

Developments in Molecular Excitation with Natural Incoherent Light

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2D photon echo studies on light harvesting systems have generated considerable interest and controversy regarding the possible role of quantum coherence effects in biological systems.

As we have previously shown, such studies rely on the response of molecular systems to pulsed laser excitation, which is significantly different than the response to natural incoherent light. Significantly, the latter produces mixed stationary states, devoid of time dependent coherences.

After briefly summarizing these results we describe new developments in this area, including the significance of various decoherence time scales for reaching mixed states in natural incoherent light, the role of doorway states in the molecular response, and the insight afforded into the quantum yield by the character of system energy eigenstates. Examples will be chosen from isomerization in retinal, radiationless transitions in pyrazine, and Rydberg atoms interacting with the cosmic microwave background.