Abstract Submitted
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Coherent vs. dissipative nonequilibrium dynamics in spectroscopy of molecular aggregates[1] DARIUS ABRAMAVICIUS, ANDRIUS GELZINIS, VYTAUTAS BUTKUS, LEONAS VALKUNAS, Physics Faculty, Vilnius University, Sauletekio al. 9, 10222 Vilnius, Lithuania — Molecular aggregates embedded in a protein environment are the core elements in photosynthetic antennae units. Photoexcitations in these systems experience multistep relaxation, which could be traced using various time-resolved spectroscopy techniques. Initiated coherent processes turn into dissipative. Understanding of these processes is still a major theoretical task. We study theoretically spectroscopic properties of simple molecular aggregates coupled to a bath, which contains main ingredients of protein environemnt: high-energy vibrations, long-range correlations, and smooth spectrum of frequencies. At short times after the optical excitation high-energy coherent vibrational resonanses can be observed in two-dimensional rephasing spectroscopy. Their beats overlap with electronic quantum coherences, responsible for the quantum transport. We show the way to discriminate between them. At the long times we find that the conventional excitonic picture of eigenstates is valid only in the Markovian regime. In the non-Markovian regime the exciton concept breaks down and renormalized system parameters must be introduced: effective intermolecular coupling, widely used in polaron theories, can be used to account for the effects of the bath.

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