not currently accessible to ^{19}F NMR of F-Trp-labeled enzyme (between residues 59 and 384).

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PHYCOBILISOMES

Macromolecular Structure and Energy Flow Dynamics

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Cyanobacteria and red algae carry out efficient photosynthesis with light over the wavelength range 450 to 650 nm. The absorption of light at these wavelengths is performed by a family of intensely colored, brilliantly fluorescent chromoproteins (biliproteins). These proteins carry covalently attached open-chain tetrapyrrole (bilin) chromophores that vary in chemical structure. The chemical nature of the particular bilins attached to a given biliprotein in large measure determines its absorption spectrum. Within cyanobacterial cells and red algal chloroplasts, the biliproteins are organized into particles of intricate structure, phycobilisomes, which are attached to the outer surface of the thylakoid membrane (1).

The structure of a typical cyanobacterial phycobilisome, that of *Synechocystis* 6701, is shown in Fig. 1. The location of the various components within this structure was determined by analysis of incomplete phycobilisomes from a large number of mutants and from studies of sub-assemblies obtained by partial dissociation of phycobilisomes (1, 2). In isolated phycobilisomes, light absorbed over a wide range of wavelengths is emitted as fluorescence at ~676 nm. In intact cells, little of this emission is observed because of efficient energy-transfer from the terminal energy acceptors to two chlorophyll-*a*–containing photosystem II reaction center complexes, lying beneath the phycobilisome in the thylakoid membrane (3).

The *Synechocystis* 6701 phycobilisome is a particle of $\sim 7.5 \times 10^6$ d that contains ~650 bilins. It is made up of two distinct kinds of substructures; rods consisting of 60 Å thick by 120 Å diameter disks of biliprotein hexamers, and a core consisting of three cylinders. The axes of the core cylinders lie at right angles to the cylindrical axes of the rods. Four rods are attached to the upper core cylinder and one rod is attached to each of the basal core cylinders. Each of the core cylinders is made up of four 30 Å thick by 115 Å diameter "trimeric" allophycocyanin complexes. Polypeptides carrying terminal acceptor bilins, α^{APB} and L^{99}_{CM} are each contained within complexes in the basal core cylinders, as illustrated in Fig. 1.

We have examined the dynamics of energy flow within complete phycobilisomes, as well as in partial structures produced by two mutants. *Synechocystis* 6701 strain CM25 produces particles lacking the two terminal phycoerythrin complexes of the rod substructures; the phycobilisome is otherwise unchanged. *Synechocystis* 6701 strain UV16 produces wild-type cores containing L^{27}_{RC} linker polypeptides, but lacking all other rod components (4). Picosecond spectroscopic studies have shown that disk-to-disk transfer (~24 ps) represents the rate-limiting step in energy flow in the rods (5). Energy transfer within the core is <11 ps.[1]

We have also found that within isolated biliproteins, intradisk energy transfer is very rapid, occurring in <8 ps (5). Consequently, from the standpoint of energy flow dynamics, each building block of the phycobilisome can be regarded as equivalent to a single chromophore. Because these building blocks are arranged in the order of decreasing excitation energies from the periphery of the structure to the terminal acceptors, the energy flow is highly directional towards the core. The structure of the phycobilisome also minimizes the possibilities for the random walk of excitation among like chromophores. Thus, only a small number of energy transfer steps of any significant length relative to the ~2 ns fluorescence lifetime of a biliprotein is

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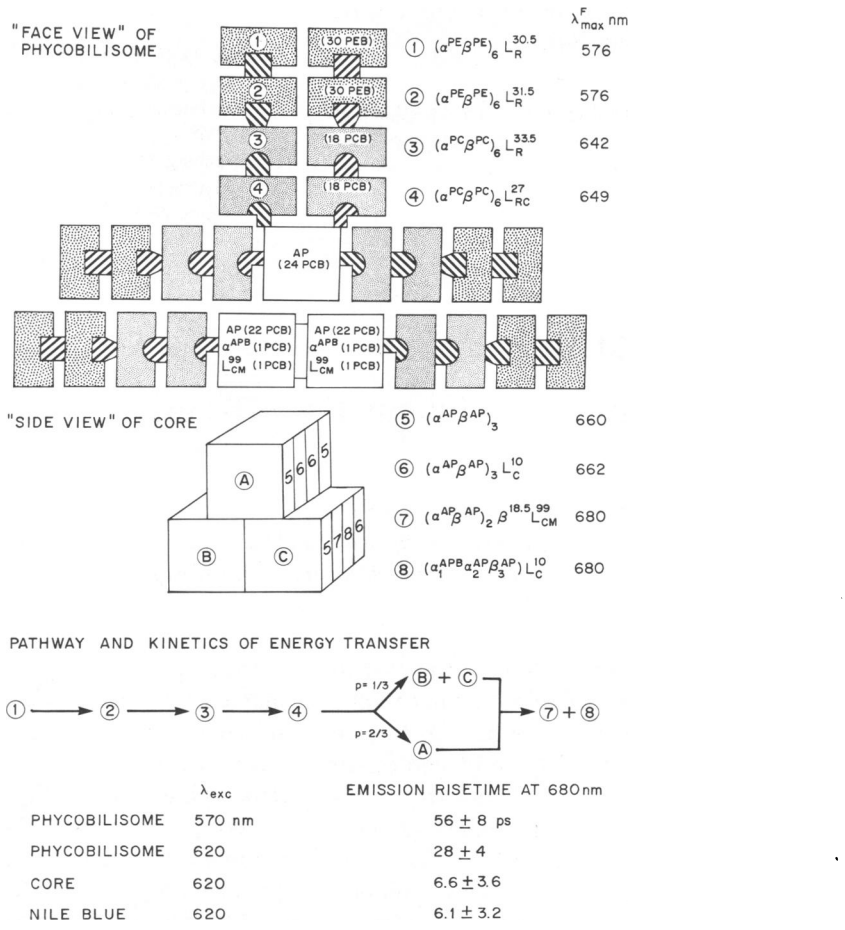

FIGURE 1 Schematic representation of the *Synechocystis* 6701 phycobilisome. A rod is made up of four hexameric biliprotein complexes, each of which is attached through its specific linker polypeptide to the component adjacent to it in the phycobilisome. The manner in which the core cylinders are held together is not known. The abbreviations AP, PC, and PE are used for the biliproteins allophycocyanin, phycocyanin, and phycoerythrin, and α^{AP}, β^{AP}, etc., for the α and β subunits of these proteins. Linker polypeptides are abbreviated *L*, with a superscript denoting the apparent size $\times 10^3$ d, and a subscript that specifies the location of the polypeptide; *R*, rod substructure; *RC*, rod-core junction; *C*, core; *CM*, core membrane junction. For other details, see reference 1. The number of bilins present in each domain of the structure is indicated in the upper diagram. Abbreviations used are PEB, phycoerythrobilin; PCB, phycocyanobilin; λ^{F}_{max}, fluorescence emission maximum. In the data on the kinetics of energy transfer, the nonzero rise time of the emission from the nile blue solution is indicative of the maximum temporal resolution available with the apparatus used for the kinetic studies. *p* = probability.

required to convey excitation from any point within the phycobilisome to the terminal acceptors (see Fig. 1).

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