Abstract

This thesis mainly explores the photo-physical properties of molecular systems under different molecular environments using time-resolved spectroscopy. This thesis starts with a brief discussion on the motivation behind the present work. The introductory chapter (**Chapter 1**) includes a brief discussion on the photo-physical properties and the dynamics of molecular systems.

Chapter 2 describes in details, the laser systems that are used to perform the experiments. It discusses laser pulse characterization using home built intensity autocorrelator and all the experimental techniques used in this thesis, namely, *Time Correlated Single Photon Counting* (TCSPC), *Femtosecond Fluorescence Upconversion* and *Degenerate Pump-probe Spectroscopy*. In **Chapter 3,** Polarization induced control of multiple fluorescence from a molecule is reported. The popular indocyanine dye, IR125 in dimethyl sulfoxide (DMSO) produces a conventional single photon fluorescence in the NIR region, (650nm - 900 nm) when excitation at 800 nm. But, when we change the excitation wavelength at 527 nm or, even bluer laser pulses, instead of 800 nm, it shows a novel dual fluorescence. The newly found fluorescence in the visible region is coming from S_2 state.In **Chapter 4**. We have used two tricarbocyanine dyes, IR144 and IR140 and studied the ultrafast dynamics in four primary alcohol.

In **Chapter 5** observations of coherent oscillations in solution due to microheterogeneous environment is presented. We observed the coherent oscillations in the ground state dynamics of the dye in binary mixtures. Coherent oscillations mainly depend on the compositions of the binary mixtures and the maximum number of coherent oscillations has been observed in 20% MeOH. **Chapter 6** deals with dynamics of IR775 across an immiscible liquid-liquid interface(DMSO and diethyl-ether). In **Chapter 7**, using *linear time of flight mass spectrometry,* we have shown the chemical transformation of dicyclopentadiene into cyclopentadiene in a supersonic molecular beam chamber.