The molecular architecture, macro-organization and functions of the major light-harvesting protein complexes of green plants – as revealed by circular dichroism spectroscopy techniques

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Light-harvesting complex II (LHCII), the major antenna pigment-protein complex of plants and green algae, is the most abundant membrane protein on Earth. The primary function of LHCII is to capture sunlight and transfer the excitation energy to the photochemical reaction centers – with up to nearly 100% quantum efficiency. As a major structure protein, which readily assembles in highly organized extended arrays, LHCII plays an important role in the stabilization of the ultrastructure of thylakoid membranes and in their reorganizations. In addition, these proteins play key roles in important regulatory mechanisms: in excess light, via controlled dissipation governed by low lumenal pH, they are capable of transiently down-regulating their light-harvesting function; via reversible phosphorylation they participate in regulation of the distribution of excitation energy between the two photosystems. Although the crystal structure of LHCII is known at near-atomic resolution, it does not account for these diverse functions, which are also documented by ultrafast time-resolved spectroscopy studies. Hence, it is essential to have detailed and accurate information about the molecular architectures and macroorganizations of these protein complexes in vivo and in vitro in their different functional states. In this, UV-VIS circular dichroism (CD) spectroscopy is an indispensable tool.

CD in the far UV is capable to provide information on the secondary structure of proteins in different physicochemical environments. In the visible region, in hierarchically organized systems with different levels of structural complexity, CD signals can be given rise by different physical mechanisms that are superimposed on each other. At the molecular level, CD arises from intrinsic asymmetry or from asymmetric perturbations. In molecular complexes or their small aggregates, excitonic CD can be induced by short-range interactions between pigment dipoles. Chirally organized densely packed macroaggregates, so-called psi-type aggregates (psi, polymer or salt-induced), exhibit large anomalously shaped psi-type CD signals, accompanied with differential scattering, revealing long-range molecular interactions. On macroscopically aligned samples, anisotropic CD (ACD) can be measured, which carries additional information on the structural basis of some bands in the CD spectrum. Magnetic CD (MCD) can also be used to identify the nature of certain bands and also, as an internal probe, to test possible spectral distortions in the presence of light scattering.

Examples will be shown, including recent experiments at the B23 Beamline of the Diamond Light Source, how CD provided information on the molecular architecture and/or macro-organization of the protein complexes and on pigment-pigment interactions in different LHCII samples, detergent-solubilized monomers and trimers and their chiral macro-assemblies, LHCII embedded in liposomes or in their native thylakoid membranes and microcrystals, and how they can be correlated with different functional states of LHCII.