Exciton-Condensate-like Amplification of Exciton Transfer in Light Harvesting

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Bose-Einstein condensation of excitons, in which excitons condense into a single coherent quantum state, known as an exciton condensate, enables frictionless energy transfer, but typically occurs under extreme conditions in highly ordered materials, such as graphene double layers. In contrast, photosynthetic light-harvesting complexes demonstrate extremely efficient transfer of energy in disordered systems under ambient conditions. Here, we establish a link between the two phenomena by investigating the potential for an exciton-condensate-like mechanism in energy transport in photosynthetic light harvesting. Using a model of the Fenna-Matthews-Olson (FMO) complex and accounting for intrachromophore electron correlation explicitly through the addition of multiple sites to the individual chromophores, we observe amplification of the exciton population in the particle-hole reduced density matrix through an exciton-condensate-like mechanism. The nature of the exciton-condensate-like mechanism is influenced by the dynamics of exciton transfer and the coupling strength, as well as the initial excitation conditions and structure of the light harvesting complex. In FMO, tuning intrachromophore coupling can tune the exciton transfer with the possibility of enhancement in transfer efficiency. The research provides fundamental connections between exciton condensation and exciton transport in light-harvesting complexes with potential applications for harnessing the exciton-condensate-like mechanism to enhance energy transfer in synthetic systems and create new materials capable of highly efficient energy transfer. [PRX Energy, Vol. 2, Iss. 2, 023002 (2023)].