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Coherent Multidimensional Spectroscopy of Photosynthetic Complexes: Manipulating Quantum Pathways by Optical Pulse Sequences

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The response of chromophore aggregates to sequences of femtosecond laser pulses is simulated using the nonlinear exciton equations. The nonlinear response can be interpreted in terms of the scattering of elementary excitations, quasiparticles, rather than as transitions among eigenstates. Applications are made to the Fenna-Matthews-Olson (FMO) and the PSI light harvesting complexes. Some fundamental symmetries of multidimensional optical signals are used to design techniques that can selectively resolve coherent quantum dynamics and incoherent energy dissipation. Simulations show damped oscillations of cross peaks corresponding to evolution of coherences, without interference from incoherent population relaxation. Energy transfer pathways are seen through the redistribution of crosspeak amplitudes. Resolution is enhanced by employing specific pulse polarization configurations to generate chirality-induced signals. New pulse sequences are designed to generate signals that are induced by correlations among elementary excitations. Specific phase-matching directions can target the correlated dynamics of double excitations. Cross peaks in 2D correlation plots are interpreted in terms of quasiparticle scattering and shown to reveal the double-exciton wavefunction, projected into products of single-excitons. Uncorrelated double-exciton states do not show up in the spectra due to quantum interference among pathways. The proposed techniques amplify cooperative dynamical features and reveal information on the robustness of quantum states to fluctuating environments. In collaboration with Darius Abramavicius and Dmitri Voronine, University of California, Irvine, CA 92697.

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