ABSTRACT: ROLE OF SYSTEM-BATH INTERACTION IN PHOTOSYNTHETIC EXCITATION ENERGY TRANSFER

In this thesis we investigate the influence of fast and slow molecular motion on the excitation energy transfer in photosynthetic light-harvesting complexes. Developing a new theoretical description for intramolecular high-frequency vibrational modes, we find that their resonance with energy gaps between the photosynthetic pigments can speed up the energy transfer. Employing single-molecule spectroscopy, we observe how slow changes of protein conformation can completely switch the state of a plant light-harvesting complex LHCII. Finally, we develop a new experimental technique of two-pulse ultrafast single-molecule spectroscopy. With its help we can observe how slow motion of a bacterial LH2 antenna protein influences the ultrafast energy relaxation within the complex. By constructing a unified model for ultrafast bulk and single-molecule experiments, we incorporate the fast and slow timescale of molecular motion into one picture of photosynthetic light harvesting.