Composite-Boson Representation of Coherent Nonlinear Optical Signals of Multiexcitons In Photosynthetic Complexes.

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The elementary optical excitations of many-Fermion systems such as molecules and semiconductors are particles made of an even number of Fermions and are known as composite Bosons (co-bosons). Here we show that the effects of many-body interactions in the nonlinear optical response to any order can be represented as a sequence of two-body interactions: the Pauli exclusion of Fermions and direct Coulomb scattering. Nonlinear optical experiments can measure the formation and evolution of interacting multi-excitons via the scattering matrix defined by a generalized n-exciton Bethe-Salpeter equation. The simulation protocol only requires the numerical diagonalization of the single exciton manifold. Coboson algebra is then used to calculate multidimensional optical signals to desired order. All many body effects are recast in terms of two tetradic two body-interactions: direct Coulomb scattering and Pauli exchange. A coherent fifth order Triple quantum coherence optical signal that directly probes three exciton states and their projections on lower exciton states is predicted.

Energy- transfer and charge-separation pathways in the reaction center of photosynthetic bacteria may be revealed by coherent two-dimensional optical spectroscopy. The excited state dynamics and relaxation of electrons and holes are simulated using a two-band tight-binding model. The dissipative exciton and charge carrier motions are calculated using a transport theory, which includes a strong coupling to a harmonic bath

1. “Composite-Boson Representation of Coherent Nonlinear Optical Signals of Multiexcitons”, O. Roslyak, K. Bennett, S. Mukamel, Phys.Rev. Lett (Submitted, 2011).

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