COHERENT EXCITATION PHENOMENA IN TIME-RESOLVED EXPERIMENTS

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The influence of coherent phenomena on femtosecond pump-probe experiments in molecular systems has been investigated. The signature of Coherent Population Return (CPR) has been observed, and satisfactorily described by means of a coherently excited two-state model. This analysis has been extended to a more general situation where N two-level subsystems interact independently with the radiation. The obtained results permit us to explain why for such complex systems the incoherent treatment provides an accurate description of the population dynamics. Furthermore, the distribution of states inside and outside the excitation laser bandwidth, rather than the state density as it is accepted, is found to be the key parameter for determining the applicability of the incoherent approach.