

Abstract Submitted
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Electronic energy transfer: vibrational control and nonlinear wavepacket interferometry OLEG V. PREZHDO, University of Washington, JEFFREY A. CINA, University of Oregon, DMITRI S. KILIN, University of Washington — The time-development of photoexcitations in molecular aggregates exhibits specific dynamics of electronic states and vibrational wavefunction. We discuss the dynamical formation of entanglement between electronic and vibrational degrees of freedom in molecular aggregates with theory of electronic energy transfer and the method of vibronic 2D wavepackets [J. Chem. Phys. 118, 46 (2003); quant-ph/0412219]. The vibronic dynamics is also described by applying Jaynes-Cummings model to the electronic energy transfer [J. Chem. Phys. 120, 11209 (2004); math-ph/0403023]. Following the ultrafast excitation of donor [J. Phys. Chem 99, 2568 (1995); chem-ph/9411004] the population of acceptor rises by small portions per each vibrational period, oscillates forward and back between donor and acceptor with later damping and partial revivals of this oscillation. The transfer rate gets larger as donor wavepacket approaches the acceptor equilibrium configuration, which is possible at specific energy differences of donor and acceptor and at maximal amount of the vibrational motion along the line that links donor and acceptor equilibria positions. The four-pulse phase-locked nonlinear wavepacket 2D interferograms reflect the shape of the relevant 2D vibronic wavepackets and have maxima at longer delay between excitation pulses for dimers with equal donor-acceptor energy difference compare to dimers with activationless energy configuration [Cina, Fleming, J. Phys. Chem. A. 108, 11196 (2004)].

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