

Abstract Submitted
for the MAR05 Meeting of
The American Physical Society

Photoinduced vibrational coherence transfer in molecular aggregates D. S. KILIN, O. V. PREZHDO, YU. V. PEREVERZEV, V. NAGARAJAN, W. W. PARSON, University of Washington — At short times faster than the time of dephasing a strong photoinduced excitation in an electron-phonon molecular structure induces evolution of the vibrational subsystem that depends on the electronic evolution. The equilibrium position and oscillation frequency of the mean nuclear coordinate depend on which electronic state keeps the major part of the population. This effect is described theoretically at a simple analytic level by applying the quantized Hamiltonian dynamics (QHD) formalism [J. Chem. Phys. **120** 11209 (2004)] to the electronic and vibrational degrees of freedom of a model molecular aggregate, as motivated by recent experimental data in a bacteriochlorophyll aggregate [J. Phys. Chem. B **103** 2297 (1999)]. The ultrafast pump-probe signal is considered. The vibronic wavepacket driven by electronic energy transfer sequentially visits different excited states. The probe signal oscillates with the nuclear configuration at the frequency that is modulated depending on the curvature of the current potential energy surface. This modulation of the frequency of the probe signal known as coherence transfer is described within simple analytic and numerical models by the QHD method that can be easily extended to many degrees of freedom.

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Date submitted: 01 Dec 2004

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