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Energy transfer: vibrational control and nonlinear wavepacket interferometry D. S. KILIN, O. V. PREZHDO, University of Washington, J. A. CINA, University of Oregon — The time-development of photoexcitations and dynamical formation of entanglement between electronic and vibrational degrees of freedom in molecular aggregates is analyzed by applying method of vibronic 2D wavepackets [J. Chem. Phys. 118, 46 (2003); quant- ph/0412219, theory of electronic energy transfer, and Jaynes-Cummings model [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 each vibrational period, oscillates force 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 [J. Phys. Chem. A. 108, 11196 (2004)].

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