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Control of functional molecules in photosynthesis and photomedicine

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Coherent control is rapidly becoming a spectroscopic tool that complements other techniques in exploring excited state dynamics and reaction mechanisms. Using a closed-loop optimization scheme, little prior knowledge of the system is required. As such, even complex systems that elude quantum mechanical description can be studied. Here, we explore functional biomolecules in (artificial) photosynthesis and photomedicine, whose efficiencies are limited by competing loss channels in the activated states. The goal is to maximize (or minimize) yield in the functional pathway, and in doing so to better understand the factors that underlie optimal performance.

Artificial photosynthetic complexes aim to mimic the salient features of their natural counterparts. We studied a donor-acceptor dyad molecule inspired by the LH2 light harvesting complex of photosynthetic bacteria. Detailed transient absorption measurements coupled with global analysis of the spectral evolution map out the pathways and efficiencies of energy flow. We find that the functional pathway of energy transfer is significantly impeded by a competing ultrafast loss channel. Closed-loop optimization experiments indicate that both pathways can be manipulated, such that the efficiency of energy transfer may be enhanced or diminished. Open loop control is further employed to extract insight to the mechanism.

Porphyrin derivatives have been successfully applied as photosensitizer molecules in photodynamic therapy (PDT). These molecules bind selectively to tumor cells and upon optical excitation facilitate the production of singlet oxygen. The efficiency of PDT depends on the competition between intramolecular deactivation channels. Here we present results on a prototype sensitizer, zinc phthalocyanine. Combining ultrafast spectroscopy and coherent control techniques, we target specific reaction pathways with the goal of enhancing function and minimizing loss.