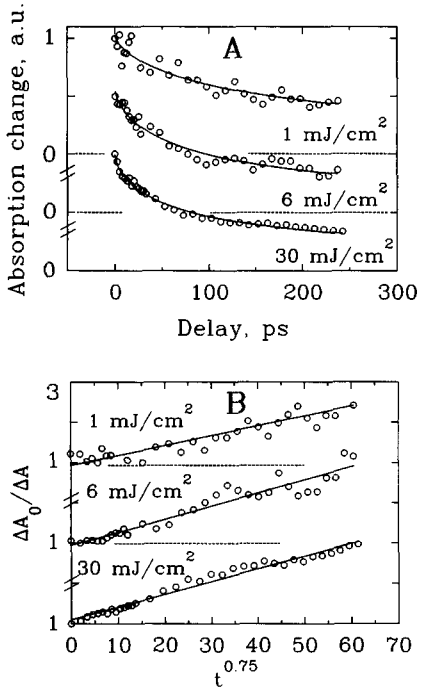

Fig. 4. Absorbance transients at 680 nm under singlet-singlet annihilation conditions in small aggregates of LHCII upon excitation with different intensities of 590 nm laser pulses (A). Data analyses are also presented in linearized plot (B). Data were fitted according to Eq. 3. Experimental data and fitted curves are represented by open circles and continuous lines, respectively. In graph (A), the initial amplitudes were normalized to unity. The kinetic traces were shifted as indicated in the figures with the baselines. (The initial amplitudes were 0.2, 0.1, and 0.02 with 30, 6 and 1 mJ/cm^2 excitations, respectively.) The optical pathway was 0.1 cm; Chl($a + b$) content, 300 μg/ml; $A_{680} = 0.8$.

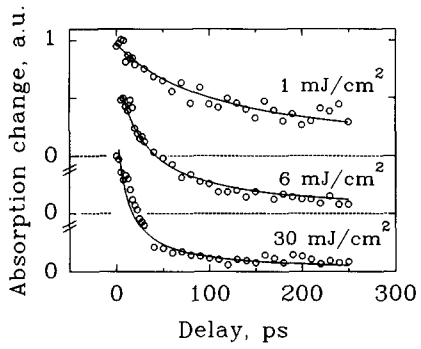

Fig. 5. Absorbance transients at 680 nm under singlet-singlet annihilation conditions in macroaggregates of LHCII upon excitation with different intensities of 590 nm laser pulses. Data were fitted according to Eq. 4. Experimental data and fitted curves are represented by open circles and continuous lines, respectively. The initial amplitudes were normalized to unity. The kinetic traces were shifted as indicated in the figures with the baselines. (The initial amplitudes were 0.2, 0.1, and 0.02 with 30, 6 and 1 mJ/cm^2 excitations, respectively.) The optical pathway was 0.1 cm; Chl($a + b$) content, 300 μg/ml; $A_{680} = 0.8$.

In macroaggregates the kinetics of absorbance transients were much faster than in the small aggregates, whereas the initial amplitudes were the same. The fact that with high intensity exciting pulse the initial amplitude became strongly non-linear is likely to be due to an essentially full bleaching of the Chl band(s) at 590 nm, which results in a bleaching 25% of Chl *a* absorbing at 680 nm. Annihilation processes in the pulse cannot be excluded. However, this seems unlikely, because the initial amplitudes were the same for small and large aggregates, whereas the decay kinetics were significantly different in the two samples. Further, the kinetics could be fitted with the same fitting parameters for 1, 6 and 30 mJ/cm^2 exciting pulses.

In the macroaggregates, the absorption transients at various excitation intensities (Fig. 5) followed a hyperbolic decay, which is typical for non-linear relaxation processes in large three-dimensional structures where the non-linear rate parameter γ becomes time-independent (see Theory).

For large aggregates, the data were fitted with the time-independent γ (Eq. 4). The best fits were obtained by assuming $C\gamma\sigma/(\sigma - \sigma_1) = 0.5$ ps^{-1} and linear relaxation lifetimes, k^{-1}, between 2 and 4 ns (Fig. 5).

Since the linear relaxation time k^{-1} was found to be slow compared to the time interval investigated, the kinetics could also be fitted with Eq. 3. With this fit we obtained $\alpha \approx 0$ (< 0.05), which means that in the macroaggregates γ becomes essentially time-independent; hence, the use of Eq. 4 is justified. Thus, it can be concluded that the annihilation in macroaggregates occurs

with kinetics that are fully consistent with the model for large three-dimensional structures.

Based on these results we analyzed further the data in large aggregates. Assuming hopping mechanism of energy migration, we estimated the diffusion radius of the excitation migration. The mere purpose of this analysis was to provide a rough estimate on the size of the domain in which the excitation can migrate.

The exact value of σ_1 which includes the absorption into higher excited states as well as the stimulated emission on the probing wavelength, is not known. However, as is evident from the absorption (Fig. 1) and fluorescence (Fig. 3) spectra, for estimations we can use $\sigma/(\sigma - \sigma_1)$ being of the order of 0.5 at 680 nm. By calculating with $a = 10$ Å (a is the interpigment distance) [31] and with $4/3\pi a^3 C = 1$ we obtain $\gamma = 4 \cdot 10^{-9}$ cm^3 s^{-1}. This value of γ can be used to estimate the excitation migration parameters for diffusion limited singlet-singlet annihilation process cf. [20]:

$$\gamma = 8\pi DR \tag{8}$$

and

$$D = \frac{a^2}{\tau_{hop}}, \tag{9}$$

where D is the excitation diffusion constant and R is the annihilation 'reaction' radius. Thus, the hopping time, τ_{hop}, can be estimated to be about 6 ps with $R = a = 10$ Å. This rough estimation, which gives an upper limit for τ_{hop}, is in line with the conclusions of [11,29,32] which indicated subpicosecond and picosecond kinetics to be present.

Based on the above values, we can estimate the excitation migration radius R_{dif} which is defined as

$$R_{dif} = \sqrt{\frac{da^2}{k\tau_{hop}}}, \tag{10}$$

where the coordination number $d = 6$ for three-dimensional structures, assuming cubic arrangement. The migration radius can be estimated to be at least 640 Å. This high value indicates that the excitation migrates over a large domain containing several hundred thousand Chl molecules.

5. Discussion

In order to attain a better understanding of the significance of macroorganization of LHCII in thylakoid membranes, in this work we performed a systematic comparison between small and large aggregates of purified LHCII.

Earlier singlet-triplet annihilation processes in LHCII aggregates were followed with the aid of analysis of fluorescence yield as a function of the intensity of the actinic flash [10]. Significant differences were observed in the functional domain sizes between LHCII complexes and their aggregates. However, theoretical analysis showed that the resolution of the measurements is very poor if the domains contain more than few hundred pigment molecules.

Absorbance transients under singlet-singlet annihilation conditions were also recorded in LHCII macroaggregates but were not compared with small aggregates [11]. The data were analyzed with the assumption that the recovery follows a three-exponential kinetic pattern, and it was concluded that energy transfer occurs over a large number ($N \geq 1000$) of Chl molecules.

By using the same technique, we compared the kinetics of the large and the small aggregates of LHCII and found that the annihilation processes were very sensitive to the aggregation state of LHCII. Analysis of the kinetics traces was based on the same kinetic equation (Eq. 1) both for the macroaggregates and the smaller aggregates. This equation was earlier introduced by Gillbro et al. [11] but has not been solved in analytical form. We solved this equation for the case when the linear decay time is slow, which is readily justifiable for small aggregates, and for large three-dimensional aggregates in which the rate of non-linear relaxation becomes time-independent.

Eq. 1 is not applicable to very small aggregates, but could be applied for our case, as our small aggregate particles ($d \approx 100$ nm) contain several dozens of trimers. (The high aggregation state of our small aggregates compared to trimers is also reflected by the fluorescence data; cf. comment on the fluorescence yield in Experimental results.) Thus, our small, mainly two-dimensional aggregates still contain a very large number of Chl molecules. Despite this fact, the excitation energy migration appears to percolate in a small cluster of molecules. This may be due to a disorder of the trimers or their loose packing, which may render the probability of inter-trimer energy transfer low. Alternatively, correlative effects of the excitation may be present, the simplest version of which can be taken into account by assuming Poisson distribution for the initial conditions.

In large aggregates, the annihilation appears to occur with a strikingly different mechanism. Our data could be fitted with curves characteristic of large three-dimensional aggregates. Thus, it can be concluded that these aggregates serve a structural basis for energy migration for long distances. This qualitative conclusion is further supported by a simple model calculation which showed that in LHCII macroaggregates the diffusion radius of the excitation, assuming hopping mechanism for the excitation energy migration, is about 640 Å.

A comparison of absorbance and CD of LHCII aggregates showed that excitonic interactions were not induced upon macroaggregation, and thus changes in short-range coupling of chromophores are highly unlikely to be responsible for the large differences between the macroaggregates and the small aggregates concerning the energy migration patterns.

On the other hand, upon the formation of macroaggregates large asymmetric CD bands emerged at 682 and 493 nm. These bands in LHCII, and similar bands in thylakoid membranes, have earlier been assigned to psi-type aggregates [4,5,26]. Psi-type aggregates have been characterized to possess large density of interacting dipoles, a size exceeding $\lambda/4$ (λ, wavelength of the light interacting with the aggregate) and long-range chirality in their molecular organization [6]. Hence, these data strongly suggest that long-range order of chromophores is essential for the long-distance migration of the excitation energy in large aggregates.

In psi-type aggregates, in addition to dipole-dipole coupling of chromophores intermediate and radiation coupling mechanisms must also be taken into account. Theory predicts that due to this type of coupling, the excitation can delocalize over regions commensurate with the wavelength of the visible light [6]. With the present investigations we cannot answer whether or not the long-range coupling of molecules in psi-type aggregates plays a role in the delocalization of the excitation energy. (For simplicity, in the calculation of the diffusion radius in the macroaggregates we used the hopping mechanism for the excitation energy transfer.) Nonetheless, our data clearly show that the excitation energy migration pathways are strikingly different in small and large aggregates of LHCII (cf. Figs. 4 and 5). The fact that the annihilations in psi-type aggregates of LHCII occur with kinetics expected for infinitely large three-dimensional domains may indicate the importance of long-range coupling of chromophores. It must be noted, however, that long range energy migration may also occur in two-dimensional aggregates. Thus, it cannot be excluded that structures which do not give rise to psi-type CD also constitute a structural basis for long range migration of the excitation.

We would also like to point out that both CD and non-linear absorbance kinetics are sensitive indicators of the parameters related to the macroorganization of chromophores. Thus, LHCII aggregates exhibit not only the characteristic psi-type spectroscopic features [26], but also appear suitable system for studies concerning excitation migration mechanisms in psi-type aggregates. As psi-type aggregates have been detected in granal thylakoids [5,26], these data may be useful in the elucidation of the role of macroorganization of PS II particles in the thylakoids.

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References

[1] Van Grondelle, R., Dekker, J.P., Gillbro, T. and Sundström, V. (1994) Biochim. Biophys. Acta 1187, 1–65.
[2] Armond, P.A., Staehelin, L.A. and Arntzen, C.J. (1977) J. Cell. Biol. 73, 400–418.
[3] Garab, G., Wells, K.S., Finzi, L. and Bustamante, C. (1988) Biochemistry 27, 5839–5843.
[4] Garab, G., Kieleczawa, J., Sutherland, J.C., Bustamante, C. and Hind, G. (1991) Photochem. Photobiol. 54, 273–281.
[5] Finzi, L., Bustamante, C., Garab, G. and Juang, C-B. (1989) Proc. Natl. Acad. Sci. USA 86, 8748–8752.
[6] Keller, D. and Bustamante, C. (1986) J. Chem. Phys. 84, 2972–2979.
[7] Liker, E. and Garab, G. (1995) Physiol. Plant. 93, 187–190.
[8] Kühlbrandt, W. and Downing, K.H. (1989) J. Mol. Biol. 207, 823–828.
[9] Garab, G., Faludi-Dániel, A., Sutherland, J.C. and Hind, G. (1988) Biochemistry 27, 2425–2430.
[10] Kolubayev, T., Geacintov, N.E., Paillotin, G. and Breton, J. (1985) Biochim. Biophys. Acta 808, 66–76.
[11] Gillbro, T., Sandström, A., Spangfort, M., Sundström, V. and Van Grondelle, R. (1988) Biochim. Biophys. Acta 934, 369–374.
[12] Wittmershaus, B., Nordlund, T.M., Knox, W., Knox, R.S., Geacintov, N.E. and Breton, J. (1985) Biochim. Biophys. Acta 806, 93–106.
[13] Joliot, P., Bennoun, P. and Joliot, A. (1973) Biochim. Biophys. Acta 305, 317–328.
[14] Lavergne, J. and Trissl, H.W. (1995) Biophys. J. 68, 2474–2492.
[15] Horton, P., Ruban, A.V., Rees, D., Pascal, A.A., Noctor, G. and Young, A.J. (1991) FEBS Lett. 292 (1,2), 1–4.
[16] Krupa, Z., Huner, N.P.A., Williams, J.P., Maissan, E. and James, D.R. (1987) Plant Physiol. 84, 19–24.
[17] Burke, J.J., Ditto, C.L. and Arntzen, C.J. (1978) Arch. Biochem. Biophys. 187, 252–263.
[18] McDonnel, A. and Staehelin, L.A. (1980) J. Cell Biol. 84, 40–56.
[19] Garab, G., Sundqvist, C., Mustardy, L. and Faludi-Dániel, A. (1980) Photochem. Photobiol. 31, 491–494.
[20] Suna, A. (1970) Phys. Rev. BV.1, 1716–1739.
[21] Klymko, P.W. and Kopelman, R. (1983) J. Chem. Phys. 87, 4565–4567.
[22] Bunde, A. and Havlin, S. (1991) Fractals and disordered systems, Springer, Berlin.
[23] Bustamante, C. and Maestre, M.F. (1988) Proc. Natl. Acad. Sci. USA 85, 8482–8486.
[24] Hemelrijk, P.W., Kwa, S.L.S., Van Grondelle, R. and Dekker, J.P. (1992) Biochim. Biophys. Acta 1098, 159–166.
[25] Jennings, R.C., Zucchelli, G., Bassi, R., Vianelli, P. and Garlaschi, F.M. (1994) Biochim. Biophys. Acta 1184, 279–283.
[26] Barzda, V., Mustardy, L. and Garab, G. (1994) Biochemistry 33, 10837–10841.
[27] Ruban, A.V. and Horton, P. (1992) Biochim. Biophys. Acta 1102, 30–38.
[28] Gülen, D., Knox, R.S. and Breton, J. (1986) Photosynthesis Res. 9, 13–20.
[29] Palsson, L.O., Spangfort, M.D., Gulbinas, V. and Gillbro, T. (1994) FEBS Lett. 339, 134–138.
[30] Kudzmauskas, S., Liuolia, V., Trinkunas, G. and Valkunas, L. (1988) in Proc. V. UPS Topical Meeting (Rudzikas, Z., Piskarskas, A. and Baltramiejunas, R., eds.), pp. 248–256, World Scientific Publishing, Singapore.
[31] Kühlbrandt, W. and Wang, D.N. (1991) Nature 350, 130–134.
[32] Kwa, S.L.S., Van Amerongen, H., Lin, S., Dekker, J.P., Van Grondelle, R. and Struve, W.S. (1992) Biochim. Biophys. Acta 1102, 202–212.