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Increased exciton annihilation in incrementally aggregated photosynthetic antenna complexes from plants

The automatic photoprotective mechanisms of photosynthetic antenna complexes that initiate light-harvesting have been a subject of great interest for potentially improved solar energy technology, enhanced crop efficiencies, and biosensing. Light-Harvesting Complex II (LHCII) is the main pigment-protein antenna in green plants and exhibits the remarkable capability to switch between a light-harvesting and a photoprotective state when exposed to fluctuating sunlight intensities. The in-vivo conditions that activate this switch can be mimicked by aggregation of LHCII. Despite more than three decades of research, the molecular mechanism responsible for the strong energy quenching in these aggregates is still unknown. We investigated LHCII aggregation in a stepwise manner and performed fluorescence correlation spectroscopy (FCS) along with time-correlated single-photon counting (TCSPC) on a home-built experimental setup to correlate the aggregate composition with their excited-state lifetimes. We discovered a non-linear relationship between the steady-state intensities and average lifetimes, which is explained well by increased annihilation of diffusing singlet excitons due to an accumulation of triplet excitons in aggregates. An approximated model of this singlet-triplet annihilation showed excellent correspondence with the experimental data. These results demonstrate the importance of distinguishing non-linear exciton annihilation from exciton quenching in photoprotective studies of plants.

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