Excitons in Light Harvesting Complexes: Quantum Delocalization, Hydrogen Bonding, and Design Principles

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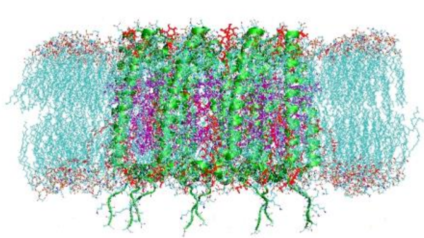
Natural organisms such as photosynthetic bacteria, algae, and plants employ complex molecular machinery to convert solar energy into biochemical fuel. An important common feature shared by most of these photosynthetic organisms is that they capture photons in the form of excitons typically delocalized over a few to tens of pigment molecules embedded in protein environments of light harvesting complexes (LHCs). Delocalized excitons created in such LHCs remain well protected despite being swayed by environmental fluctuations, and are delivered successfully to their destinations over hundred nanometer scale distances in about hundred picosecond time scales. Despite decades of research, key design principles enabling their superb light harvesting capability are not yet clearly understood at present. A representative example for this status of knowledge is the photosynthetic unit (PSU) of purple bacteria, which consists of only two types of antenna complexes called light harvesting 1 (LH1) and light harvesting 2 (LH2) with known crystal structures. Of these, LH2 complexes serve as the major initiator and carrier of the excitation energy. Based on comprehensive computational modeling of the LH2 complex and their synthetic analogues/natural mutants, we have elucidated new molecular level design principles for their robust and efficient exciton migration mechanisms. Analyses of computational results have demonstrated hidden effects of hydrogen bonding on controlling the disorder and on optimal sizes of LH2. These results provide new insights into how natural systems control negative effects of disorder through interplay of structural factors and quantum mechanical delocalization.



LH2 of purple bacteria in membrane