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Monitoring the Coherent Vibrational Control of Electronic Excitation Transfer Using Ultrafast Pump-Probe Polarization Spectroscopy JASON BIGGS, JEFFREY CINA, University of Oregon — The interplay between nuclear and electronic degrees of freedom in molecular energy-transfer complexes is a subject of current interest. We have proposed a method to use coherent nuclear motion to control the transfer of electronic excitation energy between donor and acceptor moieties in electronically coupled dimers. The underlying electronic and nuclear motion at the level of quantum mechanical amplitudes can be observed using nonlinear wave-packet interferometry(nl-WPI), a form of fluorescence-detected multidimensional electronic spectroscopy. In our control scheme, coherent nuclear motion is induced in the acceptor chromophore prior to direct electronic excitation of the donor. This nuclear motion affects the instantaneous resonance conditions between donor and acceptor moieties and thus affects subsequent energy transfer dynamics. We have developed the framework to simulate four-pulse nl-WPI experiments, and the pump-probe limit thereof, on energy-transfer systems after interaction with a control pulse that induces nuclear motion. We present simulations in the pump-probe limit from model energy-transfer systems subjected to prior impulsive vibrational excitation, and show how pulse polarization can be used to infer electronic dynamics from isotropically oriented dimers.

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