Title:	Multi-Stimuli Responsive Biomimetic Light-Harvesting Antennae and Macrocycles Towards Multifunctional
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Abstract:	Abstract The first and foremost event in natural photosynthesis is sunlight harvesting by well-organized multiple chromophores usually called light-harvesting (LH) antenna complexes, followed by ultrafast Förster resonance energy transfer (FRET) and electron transfer processes. To mimic the natural LH systems, artificial multichromophoric systems have been developed over the decades through covalent as well as supramolecular strategies. To develop highly efficient covalent light-harvesting antennae is very challenging since it encompasses synthetic challenges as well as in-depth understanding and characterization of the photophysical phenomena. Furthermore, by virtue of their multiple absorption/emission bands tunable by multiple stimuli (temperature, pH, redox potentials, metal ions), artificial LH small molecular antennae can serve as stimuli-responsive smart materials, a barely explored area and the central theme of this thesis. This work intends to address the design and synthesis of artificial LH systems that not only mimic the efficient and fast FRET and electron transfer of natural systems, but have also exhibited new and emergent multi-stimuli responsive photophysical properties. In the third chapter, molecular triads based on perylenediimide (PDI) and aza- BODIPY have been synthesized and characterized for efficient FRET as well as for appreciable electron mobility, multi-stimuli responsive photophysical behaviour towards temperature (ratiometric temperature sensors) and metal-ions (FRET turn-off metal sensors). [1] To extend the absorption of LH antennae over a broad spectral window, red-green-blue (RGB) antenna was synthesized in the fourth chapter, which showed cascade FRET from naphthalimide to PDI to aza-BODIPY. It exhibited multi-stimuli responsive selective spectral band tuning with temperature, pH, redox stimuli and applications were demonstrated as multifunctional sensors as well as tunable fluorescent inks. [2] In the fifth chapter, inspired by the cyclic arrangement of bacteriochlorophylls in the natural LH system of purple bacteria, two metal-free macrocycles [1+1] and [2+2] were synthesized composed of covalently connected PDI and aza-BODIPY and characterized for efficient FRET from PDI to aza-BODIPY, followed by the investigation of their multi-stimuli responsive behaviour and FRET-enhanced photocatalytic responses towards photooxidation reaction. [3] In the sixth chapter, dyads composed of PDI and styryl- BODIPY were synthesized, where the PDI was attached through one of the □-positions of mono- or di-styryl-BODIPYs. These dyads showed ultrafast charge transfer from styryl- BODIPYs to PDI as revealed by steady state and transient absorption studies and analysis. [4] The talk will conclude with summary and future perspectives of these developed systems in the application areas of optoelectronics as well as FRET photocatalysis.
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