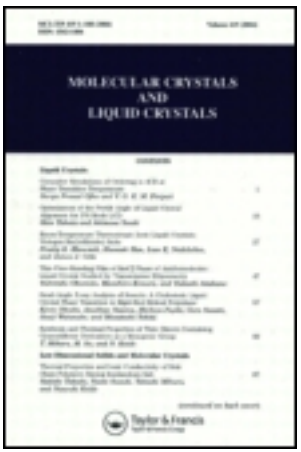

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Liquid Crystal-Like Domains of the Light Harvesting Chlorophyll a/b Protein Complex in Chloroplast Thylakoid Membranes

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LIQUID CRYSTAL-LIKE DOMAINS OF THE LIGHT HARVESTING CHLOROPHYLL a/b PROTEIN COMPLEX IN CHLOROPLAST THYLAKOID MEMBRANES

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Abstract Recent experimental results, reviewed in this paper, suggest that the light harvesting pigment protein complex in granal thylakoid membranes exists in liquid crystal like domains. These macroarrays can play an important role in the efficient photosynthetic utilization of light energy. The experimentally observed light induced reversible structural rearrangements, on the other hand, may be required for the regulatory redistribution of the excitation energy between photosystems 1 and 2.

INTRODUCTION

The light harvesting chlorophyll a/b pigment protein complex (LHC) is one of the most abundant protein complexes on Earth. It makes up about 50% of the total protein content of chloroplasts of green algae and higher plants. In addition to its basic function as a light harvesting molecular antenna, LHC regulates the distribution of excitation energy between the two types of photochemical reaction centers and mediates the stacking of granal thylakoids.

Dedicated to the memory of Prof. Á. Faludi-Dániel (1928-1986)

It has been suggested[1] that LHC in its native thylakoid membrane exists in liquid crystal-like domains. Indeed, the most recent investigations of chiroptical, magnetic and electric properties of various thylakoid membranes of different LHC contents support this hypothesis. The presently available data suggest that LHC in vivo occurs in helically organized "elastic" macroarrays which are capable of undergoing reversible structural rearrangements regulated by the photosynthetic electron transport.

CIRCULAR DICHROISM

Mature granal chloroplasts exhibit an alomalously big CD signal, the amplitude of which is about one order of magnitude higher than the CD signal of the constituent pigment protein complexes[2-4]. In addition, chloroplasts also show the long tail anomaly, i.e. an intense CD signal outside the principal absorbance bands.

Systematic comparative studies of the CD characteristics of the isolated purified liquid crystalline LHC[5,6] and chloroplasts revealed many similarities. Both mature thylakoids and the isolated liquid crystalline LHC exhibit an anomalously big ("giant") CD signal between 670 and 690 nm. The giant signal is absent in the monomeric purified complex[3]. It is also absent in immature thylakoids which are deficient in LHC[7]. It has been shown that LHC and the giant CD signal of thylakoids are synthesized together during the chloroplast development[1].

In a large three dimensional macroaggregate, the size of which is at least one quarter of the wavelength of the absorption maximum, an intense "psi type CD" can be given

rise inside the absorption band due to long range interactions between the chromophores[8]. In a chirally organized macroaggregate, which has dimensions commensurate with the wavelength, another anomalous CD signal can appear. The CD in such complexes does not originate solely from circular differential absorbance but also reflects the ability of large chiral particles to preferentially scatter right or left circularly polarized light. This circular differential scattering (CDS) signal conveys useful information regarding the macroorganization of the aggregate[9]. CDS can be recognized by the presence of an intense long tail outside the absorbance band but there may also be intense CDS inside the absorbance band. CDS displays alternating positive and negative lobes of the CD signal with respect to the direction of the measuring beam or with respect to the orientation of the scattering particle.

In a recent study[10] we investigated the possible contribution of CDS to the anomalous CD signal of chloroplasts and the purified liquid crystalline LHC. First, our attention was focussed on the long tails observed between 700 and 800 nm, and between 520 and 600 nm, where the CD signal can originate only from CDS. It was found that magnetic alignment of the chloroplasts causes the CDS signal to change sign, in agreement with the expectations. Sign-inversion of the CDS signal upon reorientation of the scattering helix has been predicted theoretically[11]. Our studies were extended to spectral regions within absorbance bands, to ascertain the extent to which the previously characterized orientation dependence of CDS was manifest. A very strong interference from CDS could be detected around the main CD bands at 690 and 510

nm. Similar results were obtained with the purified LHC aggregate.

The liquid crystalline structure of the isolated purified LHC aggregate can define a helical macroarray of the chromophores. Non-random orientation of the transition dipoles[12] may also be important in this respect. CDS properties of chloroplasts are indicative of the presence of large helically organized domains of the LHC in granal membranes.

MAGNETIC AND ELECTRIC PROPERTIES

Diamagnetic susceptibility of anisotropic particles can be calculated from their orientability in an external magnetic field[13]. (E.g. linear dichroism of absorbance or fluorescence emission can be measured as a function of field strength.) It has been shown that the aggregated purified LHC exhibits a very high diamagnetic susceptibility, relative to the monomeric complex[14]. A similarly high susceptibility was obtained with mature granal chloroplasts. However, in other type of chloroplasts which are deficient in LHC, the diamagnetic susceptibility was more than an order of magnitude lower than in the granal chloroplasts. These data show that the high diamagnetic susceptibility in chloroplasts is a collective property of the LHC macroarray. This result corroborates the conclusion that LHC in mature thylakoid membranes exists in liquid crystal-like domains.

Chloroplasts can be oriented in an external electric field[15]. The orientability of the aggregated purified LHC (J.G. Kiss, Á. Faludi-Dániel and Gy.I. Garab, unpublished) is very similar to that of chloroplasts. Both chloroplasts

and LHC aggregates can be aligned in a weak, 20-40 V/cm, external field, whereas monomeric LHC or particles deficient in LHC can be aligned only in much stronger electric fields. These results imply that the dipole moment of mature thylakoids is determined by the presence of LHC domains in the membranes. The high dipole moment of LHC, by allowing the LHC domain to respond to electric and ion gradients generated by the photosynthetic electron transport, may have a physiological importance (see below).

LIGHT INDUCED REVERSIBLE REARRANGEMENTS OF THE LHC-DOMAINS

Following the basic observations by Gregory[16] and Faludi-Dániel[17], light induced reverersible CD changes have recently been investigated[18]. It has been pointed out that, in contrast to the earlier interpretations, these slow (30-90 sec) reversible changes cannot be accounted for by changes in the short range interactions between chlorophyll molecules but can be identified as CDS changes. The same kinetics was observed in all spectral regions dominated by CDS, both in randomly oriented and magnetically aligned suspensions of chloroplasts[18]. This means that the changes reflect structural rearrangements in the thylakoid membrane, most likely reversible disintegration of the LHC domains.

It was shown that these structural changes are driven by photosystem 1, rather than by system 2. Differential sensitivity of the changes to ionophores suggest that local ion and/or electric potential gradients[19] formed along the membrane plane play an important role. A disintegration of the large domains may allow LHC to become more mobile and move toward photosystem 1 in the stroma exposed regions.

This suggestion is in line with the "mobile antenna" hypothesis[20]. According to this hypothesis a fraction of LHC dissociates itself from photosystem 2 and moves toward the stroma membrane regions enriched in photosystem 1. Our results suggest that this becomes possible by a reversible disintegration of the LHC domains, a process controlled by local electric and/or ion gradients.

EXCITATION ENERGY TRANSFER IN THE DOMAINS

An efficient utilization of light energy requires a highly organized antenna system. Even under high light intensities, direct excitation of the reaction centers is a very rare event and in most cases light is gathered by the bulk pigments, in largest part by those of LHC. The energy migrates through a maze of several hundred pigment molecules until it is trapped in the photochemical reaction centers. If it is not trapped it is emitted as fluorescence or dissipated as heat, leading to a loss in the photosynthetic utilization of light energy.

It is now well established[12,21] that the Q_y dipoles of chlorophylls, which are responsible for energy transfer, are preferentially oriented parallel to the membrane plane. This orientation favors the energy migration in this direction. This makes possible a long-range transport of excitation energy[12], and so reduces the possible losses which would occur due to inaccessibility of open reaction centers in the close vicinity of the absorbing dipoles. First of all, however, this type of energy transfer requires domains with small intermolecular distances between the adjacent complexes. This structural requirement for a long-range, fast, in-plane energy transfer in the

thylakoid membranes of chloroplasts can be met by the large liquid crystal like domains of LHC. Disintegration or rearrangement of these domains may lead to a regulatory redistribution of the excitation energy between the two types of photochemical reaction centers.

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