

Abstract Submitted
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The coherence lifetime-borrowing effect in vibronically coupled molecular aggregates under non-perturbative system-environment interactions.¹ SHU-HAO YEH, University of Chicago; Qatar Environment and Energy Research Institute, GREGORY S. ENGEL, University of Chicago, SABRE KAIS, Purdue University; Qatar Environment and Energy Research Institute — Recently it has been suggested that the long-lived coherences in some photosynthetic pigment-protein systems, such as the Fenna-Matthews-Olson complex, could be attributed to the mixing of the pigments' electronic and vibrational degrees of freedom. In order to verify whether this is the case and to understand its underlying mechanism, a theoretical model capable of including both the electronic excitations and intramolecular vibrational modes of the pigments is necessary. Our model simultaneously considers the electronic and vibrational degrees of freedom, treating the system-environment interactions non-perturbatively by implementing the hierarchical equations of motion approach. Here we report the simulated two-dimensional electronic spectra of vibronically coupled molecular dimers to demonstrate how the electronic coherence lifetimes can be extended by borrowing the lifetime from the vibrational coherences.

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