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Review

Hierarchical organization and structural flexibility of thylakoid membranes ☆

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ABSTRACT

Chloroplast thylakoid membranes accommodate densely packed protein complexes in ordered, often semi-crystalline arrays and are assembled into highly organized multilamellar systems, an organization warranting a substantial degree of stability. At the same time, they exhibit remarkable structural flexibility, which appears to play important – yet not fully understood – roles in different short-term adaptation mechanisms in response to rapidly changing environmental conditions. In this review I will focus on dynamic features of the hierarchically organized photosynthetic machineries at different levels of structural complexity: (i) isolated light harvesting complexes, (ii) molecular macroassemblies and supercomplexes, (iii) thylakoid membranes and (iv) their multilamellar membrane systems. Special attention will be paid to the most abundant systems, the major light harvesting antenna complex, LHCII, and to grana. Two physical mechanisms, which are less frequently treated in the literature, will receive special attention: (i) thermo-optic mechanism —elementary structural changes elicited by ultrafast local heat transients due to the dissipation of photon energy, which operates both in isolated antenna assemblies and the native thylakoid membranes, regulates important enzymatic functions and appears to play role in light adaptation and photoprotection mechanisms; and (ii) the mechanism by which non-bilayer lipids and lipid phases play key role in the functioning of xanthophyll cycle de-epoxidases and are proposed to regulate the protein-to-lipid ratio in thylakoid membranes and contribute to membrane dynamics. This article is part of a Special Issue entitled: Dynamic and ultrastructure of bioenergetic membranes and their components.

1. Introduction

The light reactions of oxygenic photosynthesis, the absorption of light, the regulated supply of excitation energy into the photochemical reaction centers and the functioning of the vectorial electron and proton transport system as well as the synthesis of the primary products of the energy conversion depend largely on the architecture of the photosynthetic machinery. In all organisms the photosynthetic apparatuses are highly organized systems with well recognizable hierarchical architecture. They also exhibit astounding variations between different organisms and habitats and, the topic of this review, possess substantial structural and functional flexibilities of the structural complexity in response to rapidly changing environmental conditions. This will be discussed in detail in this review, the major aim of which is to provide deeper insights into the mechanisms and physiological significances of different reorganizations at different levels of the structural complexity. When looking for examples, light-harvesting complex II (LHCII) and the granal thylakoid membranes (TMs) are the evident choices. This is justified by their high abundance in nature and the rich literature available. With regard to the molecular mechanisms the review will focus on two topics, which have received relatively little attention in the literature: thermo-optic effect, an excitation energy-dissipation-assisted mechanism of structural changes, and the role of non-bilayer lipids and lipid phases in the structure and dynamics of TMs.

Abbreviations: CD, circular dichroism; Chl, chlorophyll; DDE, diadinoxanthin de-epoxidase; DGDG, digalactosyl-diacylglycerol; EPR, electron paramagnetic resonance; FCP, fucoxanthin-chlorophyll protein complex; H_{II}, inverted hexagonal phase of lipids; LHCI, light-harvesting complex of Photosystem I; LHCII, light-harvesting complex II; LHCII-HL, LHCII isolated from high light treated leaves; MGDG, monogalactosyl-diacylglycerol; NPQ, non-photochemical quenching; PG, phosphatidyl-glycerol; PLL, poly-L-lysine; psi, polymer or salt-induced; PSI, Photosystem I; PSII, Photosystem II; qE, energy-dependent quenching; RD, repeat distance; SANS, small-angle neutron scattering; SQDG, sulfoquinovosyl diacylglycerol; TM, thylakoid membrane; VDE, violaxanthin de-epoxidase; WT, wild type

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2. Structural hierarchy and flexibility at different levels of structural complexity

As most biological entities, oxygenic photosynthetic organisms are organized in a hierarchical manner, satisfying the conditions that "each level in the hierarchy represents an increase in organizational complexity, with each object being primarily composed of the previous level's basic unit" and "the concept of emergence —the properties and functions found at a hierarchical level are not present and irrelevant at the lower levels" [1].

2.1. Pigment–protein complexes

With regard to the light reactions of photosynthesis, the basic building blocks, the pigment and protein molecules are assembled into pigment–protein complexes. In these the dense packing and non-random orientation of the pigment molecules at well defined binding sites give rise to numerous interactions, e.g. short-range pigment–pigment (dipole–dipole) excitonic interactions as well as different pigment–protein interactions [2]. The molecular architecture of the complexes generally determines the fate of excitation energy and the photophysical pathways within the complexes. The correlation between structure and function at the level of light harvesting complexes can be demonstrated best on LHCII, the most abundant membrane protein in the Biosphere, which is also one of the most studied proteins. Its high resolution crystal structure is known [3,4]. LHCII also participates in important regulatory functions [5], which require structural flexibility of the protein. Some features of this functional and structural plasticity of LHCII can be recognized in vitro, at the level of isolated complexes.

2.1.1. Light-induced changes and protein dynamics in LHCII

Jennings and coworkers [6] were the first to show that isolated, detergent-solubilized LHCII is capable of undergoing light-induced reversible fluorescence quenching with rates proportional to the intensity of the excitation. Thermodynamic analyses of the transients measured both on solubilized complexes and lamellar aggregates have shown that they are associated with conformational changes in the protein complexes —with an estimated 8.8 kJ/M enthalpy of activation for the forward direction [7]. The changes were more pronounced in lamellar aggregates than in trimers, showing that a single quenching center, presumably a trimer that had undergone a (reversible) conformational change, was capable to quench a large domain in the aggregate [8].

The light-induced reorganizations of LHCII appeared not to affect the excitonic interactions, as shown by the invariance of the excitonic CD bands —suggesting that the conformational changes are minor [9]. In lamellar aggregates of LHCII, which undergo substantial reorganizations in the long-range chiral order of the complexes (see Section 2.2), light-induced reversible release of Mg^{2+} and flash-induced fast photoelectric signal attributed to charge-displacement currents were also observed — suggesting that the changes occur in the hydrophilic loops responsible for cation binding [10,11]. Direct experimental evidence for the involvement of the stromal side loop was provided by in vitro phosphorylation experiments. It has been shown that the phosphorylation site of isolated LHCII, which can be cleaved by trypsin, is sensitive to the preillumination of the complexes [12]. These data are also in perfect harmony with recent findings that the amino acids at the N-terminus possess high structural flexibility. Pulsed EPR measurements on spin-labeled LHCII revealed that LHCII possesses rigid cores and flexible hydrophilic domains at the N-terminus [13]. (The N-terminus is missing from the LHCII crystal structure either due to disorder or heterogeneity [4].) Using single particle fluorescence microscopy, it has been shown that detergent-solubilized LHCII trimers immobilized on poly-L-lysine (PLL) undergo rapid conformational changes, switching between fluorescing and dark states [14–16]. These conformational switches, although observed with excitation light intensities much higher than in the macroscopic experiments, might explain the ability of LHCII to participate in light-induced reversible reorganizations. As suggested earlier by a different study, using time-resolved fluorescence and hydrostatic pressure on solubilized LHCII, there is a local conformational switch between the quenched and unquenched state; the estimated free energy difference of 7.0 kJ/mol is allowing the switch to operate in a controlled way [17].

2.1.2. pH stability of LHCII

Since the acidification of the lumenal pH is known to induce qE, the energy-dependent NPQ component in vivo [5], the effect of lowering the pH on LHCII is of special interest. Krüger and coworkers [14–16] have conducted systematic studies on PLL-immobilized detergent solubilized LHCII trimers and observed changes in the operation of the conformational switch using the single-molecule spectroscopy technique. The observed low-pH induced changes, with immobilized trimers allowing no aggregation, were, however, accounted for by the effect of pH on the interaction between LHCII and PLL rather than on the protein structure. In good agreement with this conclusion, it has been shown that reconstituted wild type LHCII exhibited virtually no sensitivity to pH variations; in contrast, the S123G lumenal section mutant showed a low-pH-induced reversible quenching [18]. Further, it has been shown that the lumenal loop segment between helix B and helix C plays a key role in maintaining the structural stability of the complex under acidic conditions, and the negatively charged residues in this loop regulate the pigment conformation and the structural stability under different pH environments [19]. Exchanging glutamic acid at position 94 with glycine (E94G), far from the pigment binding sites, destabilizes the 3_{10} helix in the lumenal loop structure and leads to an acquired pH sensitivity of the LHCII, and also affects the excitonic CD bands assignable to the neoxanthin-binding domain. This conclusions, i.e. that a subtle change in the lumenal loop of LHCII, induced by single amino acid mutagenesis, can generate a sensitivity to low pH and establish NPQ, have recently been confirmed [20].

2.2. Oligomers, aggregates and supercomplexes

Different pigment–protein complexes readily self-assemble into oligomeric forms and supercomplexes. At this level of hierarchy, in addition to the complexes themselves, different other compounds play important roles. For instance, digalactosyl-diacylglycerol (DGDG) facilitates the formation of the extended arrays of LHCII [21]. Different lipid molecules are found both in PSII and PSI supercomplexes in stoichiometries similar to that of the host TMs [22]. This might, in part, be the consequence of their embedding in the lipid membranes but it is more likely to indicate that bound lipid molecules play special roles in the assembly and functional regulation of the reaction center complexes, as shown e.g. for PG in the case of formation of fully functional PSII supercomplexes in vivo [23] or in the trimerization of LHCII monomeric complexes [4]. Other compounds which appear to play important roles in the self-assembly and stability of supercomplexes include small intrinsic membrane proteins, such as the Ycf4 for PSI [24] and PsbW for PSII [25] —their roles are not fully understood. The supply of reaction centers with excitation energy is regulated by varying the peripheral light harvesting antenna [26]. Most prominently, in green algae and higher plants, variations in LHCII arrays play important regulatory roles, and thus their spectroscopic features and light-induced reorganizations deserve special attention.

2.2.1. Spectral signatures of LHCII aggregates

With the assembly of isolated LHCII into large, ordered aggregates, novel spectral properties emerge. One of the most prominent change occurs in the fluorescence quantum yield of LHCII, which drops by an order of magnitude upon the formation of aggregates, when compared to the solubilized complexes [27]. This is also manifested in substantial shortening of the lifetime of the Chl-*a* singlet excited state, from about 4 ns to a few hundred picoseconds [28–30].

Significant aggregation-induced changes have been reported to occur also in the LD and CD spectra [31]. However, at least part of the differences between the detergent solubilized trimer and the LHCII aggregate might originate from a perturbation of the molecular architecture of the complexes by the detergent, as indicated first by triplet-minus-singlet transient spectroscopy [32], and also by a more recent CD and LD spectroscopic study, when comparison was made to the native TMs and small aggregates [33]. Detergents appear to affect mainly the orientation of carotenoids, and a few excitonic CD bands arising from carotenoid:chlorophyll interactions. In general, these

changes occur upon changing the physico-chemical environment of the complexes, i.e. when the trimers are removed from their native environments, lipids and proteins (mainly LHCII complexes) and are exposed to detergents. Both the absence of protein–protein interactions and the presence of detergent affect the spectra. In particular, different distortions can be recognized in the presence of different detergents; protein–protein interactions also give rise to some CD bands (P. Akhtar, K. Pawlak G. Garab and P.H. Lambrev, unpublished). Also, the 4 ns lifetime in detergent-solubilized LHCII might reflect, at least in part, a perturbation of the structure by the detergent compared to the native state [34]. The progress of solid-state magic-angle spinning (MAS) NMR technique will probably allow a detailed comparison of the structure of detergent solubilized LHCII to the crystal structure and to identify the nature of light-induced structural changes [35].

In lamellar aggregates possessing long-range order of the complexes, large, anomalously shaped, so-called 'psi-type' bands emerge both in the blue and red regions with long tails outside the principal absorbance bands. They are superimposed onto the excitonic CD bands, which are retained [36]. Hence, the CD spectra of these lamellar aggregates are not any more recomposable from the spectra (in fact, spectrum) of the constituent complexes —indicating the formation of a higher hierarchy structural unit, in which the whole is more than the sum of its parts. (At the same time, the absorption spectrum of lamellar aggregates, if corrected for scattering and sieving, is almost superposable on the spectrum obtained after disrupting the aggregates by detergents at a concentration close to its critical micelle concentration [37].) In microcrystalline lamellar aggregates, which are largely deficient in lipids, the intensity of the new bands is usually more than an order of magnitude higher than the excitonic CD bands of the solubilized LHCII [38] (Fig. 1). The long-range order of the complexes can also be recognized by the high diamagnetic anisotropy of the samples [36], and can also be visualized using scanning transmission electron microscopy [39]. In loosely stacked lamellar aggregates, obtained with an isolation method similar to the one introduced by Krupa et al. [40], which retains much more lipids than the tightly stacked microscrystalline lamellar aggregates, the psi-type bands are weaker, but they are still significantly more intense than the excitonic bands of the isolated complexes [38]. Psi-type aggregates are three-dimensional chirally organized macroaggregates, possessing sizes commensurate with the wavelength of the visible light and containing a high density of interacting chromophores [41,42].

2.2.2. Light-induced reversible structural changes in lamellar aggregates of LHCII

Highly organized molecular macroassemblies are of interest not only for their complex structures and unusual spectroscopic fingerprints but also for their outstanding structural flexibility, which was discovered by Barzda et al. [9]. Most prominently, as already mentioned in Section 2.1, loosely stacked lamellar aggregates are capable of undergoing light-induced dark-reversible reorganizations, which affect their chiral macro-organization (psi-type CD bands) but does not concern the basic architecture (excitonic CD) of the complexes [9] (Fig. 2a). Disregarding the physical mechanism, this sensitivity of the lamellar aggregates of LHCII to light is evidently correlated with the fact that, in general, the structural stability of these macroarrays is considerably lower than that of the complexes. (For the physical mechanism, see Section 3.1). Indeed, loosely stacked lamellar aggregates of LHCII exhibit a relatively low thermal stability, as shown by the disappearance of the psi-type CD bands; the chirally organized macroarrays of the complexes disassemble at ~45 °C [43]. At the same time, the excitonic CD bands remain essentially unaffected even at considerably higher temperatures. It is interesting to note here that this thermal instability is quite similar to that of the chiral macrodomains of isolated granal TMs, where the psi-type bands disappear in about the same temperature range, between 45 and 50 °C, whereas most excitonic CD bands remain essentially unaffected up to 70 °C [44]. Also, the trimer-to-monomer conversion occurs with a transition temperature at around 55 °C, well below the denaturation temperature but higher than the transition temperature between the macro-arrays and the trimers. (This transition can also be identified by its characteristic excitonic CD fingerprint). Monomerization can also be induced by light at room temperature but it requires much higher light intensities than what is efficient in the disassembly of the macroarrays. It is to be noted that tightly stacked microcrystalline LHCII, which is largely deficient in lipids, displays no light-induced changes —evidently, due to its very high stability [43].

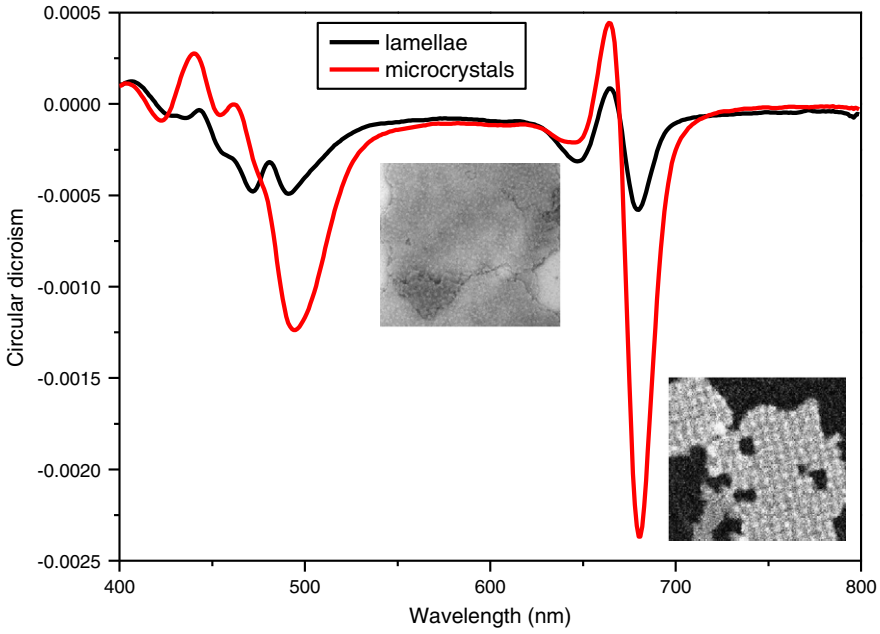

Fig. 1. CD spectra of tightly stacked, microcrystalline and loosely stacked lamellar aggregates of LHCII (red and black curves, respectively), re-plotted, with permission from Springer, based on data in [38], showing also segments of scanning transmission electron micrograph of the microcrystals, exhibiting high order with 128 ± 3 Å mean particle spacing within rows and an estimated particle of 118 ± 7 kDa, as in [39], and segments of the negative staining electron micrograph of the more loosely organized lamellae (I. Simidjiev, G. Garab and A. Holzenburg, unpublished).

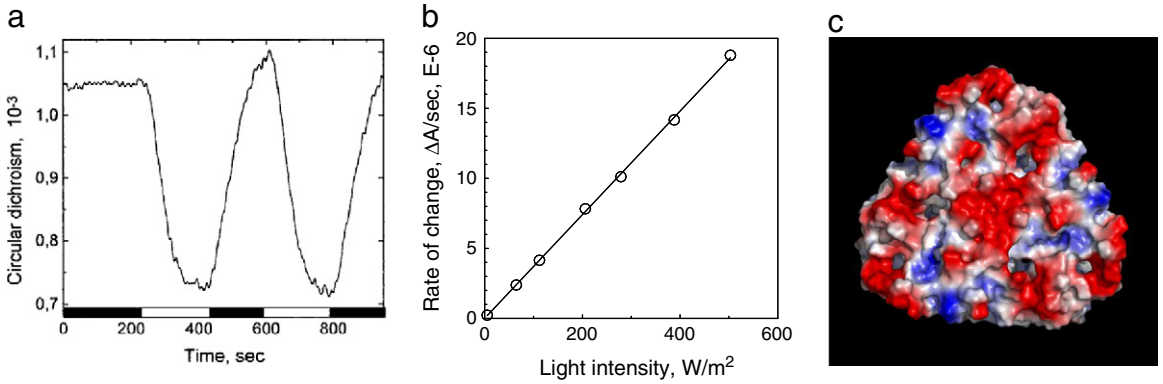

Fig. 2. Light-induced, dark-reversible changes of the psi-type CD signal of loosely stacked lamellar aggregates of LHCII (a) (light and dark periods are indicated by empty and full horizontal bars, respectively, reproduced from [38]), and its dependence on the excitation intensity (b) (reproduced from [47]) (permissions from Elsevier); panel (c) shows the Van-der-Waals surface representation of the stromal side of the trimeric LHCII (apoprotein) indicating the calculated partial charges. The figure is created by P. H. Lambrev using the "Light-Harvesting Complex II" PDB structure 2BHW [4].

It has been shown that addition of isolated thylakoid lipids substantially enhances the light-induced reversible reorganizations in LHCII macroarrays [45]. This might be due to a more complete embedding of the complexes in the 2D lipid matrix but special roles of different lipids cannot be ruled out either. This latter assumption is supported by the fact that loosely stacked LHCII and different purified thylakoid lipids assemble into different structures [45]. They form somewhat disordered loosely stacked lamellae with PG or SQDG (sulfoquinovosyl diacylglycerol), stacked unilamellar vesicles with DGDG and large loosely stacked multilamellar vesicles, often onion-like assemblies with MGDG. In a more systematic recent study, it has been reported that addition of the neutral galactolipids, MGDG and DGDG leads to the aggregation of disorganized structures of the lipid-depleted complexes, whereas the anionic lipids (SQDG and PG) exert a strong disaggregating effect, which could partly be suppressed under high proton concentration and in the presence of (other) cations, such as Mg^{2+} [46].

The structural changes of lipid:LHCII lamellar aggregates exhibit some unusual characteristics: an almost perfectly linear light intensity dependence (Fig. 2b) and a strong and characteristic, non-Arrhenius type temperature dependence – with the transients almost 'frozen' below 10 °C increased rates between ~15 and 30 °C, and gradual weakening and disappearance of the transients above 35 °C [47].

The existing data strongly suggest that electrostatic interactions play important role in the formation and stability of the LHCII macroarrays (see Section 2.1.1). In this context, it is interesting to note that lamellar aggregates of LHCII have been shown to release Mg^{2+} ions [11], mimicking the release of cations from the native TM. Oriented LHCII lamellae also displayed a fast (<200 ns, instrument-limited) photoelectric signal, attributed to a rapid charge displacement, evidently related to the release of Mg^{2+} ions. The released cations are probably liberated from the binding sites of the stromal side, negatively charged residues (Fig. 2c), which have been proposed to be involved in stacking interactions known [3,4] and also contain the phosphorylation sites (see Section 2.1.1). These features, in particular the temperature dependence as well as the linear light intensity dependence, can be interpreted within the frameworks of thermo-optic mechanism [47], which also accounts for the cation release at the N-terminal (for more details see Section 3.1).

Gruszecki et al. [48], using similar LHCII preparations, samples obtained from the isolation method of Krupa et al. [40], used different techniques to detect the light-induced changes in LHCII macroassemblies. By means of resonance Raman scattering technique, they observed that upon strong illumination, violaxanthin changed its molecular configuration from the more twisted to the more relaxed one. Since the more twisted molecular configuration of violaxanthin is characteristic of the pigment located within the binding pocket in LHCII it was concluded that overexcitation of the antenna protein results in the liberation of violaxanthin from the protein environment accompanied by its transfer into the lipid phase of the TM [48]. This is in harmony with the mechanism of the operation of the xanthophyll cycle under high light conditions and the functioning of the violaxanthin de-epoxidase (VDE) in the lipid phase of the membrane [49]. The fact that violaxanthin is localized at the border of the trimer-forming monomeric subunits suggests also that such a light-driven change of the molecular configuration of this xanthophyll might be associated with and accompany the light-induced LHCII trimer to monomer transition observed by Garab et al. [11], and might be involved in NPQ [50]. Recently Zubik et al. observed light-induced molecular configuration changes of the LHCII-bound neoxanthin from 9′-cis to 9′,13- and 9′,13′-dicis forms [51]. The fact that this light-induced conversion is spontaneously reversible implies that it can have a physiological significance. It has been suggested that such a light-driven change of molecular configuration of the LHCII-bound neoxanthin removes steric hindrance and enables close contact of the two neighboring LHCII trimers, which results in the formation of the low-energy chlorophyll spectral forms which can act as energy traps and non-radiative dissipation centers [51,52]. Earlier it has been shown by resonance Raman spectroscopy that qE is correlated with the twisting of a neoxanthin molecule in the light-harvesting antenna [53]. Very recently, Gruszecki and coworkers compared lipid–protein membranes, highly organized supramolecular assemblies composed of isolated thylakoid lipids (MGDG and DGDG) and LHCII isolated from dark-adapted leaves and from leaves preilluminated with high-intensity light (LHCII-HL), in which the complexes were partially phosphorylated and contained zeaxanthin [54]. They found drastic differences in almost all structural features between the two lipid:LHCII samples, tested with X-ray diffraction, infrared imaging microscopy, confocal laser scanning microscopy, transmission electron microscopy and fluorescence lifetime imaging. While the "dark-adapted" lipid:LHCII membranes assembled into planar multibilayers, favoring the stacking of layers, the LHCII-HL membranes formed less ordered structures. LHCII-HL proved to be active in excitation energy quenching.

All these findings, revealing remarkable structural and functional plasticity of LHCII lamellar aggregates, suggest that this structural entity, with its high susceptibility to light- and heat-induced reorganizations, lends the thylakoid membranes high flexibility. Hence, it is clear that this 'structural unit' of LHCII macroarrays participates in important light and temperature adaptation processes in vivo. The physiological significance of their reorganizations and the underlying physical mechanism are discussed in Section 3.1.

2.3. The thylakoid membrane

The structural unit that is composed primarily of supercomplexes but constitutes a significantly higher complexity is the TM. In the

hierarchy, it represents a higher level for several obvious reasons and as the consequence of higher level organization additional elements can be identified in the structural flexibility of the photosynthetic machinery.

2.3.1. Whole chain electron transport, redox sensing, state transitions

TMs are capable of performing whole chain electron transport from water to NADP but cyclic electron transport and (chloro-)respiratory pathways can also be active. Each of these different electron transport pathways evidently requires different arrangements of the components and thus changes in the regime must have structural consequences and/or are linked to some membrane reorganization which, however, might not be easily detectable if confined to the non-pigmented components.

The redox state of the plastoquinone pool depends on the activity of the two photosystems—this is sensed by the cytochrome b_6/f complex which activates the LHCII protein kinase. This redox-controlled, reversible phosphorylation of LHCII regulates the distribution of excitation energy between the two photosystems: the phosphorylated LHCII (LHCII-P) dissociates from PSII and thus reduces its absorption cross section; LHCII-P can dock to PSI and act as PSI antenna (state 2). In the reverse process, when the LHCII kinase is switched off, due to the oxidation of the plastoquinone molecules, LHCII-P becomes dephosphorylated by a phosphatase, and it returns to PSII (state 1). These transitions, called state transitions, evidently require major reorganizations in the TM. This topic has been reviewed thoroughly in the literature (for recent reviews see [55,56]), thus, it will not be discussed here in more details, except two aspects: (i) regulation of the phosphorylation of LHCII by light at the substrate level [12] and (ii) the effect of phosphorylation on the structural flexibility of LHCII in the TMs [57,58].

As already pointed out above (see Section 2.1.1) phosphorylation of LHCII is also regulated by light at the substrate level by light-induced conformational changes of LHCII [12,59]. It is tempting to suggest that this conformational change is an essential element in the molecular recognition mechanism, which has been proposed to regulate the membrane stacking and lateral heterogeneity in the TM [60].

Recently it has been shown, mainly by means of microscopic fluorescence recovery after photobleaching measurements on wild type (WT) and kinase mutants of *Arabidopsis*, that phosphorylation switches the membrane to a more fluid state, which is proposed to facilitate the PSII repair cycle [58]. This is in line with an earlier conclusion according to which phosphorylation of LHCII and other phosphoproteins enhances the structural dynamics of membranes [57]. In particular, it has been shown that in leaves phosphorylation decreases the thermal stability of the PSII:LHCII chiral macrodomains by about 6 °C, compared to the non-phosphorylated transition temperature, at around 48 °C. In addition, the trimer-to-monomer transition temperature also decreased, from 55.6 ± 1.2 to 48.6 ± 1.6 °C, measured in isolated TMs. Phosphorylation also enhanced the light-induced disassembly of the chiral macrodomains and the monomerization of the LHCII trimers at 25 °C [57] —in good accordance with the correlation between the heat and light stability of different photosynthetic assemblies and the thermo-optic mechanism [61]. The effect of phosphorylation on the light- and heat-induced monomerization of LHCII trimers strongly suggests that phosphorylation facilitates the removal of the excess LHCII, since the LHCII protease is only active on the monomeric complexes [62]. This might thus also be part of the overall, long-term regulation of the membrane composition because plastoquinone also controls the relative rates of transcription of PSI and PSII reaction center genes [63].

2.3.2. Transmembrane electrochemical potential gradient, NPQ, ion movements

In the closed membrane vesicle of the TMs, the vectorial electron transport system, coupled to proton translocation processes, creates a transmembrane electrochemical potential gradient between the inner and outer aqueous phases, the lumen and the stroma side (cytoplasmic side in cyanobacteria). This proton-motive force, consisting of an electrical field and a ΔpH component, is utilized for the synthesis of ATP; it also modulates the electron transport rate. The transmembrane electric field is also required for metabolite and protein transport across the membranes, while the ΔpH component is involved in regulatory mechanisms related to the controlled dissipation of the excess excitation by NPQ mechanisms (for general reviews, see [64,65]). With regard to the mechanism of NPQ there are numerous excellent reviews, which also deal with the possible role of major or (according to other authors) just minor changes in the macro-organization of the TM, on the role of PsbS and other proteins as well as on the xanthophyll cycle [66–70]. Here, no overview is offered on these subjects; instead, in the following paragraphs some considerations are presented on the structural and functional roles of ion movements, which accompany the formation and decay of ΔpH and which are also induced photophysically in vitro.

It has been well established that upon the formation of electrical field and ΔpH the ions must be redistributed in the electrolyte of the aqueous phase [71]. Light-induced reversible release of Mg^{2+} (and other ions) to the stroma, coupled to the inward flux of protons, has long been documented [72]. On the other hand, as mentioned above (see Section 2.1.1), light-induced dark-reversible Mg^{2+}-release has also been reported to occur in lamellar aggregates of LHCII —showing that the release of cations can be independent from and does not require photochemical activity and the formation of a transmembrane electrical field or ΔpH [10,11]. In good accordance with this, the rate of the light-induced release of Mg^{2+} ions has been shown to display an apparently non-saturable, approximately linear light intensity dependence (measured up to 500 W m^{-2}, corresponding to ~2500 μmol photons m^{-2} s^{-1}), similar to the light intensity dependence of the light-induced CD changes (Fig. 3a). These features closely resemble those seen in lamellar aggregates (see Section 2.2.2). Also, the thermal stability of the chiral macrodomains in the thylakoid membranes is very similar to that of the loosely stacked lamellar aggregates of LHCII: these macroarrays are disassembled at ~45 °C in the dark, or at room temperature by light (Fig. 3b).

It is interesting to note that the magnitude of NPQ exhibits a strong dependence on the concentration of Mg^{2+} ions between 2 and 15 mM [73] and, although ΔpH is generated, ceases when the suspension is depleted of these ions [10]. These data suggest a more intimate relationship between cation movements and NPQ than expected based on a simple control via the acidification of the lumen. Similar correlation appears to exist between the formation of chiral macrodomains (the magnitude of the psi-type CD) and their ability to undergo light-induced reorganizations, which also depend on the concentration of Mg^{2+} ions [74]. In fact, it appears to be valid not only for Mg^{2+} but, albeit with different concentration dependences, also for mono- and trivalent cations [75]. Naturally occurring polyamines, such as spermin and spermidine also had marked effects on the macro-organization and structural flexibility of granal TMs (Istokovics and Garab, unpublished). Chloroplastic polyamines have been shown to be important structure and functionality of the TMs [76]. It is also noteworthy that both the F_o and F_m levels of the Chl *a* fluorescence of granal TMs increase monotonically with the increase of the cation concentrations [77]. All these findings can most probably be explained by a complex interrelationship between the macro-organization and structural flexibility of the LHCII:PSII domains, the modulation of the fluorescence yield, and the light-induced movements of cations.

2.3.3. The lipid bilayer, non-bilayer lipids and lipid phases

The functional state of TMs is bilayer membrane, which provides electrical insulation between the two aqueous phases and an impermeability for ions. TMs are capable of holding a transmembrane uniform field of about 100 kV/cm, and allow the build-up of a ΔpH of 2–3 units; this is essential for the utilization of the proton-motive force for the synthesis of ATP. However, given the fact that about half of the thylakoid lipids belong to the class of non-bilayer (or non-lamella-forming) lipids,

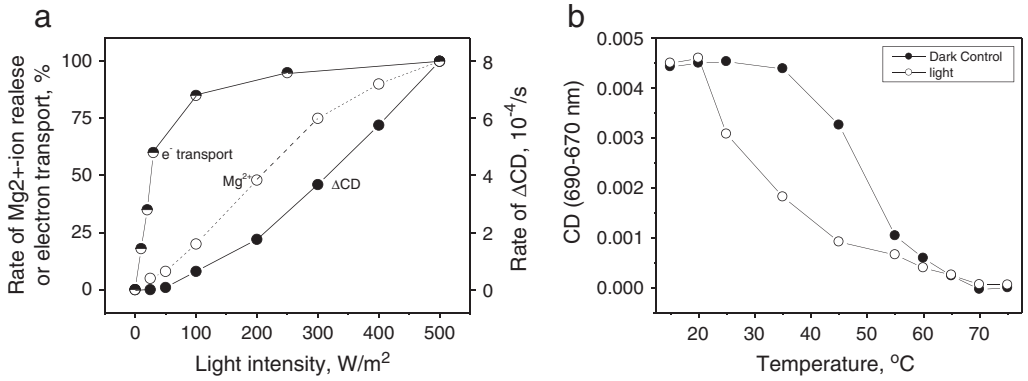

Fig. 3. Light-intensity dependences of the linear electron transport, ΔCD (light-induced CD change) and Mg^{2+}-ion release of freshly isolated pea thylakoid membranes (a) (redrawn from data published in [9] and [10]), and the effect of on the amplitude of the red psi-type CD band of pea thylakoid membranes at different temperatures (b) (reproduced from [44]). (Permissions from the American Chemical Society.).

which do not self-assemble into bilayers in aqueous media [78,79], it is not straightforward to understand how the bilayer is formed and what is the role of these lipids in the bilayer. In the native TMs more than 60% of the lipids are found in the bulk phase, i.e. not bound to proteins [80] —these structural lipids must be found in the bilayer, in which protein complexes are embedded.

It has been shown that the non-bilayer lipid MGDG can be forced into a bilayer structure upon its association with LHCII [45,81]. It has also been pointed out that, because of the high non-bilayer propensity of the thylakoid lipids, due to the presence of ~50% MGDG, the TMs must contain high concentrations of protein [82,83]. This is because this lipid composition, with only ~50% bilayer lipids (DGDG, ~30%, and PG and SQDG, ~10-10%) the presence of large protein-free areas in the membranes are not allowed since upon their exposure to water they would form non-bilayer lipid phases and segregate out from the bilayer membrane. This ability of segregation has thoroughly been documented by electron microscopy, which revealed that lipids are extruded from TMs upon different stresses and in the presence of different co-solutes (reviewed in [79]).

While MGDG might have multiple roles in the TMs and in different protein complexes, its role in the functioning of the xanthophyll cycle has been well established. The xanthophyll cycle plays important photoprotective role in plants and algae via its involvement in NPQ and the strong antioxidant properties of the de-epoxidized compounds [84–88]. It involves the enzymatic removal of epoxy groups from xanthophylls, violaxanthin and antheraxanthin in plants and green algae and diadinoxanthin in diatoms and dinoflagellates, to obtain de-epoxidized xanthophylls, zeaxanthin and diatoxanthin. These conversions are performed by the lumenal enzymes, VDE and diadinoxanthin de-epoxidase (DDE), respectively.

It has been shown that the functioning of VDE requires the presence of MGDG for its activity [89] and the presence of non-bilayer lipid phases [90]. In a series of works, the groups of Goss, Strzalka and Wilhelm conducted systematic investigations on model systems and in vivo to understand the role of this non-bilayer lipid in the de-epoxidation of the bilayer embedded xanthophylls. They have established that MGDG serves to solubilize the hydrophobic xanthophyll cycle pigments so that they become accessible to the enzymes, VDE or DDE. Otherwise the xanthophylls form aggregates in an aqueous medium which cannot be converted by these enzymes. MGDG and (the non-TM non-bilayer lipid) phosphatidyl ethanolamine have been shown to have a better solubilization capacity than the bilayer lipid DGDG and (the non-TM bilayer lipid) phosphatidyl choline [91]. MGDG has also been shown to form a lipid shield around LHCII and FCP (the fucoxanthin–chlorophyll protein complex). This shield contains free xanthophylls. In high light cultivated diatoms (which have a large xanthophyll pool) the MGDG shield incorporates a large number of diadinoxanthin and diatoxanthin molecules, which serve as antioxidants and provide a reservoir for the synthesis of light-harvesting pigments. The MGDG shield is also an attraction site for the VDE or DDE, which targets the enzymes to the place where the majority of xanthophyll cycle pigments are located [49,92]. The lipid arrangement of the thylakoid membrane. i.e. the phase behavior of lipids has also been proposed to vary between low light and high light conditions [93]. MGDG forms inverted hexagonal (H_{II}) structures which are essential for efficient de-epoxidation. In liposomes composed of H_{II} and bilayer lipids a certain percentage of non-bilayer lipids is needed for the de-epoxidation reaction to occur [94]. These data are in perfect harmony with the fact that VDE is a lipocalin type of molecule and thus readily enters the lipid phase [95]. As revealed by Arnoux et al. [96], using a truncated *Arabidopsis thaliana* VDE expressed in *Escherichia coli*, the central lipocalin domain of VDE at neutral pH is monomeric with its active site occluded within a lipocalin barrel, which opens upon acidification and appears as a dimer.

2.4. The multilamellar thylakoid membrane system

As concerns the light reactions of photosynthesis, it might be argued that the TMs represent the highest level in the hierarchy of the photosynthetic machineries. However, TMs in all mature chloroplasts and cyanobacteria are assembled into multilamellar membrane systems, which thus should be taken into account. In principle, the layers might be able to operate independently of each other. However, as it will be argued in the following paragraphs, this is usually not the case, and thus we should consider the multilamellar system as a separate, highly organized structural unit in the hierarchy.

As one of the main consequences of the multilamellarity of the TM, when compared to a single TM, is the 'replacement' of the free outer aqueous phase with narrow interthylakoidal space. In the grana, i.e. in the stacked region this space is in fact narrower than the lumenal space, 3.2 nm and 4.5 nm, respectively, according to recent cryo-electron microscopy data [97,98]. Although experiments using chymotrypsin suggest somewhat larger (4.5 nm wide) interthylakoidal space [99], in grana this region can be portrayed as being fully packed with proteins, more precisely with the polypeptide sections of PSII and LHCII, protruding to the stromal side. Hence, the packing density of the interthylakoidal space appears to be very high and, without partial or local unstacking, would restrict protein motions, such as that of the kinase but also the diffusion of D1 and LHCII proteins.

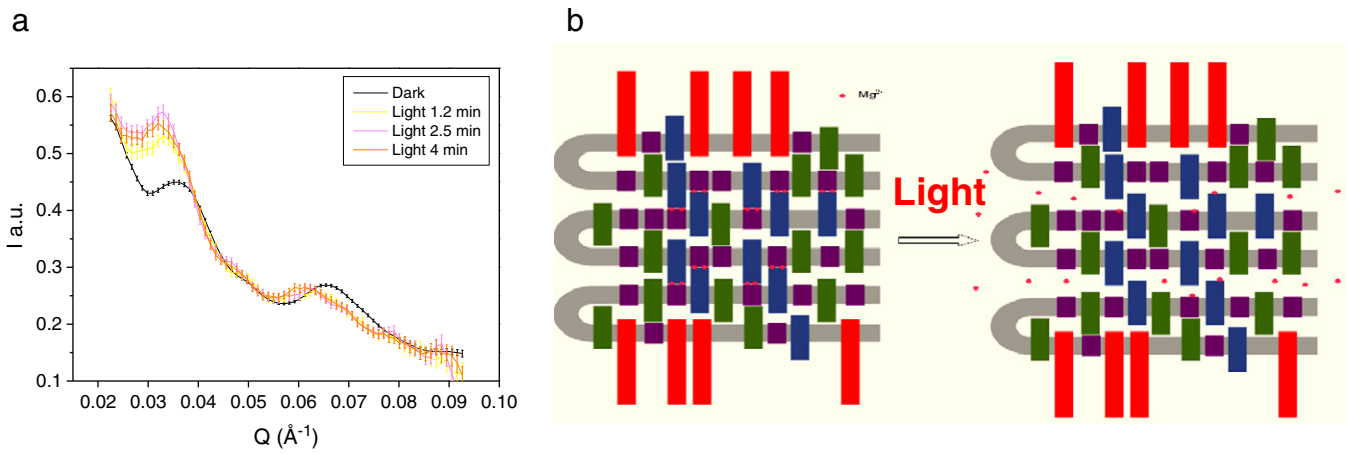

Fig. 4. Effect of illumination with white light of 2000 μmol photons m^{-2} s^{-1} photon flux density on the SANS profile of thylakoid membranes in living *Phaeodactylum tricornutum* cells (a). Similar, fully reversible changes were published in [100]. The schematic model (drawn by Gergely Nagy) shows the possible arrangement of the main protein complexes (red, ATP synthase; blue, PSI; green, PSII; purple, FCP) in the membranes, as allowed by the SANS data and the even distribution of the complexes (see text for further details); it also shows the light-induced expansion of RD —which is insensitive to NH_4Cl (b). Red dots represent Mg^{2+} ions in the interthylakoidal region, which are hypothesized to be released upon illumination.

Data obtained for the repeat distances (RDs) from small-angle neutron scattering (SANS) measurements on cyanobacteria and diatoms corroborate the conclusion on the dense packing of interthylakoidal space [100]. In the diatom *Phaeodactylum tricornutum*, a RD of ~17 nm evidently requires tight packing of membranes, given the even distribution of PSII and PSI in the TM: with an estimated membrane thickness of 4 nm (2×) and lumenal space of 4.5 nm leaves only 4.5 nm for the interthylakoidal space, which must accommodate the protruding parts of PSI, approximately 4 nm [101] and also that of the FCP, which has somewhat shorter protruding section than LHCII [102] (Fig. 4a). It is to be noted, however, that the tight packing might also reflect a partial segregation of PSII and PSI. According to the model of Lepetit et al. [69], the inner membranes, though not free of PSI, contain domains that are enriched in MGDG and PSII along with oligomerized peripheral FCP complexes, while the outer lamellae are enriched in SQDG and PSI complexes with their specific FCP antennae and accommodate the ATP synthase. Liberton et al. [103] found close correlation between the RDs and the external antenna sizes in different phycobilisome mutants (see also [100]) —showing tight packing of the interthylakoidal space also in cyanobacteria. As stressed above, this evidently restricts the mobility of the proteins in this region. In the light of these findings, it is not surprising that the TM RDs reversibly increase in these organisms upon their illumination [100,103,104]. This expansion is to a considerable extent attributed to the increase of the interthylakoidal space, which evidently can facilitate the mobility of complexes and of enzymes functioning in this region (Fig. 4b). It is interesting to note that in the case of *P. tricornutum* the RD changes closely correlate with changes in the psi-type CD bands [103]. As suggested by the decreased stacking capability of LHCII-HL (see Section 2.2.2), the interthylakoidal space might also increase in high-light-treated leaves [54].

The importance of multilamellarity is also evident when protein–protein interactions and membrane stacking are to be considered in the case of granal TMs, where the membranes are laterally separated into stacked and unstacked regions, which are enriched in PSII and PSI, respectively [105]. The formation of this highly organized multilamellar system is evidently initiated by the lateral segregation of PSII and LHCII from PSI, which can occur because of the formation of LHCII:PSII macrodomains —leaving out PSI. The LHCII:PSII macrodomains, in turn, with their flat surfaces can stack to each other and form a 3D structure of the grana, while PSI and the ATP synthase with their large protruding subunits on the stromal side reside in the stroma thylakoid membranes [106]. The CD fingerprint of this multilamellar structure is similar to that of the lamellar aggregates of LHCII [41,107]. In general, since psi-type aggregates are three-dimensional macroaggregates [41] many multilamellar systems might exhibit such features [108,109]. It must, however, be noted that multilamellarity per se does not give rise to psi-type CD bands, which also require a long-range chiral order of the complexes in the membrane system (e.g. [110]).

The multilamellar TM systems of plants and some algae have also been shown to undergo light-induced reorganizations [61,111–113]; the same is true for low-pH-induced changes in the psi-type bands [114,115]. These reorganizations might in part be related to the changes in the multilamellar organization of the membranes [100,103,104], which evidently affects the 3D array of chromophores but lateral reorganizations are also involved, as revealed mainly by CD spectroscopy [74]. It is to be stressed here, that as shown for granal TMs, the light-induced reversible changes display an apparently non-saturable light intensity dependence, with rates linearly dependent on the light intensity above the saturation of the linear electron transport and the ΔpH, tested up to 500 W m^{-2} (~2500 μmol photons m^{-2} s^{-1}) [9]. This shows that light-induced reversible changes in the chiral macrodomains are capable of 'measuring' the light intensity above the saturation of photosynthesis and to respond to it by proportional reversible reorganizations, which is thus evidently part of the light adaptation and photoprotection mechanism of plants. Very similar light-intensity dependence was obtained by using SANS and measuring the rate of RD changes [116]. This ability of the TM is evidently 'borrowed' from LHCII, which possesses the same capability of responding to excitation (see Section 2.2.2 and [111]).

In the case of *P. tricornutum*, grown at 40 μmol photons m^{-2} s^{-1} light intensity, the rates and magnitudes of CD changes increase substantially in the range of 40 and 450 μmol photons m^{-2} s^{-1} but saturated above that [117]. The RD changes, which were essentially insensitive to NH_4Cl, were also accelerated substantially at high light illumination but retained the reversibility even after several minutes of illumination with strong light (2000 μmol photons m^{-2} s^{-1}) (G. Nagy, R. Ünnep, O. Zsiros, G. Finazzi and G. Garab, unpublished). As concerns the physical mechanism and the possible physiological roles, see Section 3.1.

3. Physical mechanisms and physiological significances

In this section I would like to direct the attention to two topics, the mechanism, effects physiological significances of dissipation and the dissipation-assisted, thermo-optically driven reorganizations on the

one hand, and the why and wherefores of non-bilayer lipids and lipid phases in or associated with the bilayer TMs.

3.1. Thermo-optic mechanism

In this section, first, I briefly recall the main experimental findings which led to the proposal of a novel thermo-optic mechanism, which offers explanation for the observations that cannot be accounted for by other 'photosynthetic mechanisms', such as redox feedback or ΔpH; this will be followed by describing the essence of thermo-optic mechanism, and by outlining its possible physiological functions.

Briefly recalling, the following experimental findings on light-induced reversible conformational changes of different LHCII preparations, described in the previous sections, required explanation and were explained in terms of thermo-optic mechanism: (i) light-induced reversible fluorescence quenching [6,7]; (ii) psi-type CD

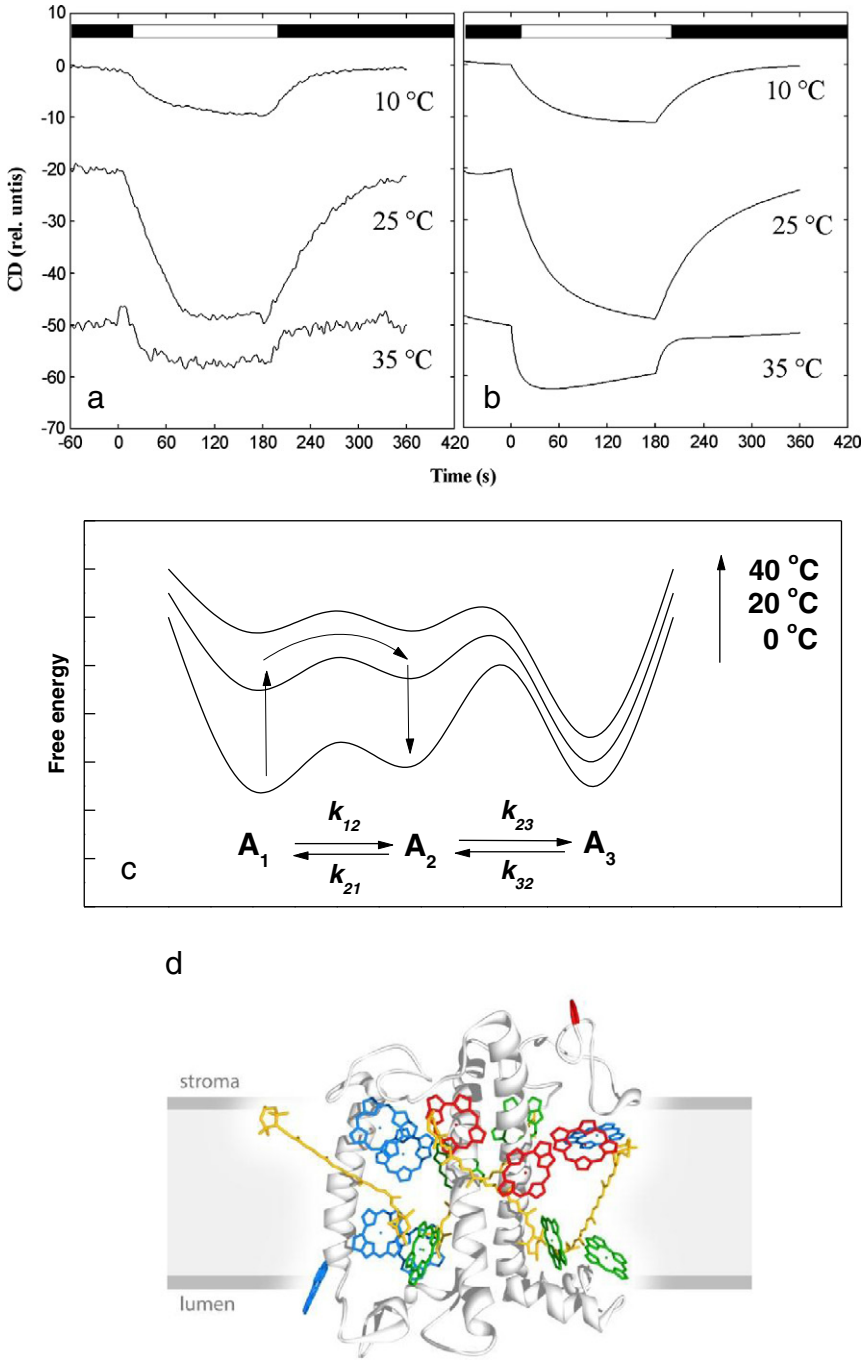

Fig. 5. Light-induced, dark reversible changes in the psi-type CD signal of loosely stacked lamellar aggregates of LHCII at different temperatures: panels a and b, kinetics observed in the experiments and calculated based on the thermo-optic mechanism, respectively (reproduced from [44]; permission from Springer). Shown in panel c, the potential profiles of simplified three-state model of LHCII, that are used in the mechanism of thermo-optic transitions marked by arrows: upon internal conversion of an excited-state molecule, the local temperature jumps, this ultrafast transition can facilitate an elementary structural transition —by this means, and because of their slow reversibility, A2 state (or the irreversible A3 state) can be accumulated (reproduced from [11]; permission from the American Chemical Society). Panel d, crystal structure of monomeric LHCII ("Light-Harvesting Complex II" PDB entry 2BHW, image by P. H. Lambrev), illustrating the site of dissipation/terminal emitter Chl-*a* molecules (drawn in red ink) and the site of missing N-terminus section (in red). This missing polypeptide section contains the phosphorylation site and very likely cation binding sites, too, and possesses high flexibility [13]; its conformation can be changed by illuminating the complexes both in vivo and in vitro [12] —most likely via thermo-optic effect, i.e. assisted by dissipation [9,11].

changes observed on loosely stacked lamellar aggregates of LHCII [38] and similar apparently non-saturable changes in granal TMs; (iii) light-induced reversible release of Mg^{2+} ions from lamellar aggregates of LHCII and, again, the release of Mg^{2+} ions with an apparently non-saturable light intensity dependence in TMs [10,11]; (iv) reversible light-induced conformational changes, which expose the phosphorylation site of LHCII, i.e. the regulation of phosphorylation by light at the substrate level, which are also observed in TMs [12,59]; (v) the peculiar, non-Arrhenius temperature dependence of the light-induced CD changes in lamellar aggregates of LHCII and TMs [47] (Fig. 5a), and (vi) light-induced monomerization of LHCII trimers in vitro and in vivo [11]. Further observations, which have been interpreted in terms of light-induced reversible effects on carotenoids in lamellar aggregates of isolated LHCII appear to be consistent with the thermo-optic mechanism include: (vii) light- and heat-induced structural changes in the temperature-dependent fluorescence quenching on LHCII suspensions and conformational changes in monomolecular films [118]; and (viii) the light-induced liberation of violaxanthin from the protein environment [48]; and (ix) light-induced formation of the low-energy chlorophyll spectral forms which are assigned to originate from a change in the supramolecular structure of LHCII —leading to closer contact of individual trimers [52].

The above experiments clearly established that isolated LHCII is capable of undergoing light-induced dark-reversible structural changes. As already discussed, the primary site of this effect appears to be at the N-terminal region, responsible for phosphorylation and cation binding (cf. also [4]), which is indeed a structurally flexible domain [13]. Structural changes in this region might also be responsible for minor variations in trimer–trimer contacts observed in [52]; it remains to be clarified whether or not light-induced changes in the conformation or binding of carotenoids can also be associated with conformational changes in the loop sections in the protein structure or the two effects are independent.

Regarding the physical mechanism, photochemical mechanisms and redox- or ΔpH-feed-back mechanisms can be ruled out for most of the cases, and thus the solution must be found in photophysics. When seeking for explanation, the analogy of thermo-optic effect in liquid crystals was used, in liquid crystals, the long-range order can be reversibly perturbed by dissipation of photon energy in the (macroscopic) path of the laser beam [119]. By using this analogy, for LHCII, it was proposed that the light-induced structural changes in LHCII originate from elementary dissipation-assisted structural changes. In other terms, these changes are accounted for by a novel, biological (or molecular, rather than macroscopic) thermo-optic mechanism: fast thermal transients, arising from dissipated excitation energy, can lead to elementary structural transitions in the close vicinity of the site of dissipation due to the presence of 'built-in' thermal structure-instabilities [11,44]. This mechanism offers explanation for the non-Arrhenius type temperature dependence of the structural changes (Fig. 5b) using a simple model, the essence of which is shown in Fig. 5c. This illustrates that the reaction temperature, transiently, is obtained from a 'combination' of the ambient temperature and the local temperature due to the heat package arising from the dissipation. This might lead to minor elementary structural changes in the close vicinity (<1 nm) of the dissipation. For LHCII, this can be envisioned in the following manner: Dissipation events occur in the domain containing the lowest energy Chl-*a* molecules [120,121], displayed in red in Fig. 5d; this, in turn, induces vibrations, most readily in the highly flexible sections near the N-terminus (a missing polypeptide section in the crystal structure, also in red) [13], which contains the phosphorylation site [12] and might also contain cation binding sites. This site might represent a 'built-in' thermally instable element that, via conformational changes and/or a cation release, might be involved in dissipation-assisted, i.e. thermo-optically driven reorganizations of the electrostatically controlled stacked 2D aggregates of LHCII in vivo and in vitro (see below and Section 2.1.1).

The presence of specific thermally instable structural elements has been shown to be involved, using CD spectroscopy, which revealed the existence of reasonably sharp transients; essentially the same CD changes could be induced by elevating the temperature as with illumination at considerably lower temperatures. (However, the heat-induced changes are irreversible, probably because they affect all units whereas the light-induced changes are confined to the actual site of dissipation). Theoretical considerations have shown that the magnitude of the heat transient due to dissipation can be sensed only at short distances, in about a 1 nm radius around the dissipation center. The occurrence of thermal transients due to dissipation in LHCII has been shown, using exciton–exciton annihilation technique. These experiments revealed that, in accordance with the expectations based on a simple model [44], the thermal transients are very fast, they exhibit two decay times of about 20 and 200 ps [122]. Thermal instabilities have also been shown to exist both in LHCII preparations and in the native thylakoid membranes system. In these samples the thermal disassembly followed the following sequence: unstacking, lateral disorganization and LHCII trimer-to-monomer transitions, and finally denaturation of the complexes; the same order was found for the susceptibility of structures to light [61]. In the hierarchically organized multilamellar structures the interactions between adjacent lamellae are evidently the weakest, held together mainly by weak, electrostatic interactions [61,74]; this is followed by the lateral order, which is destabilized after unstacking. The disassembly of thermally more stable units, such as the trimers, requires higher temperatures and higher light intensities (more dissipation) [11]. These observations, together with the strong temperature dependences [47] gave support to the thermo-optic mechanism.

In this context, it must be pointed out that since stacking is mediated largely electrostatically via protein–protein interactions [4,74], there is no reason to assume that partial unstacking occurs only at the marginal regions. Partial or transient unstacking might equally occur in the middle of the grana stack, e.g. due to local fluctuations, or induced thermo-optically, via the release of cations due to local dissipation events. This mechanism would largely facilitate the mobility of the complexes, which are required, e.g. for the operation of PSII repair mechanisms [123,124].

So-called hot molecules, formed by internal conversion from an excited singlet state to a highly vibrationally excited ground electronic state have earlier been shown to facilitate different reactions and can play a role as an intermediate step of e.g. multiphoton dissociations [125]. Also, photoinduced reduction has been shown to occur mainly in the ground state of hot cytochrome c, i.e. following an ultrafast internal conversion of oxidized cytochrome c; modest heating of the sample has also been shown to lead to its efficient thermal reduction [126]. Nevertheless, our understanding of the mechanism of dissipation is still rudimentary mainly because the detection of the rise and dispersal of the heat packages in the molecular microenvironment is not straightforward. To this end, light harvesting complexes with designed dissipation centers and very fast dissipation would be needed —this is presently available only for reconstituted bacterial light harvesting complex (LH1) containing Ni-bacteriochlorophyll [127,128].

The better understanding of the processes related to dissipation in light harvesting antennae would be important because dissipation is a basic phenomenon, its statistical frequency under natural conditions might probably be as high as that of the energy utilization. Evidently, the molecular architecture of light harvesting complexes must be stable enough to resist the dissipation-induced heat transient. On the other hand, as shown for LHCII (and some other complexes, see below), they might possess the ability to respond to the unused excitation with well defined structural changes, due to the presence of a built-in thermally instable structural elements (see above). These thermo-optically induced reorganizations appear to occur with a very low quantum yield (of about 10^{-6} or smaller —Lambrev and Garab,

unpublished), and by this means respond only to persistently excess excitation, as one would expect for a light adaptation mechanism.

Despite the low quantum yield of these dissipation-assisted reorganizations, they appear to have important physiological roles —which can be summed up as follows: (i) As pointed out above, in LHCII, they regulate the phosphorylation of LHCII, not only in vitro but also in vivo [12,59,129]. (ii) The trimer-to-monomer transition is required for the enzymatic removal of LHCII because trimers cannot be digested by the specific protease [62]. (iii) Several lines of evidence suggest that the reorganizations, which are very similar in the grana and in loosely stacked lamellar aggregates of LHCII, play important roles in NPQ — this is indicated by similarities in their thermal and light instabilities, cation releases, and by the effects of inhibitors, as well as by the (limited) sensitivity of thylakoids ΔpH [9,130,131], and especially by the features observed above the saturation of photosynthesis. Data in Janik et al. [54] have demonstrated the physiological significance of the conformational changes of LHCII induced by high light in the membrane. These affected their associations and macrostructures in vitro, which in turn influenced their abilities to participate in fluorescence quenching. Nevertheless, direct correlation between the thermo-optically induced reorganizations in the LHCII:PSII macroarrays and NPQ may not exist because NPQ also requires additional components, such as the PsbS protein and/or a de-epoxidized xanthophyll [66]. Thus it seems most likely that these reorganizations do not induce directly but rather facilitate the quenching, or may even be required for an efficient NPQ.

The ability of LHCII to undergo thermo-optically driven reorganizations is not unique among the light harvesting antenna complexes. Light- or heat-induced detachment of the phycobilisome antenna has also been observed, which could also be accounted for by a thermo-optic mechanism [132]. In good agreement with these data, Tamary et al. [133] have shown that nonradiative energy dissipation in the phycobilisome induces alterations in thermo-labile elements, most likely in rod and core linker polypeptides, which then leads to the disruption of the excitation energy migration and promotes the their dissociation from the thylakoid membranes. Although these changes, via energetically disconnecting part of the phycobilisomes from the membrane, evidently play important roles in the photoprotection, they are not directly involved in NPQ, which also require the involvement of the so-called orange carotenoid protein (OCP) [134,135]. The same appears to apply to the ultrastructural changes, revealed by small-angle neutron scattering both in wild-type and phycobilisome mutant *Synechocystis* cells, i.e. also in the absence of OCP [100,103].

3.2. Roles of non-bilayer lipids and lipid phases

As already pointed out, the non-bilayer lipid is the most abundant lipid MGDG is the major lipid in the bilayer membrane of TMs. Non-bilayer lipids, which in aqueous media do not spontaneously form bilayers, are contained probably in all biological membranes [136] but are most abundant in energy converting membranes (cf. [82]). It has been proposed that by this means, due to the high non-bilayer propensity of the lipid mixture, and thus its ability to segregate if in excess, the protein content of TMs can be self-regulated via controlling their MGDG:DGDG ratios (see Section 2.3.3 and [82]). The galactolipid synthesis, on the other hand, is coordinated with gene expressions in the nucleus and plastids, protein transport into plastids, and the pigment biosynthesis [137]. These mechanisms might be involved in controlling the membrane fluidity under stress conditions [138,139], and might also be responsible, at least in part, for the variations in the lipid:protein ratios and protein crowding in isolated PSII (BBY) membranes and in bundle-sheath TMs, where the variations also affect the protein diffusions [140,141]. These lipids also play important roles in determining the lateral pressure profile within the membrane and thus exert effects on integral membrane proteins [142].

As pointed out in Section 2.3.3, the functioning of VDE requires the presence of a non-bilayer lipid phase in the TM, which must basically remain bilayer, or associated with it. Using ^{31}P NMR, [143] observed the formation of H_{II} phase in Tris-washed TMs; these authors also reported that the lamellar (bilayer) phase co-exists with an isotropic phase, this latter phase could be enhanced by the addition of glycerol and by increasing the temperature to 35 °C. More recent experiments on intact thylakoid membranes revealed that the characteristic lamellar signal is observed only below 20 °C but even at lower temperatures an isotropic phase is present, which becomes dominant between 14 and 28 °C despite the presence of fully functional TMs capable of generating and maintaining a transmembrane electric field [144] (Fig. 6). These data show that in fully functional TMs the lamellar (bilayer) phase co-exists with a non-bilayer isotropic lipid phase. In good agreement with this conclusion, steady state and time-resolved experiments using the fluorescent lipid probe Merocyanine 540, have also shown that TMs cannot be described satisfactorily with the properties of mono-phasic bilayer lipid phase and the mix of the phase behavior depends strongly on temperature [145].

It has also been concluded from NMR experiments that the non-bilayer structures remain in contact with the bilayer membrane and lipid molecules are exchanged between the two phases [144]. Since the enzymatic activity of VDE (and DDE) depends on the presence of non-bilayer lipid phases (see Section 2.3.3) it is reasonable to assume that this lipocalin-type protein (and perhaps other lipocalin proteins in the lumen) is (are) involved in the formation of non-bilayer lipid phases and in the lipid-trafficking between the two phases. This requires further investigations. At any rate, these data show that non-bilayer lipids contribute significantly to the dynamic features of the TMs, in accordance with the hypothesis that has been put forward earlier [82]. Variations in the MGDG:DGDG ratios and thus in the non-bilayer propensity of the lipid mixture has consequences also on the structural flexibility of the TMs, as shown by the significant alterations in the overall organization of the thylakoid membranes and decreased thermal stabilities in dgd1 mutant, deficient in DGDG [146]. In contrast, as concluded from a study using *mgd1* mutant, MGDG appears to play an important role during thylakoid membrane reorganizations under high light conditions —these reorganizations are largely inhibited in the MGDG-deficient mutant [147]. In general, MGDG deficient plants exhibit retarded growth and a chlorotic phenotype, the chloroplasts contain many fewer thylakoid membranes and both PSII activity and the whole chain electron transport are inhibited [148] These data point to the importance of the role of non-bilayer lipids and lipid phases in the structure and function of TMs but our understanding is still quite rudimentary. Molecular dynamics simulations [149] would be needed to understand the coupling between the bilayers, lipocalin proteins and non-bilayer lipid phases in TMs.

4. Conclusions and outlook

Oxygenic photosynthetic organisms evolved multilevel regulatory mechanisms by the aid of which cyanobacteria, algae and plants, living in different habitats, are capable to fine-adjust their functions to rapidly changing environmental conditions, most notably to light quality and intensity and temperature. The functional plasticity evidently requires structural flexibility of the photosynthetic machinery. Conversely, environmental factors may directly cause structural changes that might bring about changes in the functions —these in turn might be coupled to regulatory mechanisms. In either case, the organism responds with structural changes, which can occur at different levels of the structural complexity of the hierarchically organized machinery. It turns out that some of the regulatory functions and structural changes can be traced back to the level of the pigment–protein complexes, which 'lend' their flexibility to the higher level assemblies, while others appear only at the highest level of the hierarchy, the multilamellar membrane systems. The present review focuses on several somewhat arbitrarily selected

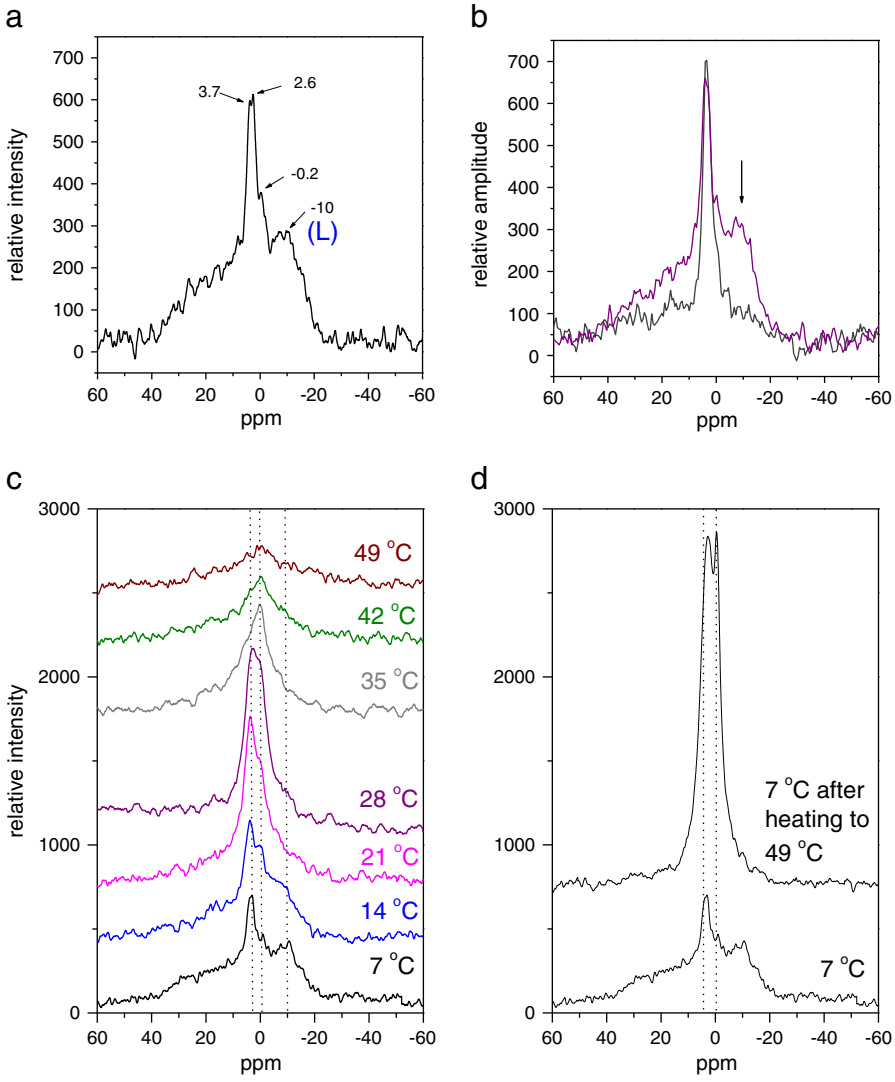

Fig. 6. ^{31}P NMR spectra of freshly isolated intact spinach thylakoid membranes reproduced from [144] with permission from Elsevier. The spectrum recorded at 7 °C clearly shows the signature of the lamellar (bilayer) phase (L); however, it also contains additional intense signals, indicating the presence of other lipid phases (a). The saturation transfer experiment (b), where the irradiation frequency was set at the high intensity peak of the lamellar phase (at −10 ppm), revealed that the intensity of the remaining 4 ppm peak, which is isotropic in nature, is not decreased, suggesting that at 7 °C, there is no considerable magnetization transfer between the lipids experiencing isotropic motion and the ones motionally restricted in the bilayer. As shown by the spectra recorded at elevated temperatures (c), the isotropic phase becomes dominant already between 14 and 28 °C, suggesting that these non-bilayer structures are in contact with the bilayer membrane and the exchange of lipids between the two phases is temperature dependent. After heat treatment the lamellar phase cannot be restored (d).

topics, which are either less frequently reviewed or, in my opinion, might deserve more attention for their physiological importance or just problems which should be solved in order to better understand the structure and function of the given systems.

The main focal points and problems are as follows:

(i) The nature and mechanism of dissipation of the excess excitation—a problem which concerns the molecular architecture and stability vs flexibility of light harvesting complexes, which should allow efficient energy utilization under normal conditions but should be able to switch to a safe dissipation regime upon overexcitation. In this regard, the dissipation-assisted, thermo-optically driven reorganizations deserve special attention because they have been shown to play important roles in enzymatic regulatory processes, such as the phosphorylation and the proteolysis of LHCII. In addition, this mechanism can be identified at all levels of the structural hierarchy, from isolated complexes to the intact membrane system, and appears to provide the structural basis of light adaptation and photoprotection mechanisms.

(ii) An apparently technical question, but important to clarify, what is the relation between the crystal structure and the detergent solubilized proteins, and how these two can be related to the native state of the same protein in the TM. This concerns both the static and dynamic features. Although the basic in vivo properties can evidently be understood using the crystal structure or the detergent-solubilized data, both fall short when the fine details are considered.

(iii) The light-induced reversible changes in the periodic organization of the thylakoid membranes, which are manifested in small (max 1–2 nm) but well discernible and relatively rapid RD changes, occurring on the time scale of seconds and minutes, are apparently present at all levels of the evolutionary tree, from cyanobacteria to whole leaves. The mechanism(s) and significance of these changes are not very well understood.

(iv) One of the enigmatic questions of membrane biology is the role of bulk non-bilayer lipids in the bilayer membranes, here TM, and the properties and roles of non-bilayer lipid phases that are associated with the TM. As in all energy converting membranes, in

TM, too, the major lipid is a non-bilayer lipid, which (MGDG) can be forced into the bilayer by protein aggregation but their presence appears to lend the membranes an additional flexibility. While the role of non-bilayer lipid phase associated with the bilayer membrane and the light harvesting complexes has been satisfactorily clarified in the case of VDE and DDE, these phases might have further, probably more general functions in the overall organization of the membranes, such as the self-regulation of the protein content of the membranes and the self-assembly and dynamics of membranes. If the conclusion, derived from ^{31}P NMR experiments, that there is an intense communication between the bilayer phase and the non-bilayer lipid phase, gets further experimental or molecular dynamics support, we may have to think over the presently accepted model of TMs.

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References

[1] A. Pave, Biological and ecological systems hierarchical organization, in: D. Pumain (Ed.), Hierarchy in Natural and Social Sciences, Springer, New York, 2006, pp. 39–70.
[2] H. van Amerongen, L. Valkunas, R. van Grondelle, Photosynthetic Excitons, World Scientific, Singapore, 2000.
[3] Z. Liu, H. Yan, K. Wang, T. Kuang, J. Zhang, L. Gui, X. An, W. Chang, Crystal structure of spinach major light-harvesting complex at 2.72 Angstrom resolution, Nature 428 (2004) 287–292.
[4] J. Standfuss, A.C.T. van Scheltinga, M. Lamborghini, W. Kühlbrandt, Mechanisms of photoprotection and nonphotochemical quenching in pea light-harvesting complex at 2.5 Å resolution, EMBO J. 24 (2005) 919–928.
[5] P. Horton, A.V. Ruban, R.G. Walters, Regulation of light harvesting in green plants, Annu. Rev. Plant Physiol. Plant Mol. Biol. 47 (1996) 655–684.
[6] R.C. Jennings, F.M. Garlaschi, G. Zucchelli, Light-induced fluorescence quenching in the light-harvesting chlorophyll *a*/*b* protein complex, Photosynth. Res. 27 (1991) 57–64.
[7] V. Barzda, R.C. Jennings, G. Zucchelli, G. Garab, Kinetic analysis of the light-induced fluorescence quenching in light-harvesting chlorophyll *a*/*b* pigment–protein complex of photosystem II, Photochem. Photobiol. 70 (1999) 751–759.
[8] P.H. Lambrev, F.J. Schmitt, S. Kussin, M. Schoengen, Z. Várkonyi, H.J. Eichler, G. Garab, G. Renger, Functional domain size in aggregates of light-harvesting complex II and thylakoid membranes, Biochim. Biophys. Acta 1807 (2011) 1022–1031.
[9] V. Barzda, A. Istokovics, I. Simidjiev, G. Garab, Structural flexibility of chiral macroaggregates of light-harvesting chlorophyll *a*/*b* pigment–protein complexes. Light-induced reversible structural changes associated with energy dissipation, Biochemistry 35 (1996) 8981–8985.
[10] G. Garab, A. Istokovics, A. Butiuc, I. Simidjiev, A. Dér, Light-induced ion movements in thylakoid membranes and isolated LHC II, in: G. Garab (Ed.), Photosynthesis: Mechanisms and Effects, Springer, Netherlands, 1998, pp. 341–344.
[11] G. Garab, Z. Cseh, L. Kovács, S. Rajagopal, Z. Várkonyi, M. Wentworth, L. Mustárdy, A. Der, A.V. Ruban, E. Papp, A. Holzenburg, P. Horton, Light-induced trimer to monomer transition in the main light-harvesting antenna complex of plants: thermo-optic mechanism, Biochemistry 41 (2002) 15121–15129.
[12] H. Zer, M. Vink, N. Keren, H.G. Dilly-Hartwig, H. Paulsen, R.G. Herrmann, B. Andersson, I. Ohad, Regulation of thylakoid protein phosphorylation at the substrate level: reversible light-induced conformational changes expose the phosphorylation site of the light-harvesting complex II, Proc. Natl. Acad. Sci. U. S. A. 96 (1999) 8277–8282.
[13] C. Dockter, A.H. Müller, C. Dietz, A. Volkov, Y. Polyhach, G. Jeschke, H. Paulsen, Rigid core and flexible terminus: structure of solubilized light-harvesting chlorophyll *a*/*b* complex (LHCII) measured by EPR, J. Biol. Chem. 287 (2012) 2915–2925.
[14] T.P.J. Krüger, C. Ilioaia, L. Valkunas, R. van Grondelle, Fluorescence intermittency from the main plant light-harvesting complex: sensitivity to the local environment, J. Phys. Chem. B 115 (2011) 5083–5095.
[15] T.P.J. Krüger, C. Ilioaia, R. van Grondelle, Fluorescence intermittency from the main plant light-harvesting complex: resolving shifts between intensity levels, J. Phys. Chem. B 115 (2011) 5071–5082.
[16] T.P.J. Krüger, C. Ilioaia, M.P. Johnson, A.V. Ruban, E. Papagiannakis, P. Horton, R. van Grondelle, Controlled disorder in plant light-harvesting complex II explains its photoprotective role, Biophys. J. 102 (2012) 2669–2676.
[17] B. van Oort, A. van Hoek, A.V. Ruban, H. van Amerongen, Equilibrium between quenched and nonquenched conformations of the major plant light-harvesting complex studied with high-pressure time-resolved fluorescence, J. Phys. Chem. B 111 (2007) 7631–7637.
[18] C. Liu, Y. Zhang, D. Cao, Y. He, T. Kuang, C. Yang, Structural and functional analysis of the antiparallel strands in the lumenal loop of the major light-harvesting chlorophyll *a*/*b* complex of photosystem II (LHCIIb) by site-directed mutagenesis, J. Biol. Chem. 283 (2008) 487–495.
[19] C. Yang, P. Lambrev, Z. Chen, T. Jávorfi, A.Z. Kiss, H. Paulsen, G. Garab, The negatively charged amino acids in the lumenal loop influence the pigment binding and conformation of the major light-harvesting chlorophyll *a*/*b* complex of photosystem II, Biochim. Biophys. Acta 1777 (2008) 1463–1470.
[20] E. Belgio, C.D.P. Duffy, A.V. Ruban, Switching light harvesting complex II into photoprotective state involves the lumen-facing apoprotein loop, Phys. Chem. Chem. Phys. 15 (2013) 12253–12261.
[21] S. Hobe, S. Prytulla, W. Kühlbrandt, H. Paulsen, Trimerization and crystallization of reconstituted light-harvesting chlorophyll *a*/*b* complex, EMBO J. 13 (1994) 3423–3429.
[22] N. Mizusawa, H. Wada, The role of lipids in photosystem II, Biochim. Biophys. Acta 1817 (2012) 194–208.
[23] Z. Gombos, Z. Várkonyi, M. Hagio, M. Iwaki, L. Kovács, K. Masamoto, S. Itoh, H. Wada, Phosphatidylglycerol requirement for the function of electron acceptor plastoquinone QB in the photosystem II reaction center, Biochemistry 41 (2002) 3796–3802.
[24] E. Boudreau, Y. Takahashi, C. Lemieux, M. Turmel, J.D. Rochaix, The chloroplast ycf3 and ycf4 open reading frames of *Chlamydomonas reinhardtii* are required for the accumulation of the photosystem I complex, EMBO J. 16 (1997) 6095–6104.
[25] J.G. Garcia-Cerdan, L. Kovács, T. Tóth, S. Kereïche, E. Aseeva, E.J. Boekema, F. Mamedov, C. Funk, W.P. Schröder, The PsbW protein stabilizes the supramolecular organization of photosystem II in higher plants, Plant J. 65 (2011) 368–381.
[26] R. Croce, H. van Amerongen, Light-harvesting and structural organization of photosystem II: from individual complexes to thylakoid membrane, J. Photochem. Photobiol. B 104 (2011) 142–153.
[27] P. Horton, A.V. Ruban, D. Rees, A.A. Pascal, G. Noctor, A.J. Young, Control of the light-harvesting function of chloroplast membranes by aggregation of the LHCII chlorophyll–protein complex, FEBS Lett. 292 (1991) 1–4.
[28] S. Vasil'ev, K.-D. Irrgang, T. Schrötter, A. Bergmann, H.-J. Eichler, G. Renger, Quenching of chlorophyll a fluorescence in the aggregates of LHCII: steady state fluorescence and picosecond relaxation kinetics, Biochemistry 36 (1997) 7503–7512.
[29] V. Barzda, C.J. de Grauw, J. Vroom, F.J. Kleima, R. van Grondelle, H. van Amerongen, H.C. Gerritsen, Fluorescence lifetime heterogeneity in aggregates of LHCII revealed by time-resolved microscopy, Biophys. J. 81 (2001) 538–546.
[30] Y. Miloslavina, A. Wehner, E. Wientjes, M. Reus, P. Lambrev, G. Garab, R. Croce, A.R. Holzwarth, Far-red fluorescence: a direct spectroscopic marker for LHCII oligomers forming in non-photochemical quenching, FEBS Lett. 582 (2008) 3625–3631.
[31] A.V. Ruban, F. Calkoen, S.L.S. Kwa, R. van Grondelle, P. Horton, J.P. Dekker, Characterisation of LHC II in the aggregated state by linear and circular dichroism spectroscopy, Biochim. Biophys. Acta 1321 (1997) 61–70.
[32] K.R. Naqvi, T. Jávorfi, T.B. Melo, G. Garab, More on the catalysis of internal conversion in chlorophyll *a* by an adjacent carotenoid in light-harvesting complex (Chl *a*/*b* LHCII) of higher plants: time-resolved triplet-minus-singlet spectra of detergent-perturbed complexes, Spectrochim. Acta A 55 (1999) 193–204.
[33] P. Lambrev, Z. Várkonyi, S. Krumova, L. Kovács, Y. Miloslavina, A.R. Holzwarth, G. Garab, Importance of trimer–trimer interactions for the native state of the plant light-harvesting complex II, Biochim. Biophys. Acta 1767 (2007) 847–853.
[34] E. Belgio, M.P. Johnson, S. Juric, A.V. Ruban, Higher plant photosystem II light-harvesting antenna, not the reaction center, determines the excited-state lifetime—both the maximum and the nonphotochemically quenched, Biophys. J. 102 (2012) 2761–2771.
[35] A. Pandit, T. Morosinotto, M. Reus, A.R. Holzwarth, R. Bassi, H.J.M. de Groot, First solid-state NMR analysis of uniformly ^{13}C-enriched major light-harvesting complexes from *Chlamydomonas reinhardtii* and identification of protein and cofactor spin clusters, Biochim. Biophys. Acta 1807 (2011) 437–443.
[36] V. Barzda, L. Mustárdy, G. Garab, Size dependency of circular dichroism in macroaggregates of photosynthetic pigment–protein complexes, Biochemistry 33 (1994) 10837–10841.
[37] K.R. Naqvi, T.B. Melo, B. Bangar Raju, T. Jávorfi, G. Garab, Comparison of the absorption spectra of trimers and aggregates of chlorophyll *a*/*b* light-harvesting complex LHC II, Spectrochim. Acta A 53 (1997) 1925–1936.
[38] I. Simidjiev, V. Barzda, L. Mustárdy, G. Garab, Isolation of lamellar aggregates of the light-harvesting chlorophyll *a*/*b* protein complex of photosystem II with long-range chiral order and structural flexibility, Anal. Biochem. 250 (1997) 169–175.
[39] Y. Miloslavina, P. Lambrev, T. Jávorfi, Z. Várkonyi, V. Karlický, J. Wall, G. Hind, G. Garab, Anisotropic circular dichroism signatures of oriented thylakoid membranes and lamellar aggregates of LHCII, Photosynth. Res. 111 (2012) 29–39.
[40] Z. Krupa, N.P.A. Huner, J.P. Williams, E. Maissan, D.R. James, Development at cold-hardening temperatures. The structure and composition of purified rye light harvesting complex II, Plant Physiol. 84 (1987) 19–24.
[41] G. Garab, H. van Amerongen, Linear dichroism and circular dichroism in photosynthesis research, Photosynth. Res. 101 (2009) 135–146.

[42] G. Garab, Linear and circular dichroism, in: J. Amesz, A. Hoff (Eds.), Biophysical Techniques in Photosynthesis, Springer, Netherlands, 2004, pp. 11–40.
[43] I. Simidjiev, Z. Várkonyi, P.H. Lambrev, G. Garab, Isolation and characterization of lamellar aggregates of LHCII and LHCII-lipid macro-assemblies with light-inducible structural transitions, in: R. Carpentier (Ed.), Photosynthesis Research Protocols, Springer, Dordrecht, 2011.
[44] Z. Cseh, S. Rajagopal, T. Tsonev, M. Busheva, E. Papp, G. Garab, Thermooptic effect in chloroplast thylakoid membranes. Thermal and light stability of pigment arrays with different levels of structural complexity, Biochemistry 39 (2000) 15250–15257.
[45] I. Simidjiev, V. Barzda, L. Mustárdy, G. Garab, Role of thylakoid lipids in the structural flexibility of lamellar aggregates of the isolated light-harvesting chlorophyll *a*/*b* complex of photosystem II, Biochemistry 37 (1998) 4169–4173.
[46] S. Schaller, D. Latowski, M. Jemiola-Rzeminska, A. Dawood, C. Wilhelm, K. Strzalka, R. Goss, Regulation of LHCII aggregation by different thylakoid membrane lipids, Biochim. Biophys. Acta 1807 (2011) 326–335.
[47] Z. Cseh, A. Vianelli, S. Rajagopal, S. Krumova, L. Kovács, E. Papp, V. Barzda, R. Jennings, G. Garab, Thermo-optically induced reorganizations in the main light harvesting antenna of plants. I. Non-Arrhenius type of temperature dependence and linear light-intensity dependencies, Photosynth. Res. 86 (2005) 263–273.
[48] W.I. Gruszecki, M. Gospodarek, W. Grudzinski, R. Mazur, K. Gieczewska, M. Garstka, Light-induced change of configuration of the LHCII-bound xanthophyll (tentatively assigned to violaxanthin): a resonance Raman study, J. Phys. Chem. B 113 (2009) 2506–2512.
[49] S. Schaller, D. Latowski, M. Jemiola-Rzeminska, C. Wilhelm, K. Strzalka, R. Goss, The main thylakoid membrane lipid monogalactosyldiacylglycerol (MGDG) promotes the de-epoxidation of violaxanthin associated with the light-harvesting complex of photosystem II (LHCII), Biochim. Biophys. Acta 1797 (2010) 414–424.
[50] W.I. Gruszecki, R. Luchowski, M. Zubik, W. Grudzinski, E. Janik, M. Gospodarek, J. Goc, Z. Gryczynski, I. Gryczynski, Blue-light-controlled photoprotection in plants at the level of the photosynthetic antenna complex LHCII, J. Plant Physiol. 167 (2010) 69–73.
[51] M. Zubik, R. Luchowski, W. Grudzinski, M. Gospodarek, I. Gryczynski, Z. Gryczynski, J.W. Dobrucki, W.I. Gruszecki, Light-induced isomerization of the LHCII-bound xanthophyll neoxanthin: possible implications for photoprotection in plants, Biochim. Biophys. Acta 1807 (2011) 1237–1243.
[52] M. Zubik, R. Luchowski, M. Puzio, E. Janik, J. Bednarska, W. Grudzinski, W.I. Gruszecki, The negative feedback molecular mechanism which regulates excitation level in the plant photosynthetic complex LHCII: towards identification of the energy dissipative state, Biochim. Biophys. Acta 1827 (2013) 355–364.
[53] A.V. Ruban, R. Berera, C. Ilioaia, I.H.M. van Stokkum, J.T.M. Kennis, A.A. Pascal, H. van Amerongen, B. Robert, P. Horton, R. van Grondelle, Identification of a mechanism of photoprotective energy dissipation in higher plants, Nature 450 (2007) 575–578.
[54] E. Janik, J. Bednarska, M. Zubik, M. Puzio, R. Luchowski, W. Grudzinski, R. Mazur, M. Garstka, W. Maksymiec, A. Kulik, G. Dietler, W.I. Gruszecki, Molecular architecture of plant thylakoids under physiological and light stress conditions: a study of lipid–light-harvesting complex II model membranes, Plant Cell 25 (2013) 2155–2170.
[55] M. Tikkanen, M. Suorsa, P.J. Gollan, E.-M. Aro, Post-genomic insight into thylakoid membrane lateral heterogeneity and redox balance, FEBS Lett. 586 (2012) 2911–2916.
[56] J. Minagawa, State transitions—the molecular remodeling of photosynthetic supercomplexes that controls energy flow in the chloroplast, Biochem. Biophys. Acta 1807 (2011) 897–905.
[57] Z. Várkonyi, G. Nagy, P. Lambrev, A. Kiss, N. Székely, L. Rosta, G. Garab, Effect of phosphorylation on the thermal and light stability of the thylakoid membranes, Photosynth. Res. 99 (2009) 161–171.
[58] T.K. Goral, M.P. Johnson, A.P. Brain, H. Kirchhoff, A.V. Ruban, C.W. Mullineaux, Visualizing the mobility and distribution of chlorophyll proteins in higher plant thylakoid membranes: effects of photoinhibition and protein phosphorylation, Plant J. 62 (2010) 948–959.
[59] M. Vink, H. Zer, N. Alumot, A. Gaathon, K. Niyogi, R.G. Herrmann, B. Andersson, I. Ohad, Light-modulated exposure of the light-harvesting complex II (LHCII) to protein kinase(s) and state transition in *Chlamydomonas reinhardtii* xanthophyll mutants, Biochemistry 43 (2004) 7824–7833.
[60] J.F. Allen, J. Forsberg, Molecular recognition in thylakoid structure and function, Trends Plant Sci. 6 (2001) 317–326.
[61] A.G. Dobrikova, Z. Várkonyi, S.B. Krumova, L. Kovács, G.K. Kostov, S.J. Todinova, M.C. Busheva, S.G. Taneva, G. Garab, Structural rearrangements in chloroplast thylakoid membranes revealed by differential scanning calorimetry and circular dichroism spectroscopy. Thermo-optic effect, Biochemistry 42 (2003) 11272–11280.
[62] D.-H. Yang, H. Paulsen, B. Andersson, The N-terminal domain of the light-harvesting chlorophyll *a*/*b*-binding protein complex (LHCII) is essential for its acclimative proteolysis, FEBS Lett. 466 (2000) 385–388.
[63] T. Pfannschmidt, A. Nilsson, J.F. Allen, Photosynthetic control of chloroplast gene expression, Nature 397 (1999) 625–628.
[64] D.M. Kramer, J.A. Cruz, A. Kanazawa, Balancing the central roles of the thylakoid proton gradient, Trends Plant Sci. 8 (2003) 27–32.
[65] B. Bailleul, P. Cardol, C. Breyton, G. Finazzi, Electrochromism: a useful probe to study algal photosynthesis, Photosynth. Res. 106 (2010) 179–189.
[66] K.K. Niyogi, Photoprotection revisited: genetic and molecular approaches, Annu. Rev. Plant Physiol. Plant Mol. Biol. 50 (1999) 333–359.
[67] P. Jahns, A.R. Holzwarth, The role of the xanthophyll cycle and of lutein in photoprotection of photosystem II, Biochim. Biophys. Acta Bioenerg. 1817 (2012) 182–193.
[68] P. Horton, Optimization of light harvesting and photoprotection: molecular mechanisms and physiological consequences, Philos. Trans. R. Soc. Lond. B Biol. Sci. 367 (2012) 3455–3465.
[69] B. Lepetit, R. Goss, T. Jakob, C. Wilhelm, Molecular dynamics of the diatom thylakoid membrane under different light conditions, Photosynth. Res. 111 (2012) 245–257.
[70] S. Matsubara, B. Förster, M. Waterman, S.A. Robinson, B.J. Pogson, B. Gunning, B. Osmond, From ecophysiology to phenomics: some implications of photoprotection and shade–sun acclimation in situ for dynamics of thylakoids in vitro, Philos. Trans. R. Soc. Lond. B Biol. Sci. 367 (2012) 3503–3514.
[71] L. Zimányi, G. Garab, Configuration of the electric field and distribution of ions in energy transducing biological membranes: model calculations in a vesicle containing discrete charges, J. Theor. Biol. 138 (1989) 59–76.
[72] G. Hind, H.Y. Nakatani, S. Izawa, Light-dependent redistribution of ions in suspensions of chloroplast thylakoid membranes, Proc. Natl. Acad. Sci. U. S. A. 71 (1974) 1484–1488.
[73] G. Noctor, D. Rees, A. Young, P. Horton, The relationship between zeaxanthin, energy-dependent quenching of chlorophyll fluorescence, and trans-thylakoid pH gradient in isolated chloroplasts, Biochim. Biophys. Acta 1057 (1991) 320–330.
[74] G. Garab, J. Kieleczawa, J.C. Sutherland, C. Bustamante, G. Hind, Organization of pigment–protein complexes into macrodomains in the thylakoid membranes of wild-type and chlorophyll *b*-less mutant of barley as revealed by circular dichroism, Photochem. Photobiol. 54 (1991) 273–281.
[75] A. Istokovics, Királis makrodomének struktúraváltozásai kloroplasztisz tilakoid membránjaiban, Ph.D. thesis Szeged University, Hungary, 1997.
[76] N.E. Ioannidis, K. Kotzabasis, Effects of polyamines on the functionality of photosynthetic membrane in vivo and in vitro, Biochim. Biophys. Acta 1767 (2007) 1372–1382.
[77] J. Barber, G.F.W. Searle, Cation-induced increase in chlorophyll fluorescence yield and effect of electrical charge, FEBS Lett. 92 (1978) 5–8.
[78] V. Luzzati, X-ray diffraction studies of lipid–water systems, in: D. Chapman (Ed.), Biological Membranes, Academic Press, New York, 1968, pp. 71–123.
[79] W.P. Williams, The physical properties of thylakoid membrane lipids and their relation to photosynthesis, in: P.-A. Siegenthaler, N. Murata (Eds.), Advances in Photosynthesis. Lipids in Photosynthesis, Kluwer Academic Publishers, Dordrecht, The Netherlands, 1998, pp. 103–118.
[80] T. Páli, G. Garab, L.I. Horváth, Z. Kóta, Functional significance of the lipid–protein interface in photosynthetic membranes, Cell. Mol. Life Sci. 60 (2003) 1591–1606.
[81] I. Simidjiev, S. Stoylova, H. Amenitsch, T. Jávorfi, L. Mustárdy, P. Laggner, A. Holzenburg, G. Garab, Self-assembly of large, ordered lamellae from non-bilayer lipids and integral membrane proteins in vitro, Proc. Natl. Acad. Sci. U. S. A. 97 (2000) 1473–1476.
[82] G. Garab, K. Lohner, P. Laggner, T. Farkas, Self-regulation of the lipid content of membranes by non-bilayer lipids: a hypothesis, Trends Plant Sci. 5 (2000) 489–494.
[83] H. Kirchhoff, U. Mukherjee, H.J. Galla, Molecular architecture of the thylakoid membrane: lipid diffusion space for plastoquinone, Biochemistry 41 (2002) 4872–4882.
[84] B. Demmig-Adams, Carotenoids and photoprotection in plants: a role for the xanthophyll zeaxanthin, Biochim. Biophys. Acta 1020 (1990) 1–24.
[85] B. Demmig-Adams, W.W. Adams III, Antioxidants in photosynthesis and human nutrition, Science 298 (2002) 2149–2153.
[86] M. Havaux, K.K. Niyogi, The violaxanthin cycle protects plants from photooxidative damage by more than one mechanism, Proc. Natl. Acad. Sci. U. S. A. 96 (1999) 8762–8767.
[87] M. Havaux, L. Dall'Osto, R. Bassi, Zeaxanthin has enhanced antioxidant capacity with respect to all other xanthophylls in *Arabidopsis* leaves and functions independent of binding to PSII antennae, Plant Physiol. 145 (2007) 1506–1520.
[88] M.P. Johnson, M. Havaux, C. Triantaphylides, B. Ksas, A.A. Pascal, B. Robert, P.A. Davison, A.V. Ruban, P. Horton, Elevated zeaxanthin bound to oligomeric LHCII enhances the resistance of *Arabidopsis* to photooxidative stress by a lipid-protective, antioxidant mechanism, J. Biol. Chem. 282 (2007) 22605–22618.
[89] H.Y. Yamamoto, R.M. Higashi, Violaxanthin de-epoxidase. Lipid composition and substrate specificity, Arch. Biochem. Biophys. 190 (1978) 514–522.
[90] D. Latowski, H.E. Akerlund, K. Strzalka, Violaxanthin de-epoxidase, the xanthophyll cycle enzyme, requires lipid inverted hexagonal structures for its activity, Biochemistry 43 (2004) 4417–4420.
[91] R. Goss, M. Lohr, D. Latowski, J. Grzyb, A. Vieler, C. Wilhelm, K. Strzalka, Role of hexagonal structure-forming lipids in diadinoxanthin and violaxanthin solubilization and de-epoxidation, Biochemistry 44 (2005) 4028–4036.
[92] B. Lepetit, D. Volke, M. Gilbert, C. Wilhelm, R. Goss, Evidence for the existence of one antenna-associated, lipid-dissolved and two protein-bound pools of diadinoxanthin cycle pigments in diatoms, Plant Physiol. 154 (2010) 1905–1920.
[93] R. Goss, C. Wilhelm, Lipids in algae, lichens and mosses, in: H. Wada, N. Murata (Eds.), Lipids in Photosynthesis, Springer, Netherlands, 2010, pp. 117–137.
[94] R. Goss, S. Oroszi, C. Wilhelm, The importance of grana stacking for xanthophyll cycle-dependent NPQ in the thylakoid membranes of higher plants, Physiol. Plant. 131 (2007) 496–507.
[95] A.D. Hieber, R.C. Bugos, H.Y. Yamamoto, Plant lipocalins: violaxanthin de-epoxidase and zeaxanthin epoxidase, Biochim. Biophys. Acta 1482 (2000) 84–91.
[96] P. Arnoux, T. Morosinotto, G. Saga, R. Bassi, D. Pignol, A structural basis for the pH-dependent xanthophyll cycle in *Arabidopsis thaliana*, Plant Cell 21 (2009) 2036–2044.
[97] B. Daum, D. Nicastro, J. Austin II, J.R. McIntosh, W. Kühlbrandt, Arrangement of photosystem II and ATP synthase in chloroplast membranes of spinach and pea, Plant Cell 22 (2010) 1299–1312.

[98] H. Kirchhoff, C. Hall, M. Wood, M. Herbstová, O. Tsabari, R. Nevo, D. Charuvi, E. Shimoni, Z. Reich, Dynamic control of protein diffusion within the granal thylakoid lumen, Proc. Natl. Acad. Sci. U. S. A. 108 (2011) 20248–20253.
[99] R.C. Jennings, F.M. Garlaschi, G. Forti, P.D. Gerola, Evidence for a structural role for chlorophyll in chlorophyll–protein complexes, Biochim. Biophys. Acta 581 (1979) 87–95.
[100] G. Nagy, D. Posselt, L. Kovács, J.K. Holm, M. Szabó, B. Ughy, L. Rosta, J. Peters, P. Timmins, G. Garab, Reversible membrane reorganizations during photosynthesis in vivo: revealed by small-angle neutron scattering, Biochem. J. 436 (2011) 225–230.
[101] J.P. Dekker, E.J. Boekema, Supramolecular organization of thylakoid membrane proteins in green plants, Biochim. Biophys. Acta 1706 (2005) 12–39.
[102] C. Wilhelm, C. Büchel, J. Fisahn, R. Goss, T. Jakob, J. LaRoche, J. Lavaud, M. Lohr, U. Riebesell, K. Stehfest, K. Valentin, P.G. Kroth, The regulation of carbon and nutrient assimilation in diatoms is significantly different from green algae, Protist 157 (2006) 91–124.
[103] M. Liberton, L.E. Page, W.B. O'Dell, H. O'Neill, E. Mamontov, V.S. Urban, H.B. Pakrasi, Organization and flexibility of cyanobacterial thylakoid membranes examined by neutron scattering, J. Biol. Chem. 288 (2013) 3632–3640.
[104] G. Nagy, M. Szabó, R. Ünnep, G. Kali, Y. Miloslavina, P.H. Lambrev, O. Zsiros, L. Porcar, P. Timmins, L. Rosta, G. Garab, Modulation of the multilamellar membrane organization and of the chiral macrodomains in the diatom *Phaeodactylum tricornutum* revealed by small-angle neutron scattering and circular dichroism spectroscopy, Photosynth. Res. 111 (2012) 71–79.
[105] J.M. Anderson, B. Andersson, The dynamic photosynthetic membrane and regulation of solar-energy conversion, Trends Biochem. Sci. 13 (1988) 351–355.
[106] G. Garab, L. Mustárdy, Role of LHCII-containing macrodomains in the structure, function and dynamics of grana, Aust. J. Plant Physiol. 26 (1999) 649–658.
[107] D. Keller, C. Bustamante, Theory of the interaction of light with large inhomogeneous molecular aggregates. II. Psi-type circular dichroism, J. Chem. Phys. 84 (1986) 2972–2980.
[108] C. Büchel, G. Garab, Molecular organisation of the chlorophyll *a*/*c* light-harvesting complex of *Pleurochloris meiringensis* (Xanthophyceae). Pigment binding and secondary structure of the protein, J. Photochem. Photobiol. B 42 (1998) 191–194.
[109] R. Goss, C. Wilhelm, G. Garab, Organization of the pigment molecules in the chlorophyll *a*/*b*/*c* containing alga *Mantoniella squamata* studied by means of absorption, circular and linear dichroism spectroscopy, Biochim. Biophys. Acta 1457 (2000) 190–199.
[110] L. Kovács, J. Damkjaer, S. Kereïche, C. Ilioaia, A.V. Ruban, E.J. Boekema, S. Jansson, P. Horton, Lack of the light-harvesting complex CP24 affects the structure and function of the grana membranes of higher plant chloroplasts, Plant Cell 18 (2006) 3106–3120.
[111] J.K. Holm, Z. Várkonyi, L. Kovács, D. Posselt, G. Garab, Thermo-optically induced reorganizations in the main light harvesting antenna of plants. II. Indications for the role of LHCII-only macrodomains in thylakoids, Photosynth. Res. 86 (2005) 275–282.
[112] G. Garab, R.C. Leegood, D.A. Walker, J.C. Sutherland, G. Hind, Reversible changes in macroorganization of the light-harvesting chlorophyll *a*/*b* pigment protein complex detected by circular-dichroism, Biochemistry 27 (1988) 2430–2434.
[113] M. Szabó, B. Lepetit, R. Goss, C. Wihelm, L. Mustárdy, G. Garab, Structurally flexible macro-organization of the pigment–protein complexes of the diatom *Phaeodactylum tricornutum*, Photosynth. Res. 95 (2008) 237–245.
[114] R. Goss, G. Garab, Non-photochemical chlorophyll fluorescence quenching and structural rearrangements induced by low pH in intact cells of *Chlorella fusca* (Chlorophyceae) and *Montaniella squamata* (Prasinophyceae), Photosynth. Res. 67 (2001) 185–197.
[115] A. Jajoo, M. Szabó, O. Zsiros, G. Garab, Low pH induced structural reorganization in thylakoid membranes, Biochim. Biophys. Acta 1817 (2012) 1388–1391.
[116] G. Nagy, L. Kovács, R. Ünnep, O. Zsiros, L. Almásy, L. Rosta, P. Timmins, J. Peters, D. Posselt, G. Garab, Kinetics of structural reorganizations in multilamellar photosynthetic membranes monitored by small-angle neutron scattering, Eur. Phys. J. E 36 (2013), http://dx.doi.org/10.1140/epje/i2013-13069-0.
[117] M. Szabó, Macro-organization and structural flexibility of the photosynthetic pigment system in diatoms, Ph.D. ThesisSzeged University, Hungary, 2011.
[118] W.I. Gruszecki, W. Grudzinski, M. Matula, P. Kernen, Z. Krupa, Light-induced excitation quenching and structural transition in light-harvesting complex II, Photosynth. Res. 59 (1999) 175–185.
[119] I. Jánossy, Optical Effects in Liquid Crystals, Kluwer Academic, Dordrecht, The Netherlands, 1991.
[120] R. Remelli, C. Varotto, D. Sandona, R. Croce, R. Bassi, Chlorophyll binding to monomeric light-harvesting complex —a mutation analysis of chromophore-binding residues, J. Biol. Chem. 274 (1999) 33510–33521.
[121] V.I. Novoderezhkin, M.A. Palacios, H. van Amerongen, R. van Grondelle, Excitation dynamics in the LHCII complex of higher plants: modeling based on the 2.72 Angstrom crystal structure, J. Phys. Chem. B 109 (2005) 10493–10504.
[122] V. Gulbinas, R. Karpicz, G. Garab, L. Valkunas, Nonequilibrium heating in LHCII complexes monitored by ultrafast absorbance transients, Biochemistry 45 (2006) 9559–9565.
[123] M. Khatoon, K. Inagawa, P. Pospisil, A. Yamashita, M. Yoshioka, B. Lundin, J. Horie, N. Morita, A. Jajoo, Y. Yamamoto, Y. Yamamoto, Quality control of photosystem II: thylakoid unstacking is necessary to avoid further damage to the D1 protein and to facilitate D1 degradation under light stress in spinach thylakoids, J. Biol. Chem. 284 (2009) 25343–25352.
[124] Y. Yamamoto, R. Aminaka, M. Yoshioka, M. Khatoon, K. Komayama, D. Takenaka, A. Yamashita, N. Nijo, K. Inagawa, N. Morita, T. Sasaki, Y. Yamamoto, Quality control of photosystem II: impact of light and heat stresses, Photosynth. Res. 98 (2008) 589–608.
[125] N. Nakashima, K. Yoshihara, Role of hot molecules formed by internal conversion in UV single-photon and multiphoton chemistry, J. Phys. Chem. 93 (1989) 7763–7771.
[126] D. Löwenich, K. Kleinermanns, V. Karunakaran, S.A. Kovalenko, Transient and stationary spectroscopy of cytochrome *c*: ultrafast internal conversion controls photoreduction, Photochem. Photobiol. 84 (2008) 193–201.
[127] L. Fiedor, H. Scheer, C.N. Hunter, F. Tschirschwitz, B. Voigt, J. Ehlert, E. Nibbering, D. Leupold, T. Elsaesser, Introduction of a 60 fs deactivation channel in the photosynthetic antenna LH1 by Ni-bacteriopheophytin a, Chem. Phys. Lett. 319 (2000) 145–152.
[128] P.H. Lambrev, Y. Miloslavina, I.H.M. van Stokkum, A.D. Stahl, M. Michalik, A. Susz, J. Tworzydło, J. Fiedor, G. Huhn, M.-L. Groot, R. van Grondelle, G. Garab, L. Fiedor, Excitation energy trapping and dissipation by Ni-substituted bacteriochlorophyll *a* in reconstituted LH1 complexes from *Rhodospirillum rubrum*, J. Phys. Chem. B 117 (2013) 11260–11271.
[129] H. Zer, M. Vink, S. Shochat, R.G. Herrmann, B. Andersson, I. Ohad, Light affects the accessibility of the thylakoid light harvesting complex II (LHCII) phosphorylation site to the membrane protein kinase(s), Biochemistry 42 (2003) 728–738.
[130] A. Istokovics, I. Simidjiev, F. Lajkó, G. Garab, Characterization of the light induced reversible changes in the chiral macroorganization of the chromophores in chloroplast thylakoid membranes. Temperature dependence and effect of inhibitors, Photosynth. Res. 54 (1997) 45–53.
[131] A.K. Holm, O.F. Mohammed, M. Rini, E. Mukhtar, E.T.J. Nibbering, H. Fidder, Sequential merocyanine product isomerization following femtosecond UV excitation of a spiropyran, J. Phys. Chem. A 109 (2005) 8962–8968.
[132] K. Stoitchkova, O. Zsiros, T. Jávorfi, T. Páli, A. Andreeva, Z. Gombos, G. Garab, Heat- and light-induced reorganizations in the phycobilisome antenna of *Synechocystis* sp PCC 6803. Thermo-optic effect, Biochim. Biophys. Acta 1767 (2007) 750–756.
[133] E. Tamary, V. Kiss, R. Nevo, Z. Adam, G. Bernat, S. Rexroth, M. Rogner, Z. Reich, Structural and functional alterations of cyanobacterial phycobilisomes induced by high-light stress, Biochim. Biophys. Acta 1817 (2012) 319–327.
[134] M.G. Rakhimberdieva, I.N. Stadnichuk, T.V. Elanskaya, N.V. Karapetyan, Carotenoid-induced quenching of the phycobilisome fluorescence in photosystem II-deficient mutant of *Synechocystis* sp, FEBS Lett. 574 (2004) 85–88.
[135] A. Wilson, G. Ajlani, J.-M. Verbavatz, I. Vass, C.A. Kerfeld, D. Kirilovskya, A soluble carotenoid protein involved in phycobilisome-related energy dissipation in cyanobacteria, Plant Cell 18 (2006) 992–1007.
[136] R.M. Epand, Lipid polymorphism and protein–lipid interactions, Biochim. Biophys. Acta 1376 (1998) 353–368.
[137] K. Kobayashi, T. Narise, K. Sonoike, H. Hashimoto, N. Sato, M. Kondo, M. Nishimura, M. Sato, K. Toyooka, K. Sugimoto, H. Wada, T. Masuda, H. Ohta, Role of galactolipid biosynthesis in coordinated development of photosynthetic complexes and thylakoid membranes during chloroplast biogenesis in *Arabidopsis*, Plant J. 73 (2013) 250–261.
[138] J.L. Harwood, Involvement of chloroplast lipids in the reaction of plants submitted to stress, in: P.-A. Siegenthaler, N. Murata (Eds.), Advances in Photosynthesis. Lipids in Photosynthesis, Kluwer Academic Publishers, Dordrecht, The Netherlands, 1998, pp. 287–302.
[139] P. Dörmann, Galactolipids in plant membranes, eLS, John Wiley & Sons, Ltd., 2001
[140] S. Haferkamp, W. Haase, H. van Amerongen, H. Kirchhoff, Efficient light harvesting by photosystem II requires an optimized protein packing density in grana thylakoids, J. Biol. Chem. 285 (2010) 17020–17028.
[141] H. Kirchhoff, R.M. Sharpe, M. Herbstova, R. Yarbrough, G.E. Edwards, Differential mobility of pigment-protein complexes in granal and agranal thylakoid membranes of c3 and c4 plants, Plant Physiol. 161 (2013) 497–507.
[142] E.V. Brink-van der Laan, J.A. Killian, B. de Kruijff, Nonbilayer lipids affect peripheral and integral membrane proteins via changes in the lateral pressure profile, Biochim. Biophys. Acta 1666 (2004) 275–288.
[143] H. Haranczyk, K. Strzalka, W. Dietrich, J.S. Blicharski, ^{31}P-NMR observation of the temperature and glycerol induced non-lamellar phase-formation in wheat thylakoid membranes, J. Biol. Phys. 21 (1995) 125–139.
[144] S.B. Krumova, C. Dijkema, P. de Waard, H. Van As, G. Garab, H. van Amerongen, Phase behavior of phosphatidylglycerol in spinach thylakoid membranes as revealed by ^{31}P NMR, Biochim. Biophys. Acta 1778 (2008) 997–1003.
[145] S.B. Krumova, R.B.M. Koehorst, A. Bota, T. Pali, A. van Hoek, G. Garab, H. van Amerongen, Temperature dependence of the lipid packing in thylakoid membranes studied by time- and spectrally resolved fluorescence of merocyanine, Biochim. Biophys. Acta 1778 (2008) 2823–2833.
[146] S.B. Krumova, S.P. Laptenok, L. Kovács, T. Toth, A. van Hoek, G. Garab, H. van Amerongen, Digalactosyl-diacylglycerol-deficiency lowers the thermal stability of thylakoid membranes, Photosynth. Res. 105 (2010) 229–242.
[147] H. Aronsson, M.A. Schottler, A.A. Kelly, C. Sundqvist, P. Dormann, S. Karim, P. Jarvi, Monogalactosyldiacylglycerol deficiency in *Arabidopsis* affects pigment composition in the prolamellar body and impairs thylakoid membrane energization and photoprotection in leaves, Plant Physiol. 148 (2008) 580–592.
[148] W. Wu, W. Ping, H. Wu, M. Li, D. Gu, Y. Xu, Monogalactosyldiacylglycerol deficiency in tobacco inhibits the cytochrome b6f-mediated intersystem electron transport process and affects the photostability of the photosystem II apparatus, Biochim. Biophys. Acta 1827 (2013) 709–722.
[149] J. Kapla, B. Stevensson, M. Dahlberg, A. Maliniak, Molecular dynamics simulations of membranes composed of glycolipids and phospholipids, J. Phys. Chem. B 11 (2012) 244–252.