MAR07-2006-003722

Abstract for an Invited Paper
for the MAR07 Meeting of
the American Physical Society

Coherent 2D Spectroscopy and Control of Molecular Complexes

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Coherent two-dimensional femtosecond spectroscopy is used to investigate electronic couplings within molecular complexes. Third-order optical response functions are measured in a non-collinear three-pulse photon echo geometry with heterodyne signal detection. In combination with suitable simulations this allows recovering the delocalization of excited-state wavefunctions, their coupling, and the corresponding energy transport pathways, with nanometer spatial and femtosecond temporal resolution. Examples of multichromophoric systems are the FMO and the LH3 light-harvesting complexes from green sulfur bacteria and purple bacteria, respectively, for which energy transfer processes have been determined. Additional challenges arise if one is interested in the spectroscopy of photochemical rather than photophysical processes in molecular complexes: The product yields attained by a single femtosecond laser pulse are often very small, and hence time-dependent signals are hard to measure with good signal-to-noise ratio. In the context of coherent control, this implies that bond-breaking photochemistry in liquids is still difficult despite the many successes of optimal control in gas-phase photodissociation. In a novel accumulative scheme, macroscopic amounts of stable photoproducts are generated in an optimal fashion and with high product detection sensitivity. In connection with time-resolved spectroscopy, the accumulative scheme furthermore provides kinetic information on the pathways of low-efficiency chemical reaction channels. This was applied to investigate the photoconversion of green fluorescent protein.