A Step Towards A Comprehensive Steady-State Picture Of Photosynthetic Solar Energy Conversion

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Abstract. The interpretation of oscillating experimental signals observed in ultrafast nonlinear spectroscopies [1] has been motivating vigorous interest in quantum effects in photoinduced biological processes. However, electronic dynamics triggered by natural light, which is stationary and incoherent, is generally substantially different from the one observed in pulsed laser experiments. It has been suggested that the physically correct picture of photosynthetic excitation energy transfer (EET) should be in terms of a steady state [2], which is formed when the photosynthetic antenna is continuously photoexcited and continuously delivers the excitation energy to the reaction center, in which charge separation takes place.

We study EET triggered by a low-intensity photoexcitation of an initially unexcited molecular aggregate, which interacts with its environment and is coupled to the reaction center. We treat the aggregate–environment coupling in a numerically exact manner and extend previous theoretical treatments [3, 4] by formulating the hierarchy of equations of motion (HEOM) which explicitly takes into account the photoexcitation process. We investigate the properties of the steady state arising when the aggregate is subjected to a continuous-wave excitation, while the charge separation from the reaction center occurs at a constant rate. The developed theoretical formalism enables us to approach questions ranging from the influence of the short-time dynamics (which is accessible in pulsed laser experiments) on the steady state to the relevance of steady-state coherences for the EET process.

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