Thesis Abstract

The present thesis uses a multidimensional technique, *Spectrally Resolved Photon Echo Spectroscopy* (SRPE) to study the vibrational and electronic dynamics in complex molecular systems. The thesis also details out the theory and construction of a three pulse photon echo experimental setup. This also provides an overview and background of SRPE spectroscopy and discusses our femtosecond SRPE experimental system. The entire experimental setup, including the ultrafast laser systems is also describe. The functioning of home built femtosecond oscillator and Multipass amplifier (ODIN) has been described in details. The ODIN is pumped by a Nd:YAG laser (Evolution-15, Coherent Inc.) that generates nanosecond pulses at 532 nm wavelength and has a repetition rate of 1 KHz. The seed pulses for the ODIN comes from the home-built Ti:Sapphire oscillator (KM–Labs kit). The optical parametric amplifier, TOPAS (Quantronix Inc.) generates tunable laser pulses that cover the whole visible spectral region. We also describe the second laser system, which consists of a tunable repetition rate commercial Regenerative Amplifier (Spitfire Pro, Spectra Physics). This amplifier is seeded by femtosecond pulses from a 82 MHz Ti:Sapphire oscillator (Mai Tai, Spectra Physics) and amplified by a Nd:YLF Pump laser system (Empower, Spectra Physics). The technique used to measure the pulses coming out of these lasers is also described. Spectrally Resolved Photon Echo spectroscopy (SRPE) of Astaxanthin in two different solvents by using amplified laser system is discussed. This technique is also used to study the Rhodamine 6G (Rh6G) dye at two different excitation wavelengths. The study of Zinc substitution effect on the molecular dynamics of Protoporphyrin-IX in dichloromethane solvent is also presented by SRPE technique. This technique is further used in off-resonance condition for the first time to study the coherence and population dynamics of CdSe quantum-dots. The comparative study of the molecular dynamics of two xanthene dye derivatives (Rhodamine 6G and Fluorescein 548) is presented by using the SRPE spectroscopy.