Name	:	Darakhshan Qaiser
Supervisor	:	Prof. Zahid Husain Khan
Co-Supervisors	:	Dr. Mohd. Shahid Khan & Dr. R.D. Singh
Institute	:	Department of Physics, Jamia Millia Islamia, New Delhi-110025.
Title of Ph. D. Thesis	:	Study of Optical Gain and Relaxation Mechanism of Fullerenes in Solution.

Abstract

The most important single feature behind selecting this topic has been the fact that all the fullerenes have very good absorption near the nitrogen laser frequency. Since nitrogen laser is quite common for pumping a dye laser system and many dyes are popular as laser materials, this has tempted us to study the possibility of using fullerenes as laser material. In the present work we have studied the optical properties of fullerene C_{60} and fullerene C_{70} in pure form and in mixture. The thesis is based on the following studies:

- Effect of fullerene C_{60} and fullerene C_{70} concentration on the fluorescence spectra.
- Forster's Resonance Energy Transfer (FRET) between fullerene C_{60} and Coumarin C440.
- FRET study of fullerene C_{70}-quinizarine and its FRET study.
- Time Resolved Spectroscopy and Optical Gain measurements of fullerene C_{60} and fullerene C_{70}.
- Study of band gap, X-ray diffraction and SEM for fullerene C_{60} thin films

Effect of Fullerene C_{60} and Fullerene C_{70} concentration on the fluorescence spectra

The optical absorption spectra of fullerenes C_{60} and fullerene C_{70} of different concentrations, varying from 5 µM to 50 µM, in the wavelength range of 250 nm - 450 nm were measured. It is noticed that the absorption characteristics do not change with concentration and the absorbance follows the Beer-Lambert law. The spectrum is red shifted with increase in the concentration of fullerene C_{60} in the solution, which is due to aggregate formation and is reversible with dilution.

Forster's Resonance Energy Transfer (FRET) between Fullerene C_{60} and Coumarin C440.

This study is focused on the measurement of FRET radius (Forster's Distance), Stern Volmer Characteristics and FRET Efficiency. We find that in fullerene C_{60}- Coumarin pair the fluorescence energy transfer from Coumarin C440 to fullerene C_{60} takes place. The Stern Volmer plot of the pair indicates that for both types of quenching, i.e. static as well as dynamic, the Forster's radius (R_0) at 50% energy transfer efficiency is found to be 34.1 Å. Since the

Foster's radius lies in the range 20 Å to 90 Å, therefore the present pair can be used as spectroscopic ruler for biomedical applications.

FRET study of Fullerene C_{70}-Quinizarine

In this study we measured the FRET distance in fullerene C_{70}-quinizarine pair and compared the results obtained by steady state fluorescence and laser induced fluorescence (LIF) techniques using 532 nm excitation. From the spectra measured by steady state method, it is found that the fluorescence energy transfer from quinizarine to fullerene C_{70} takes place. Also the Stern Volmer plot is linear throughout the concentration of the acceptor which indicates collisional quenching process. In the steady state fluorescence technique, the Forster's radius for the pair is found to be 25.4 Å, whereas in the LIF technique it is 72.0 Å. Since the Forster's distance calculated by both the methods is within the range 20 Å - 90 Å, it demonstrates that both the methods for the pair are suitable as spectroscopic ruler.

Time Resolved Spectroscopy and Optical Gain measurements of Fullerene C_{60} and Fullerene C_{70}.

In this study we measured the fluorescence decay of both the fullerenes in pure form as well as in mixture. It is found that the fluorescence decay of pure fullerenes does not vary with concentration and that the fluorescence decay of fullerene C_{60} is faster than that of fullerene C_{70}. In the case of mixed solution, the results are quite different and interesting where it is found that the lifetime varies with concentration of the acceptor indicating cluster formation and energy transfer due to size effect. From the study of optical gain we find that the optical gain is highly concentration dependent and it is positive for a narrow range of concentration. Furthermore, fullerene C_{70} has positive optical gain and thus it can be used as a gain material.

Study of band gap, X-ray diffraction and SEM for Fullerene C_{60} thin films

The study of fullerene C_{60} thin film shows that it is more conducting at low temperature. Also, the XRD of fullerene C_{60} film reveals that the grain size is in the nanometer range. This matches well with the results obtained from SEM measurements.

The main findings of this work are:

- Fluorescence spectra of fullerene C_{60} and fullerene C_{70} show aggregates formation in single solution.
- Forster's radius of both the pairs (fullerene C_{60}–coumarin C440 and fullerene C_{70}–quinizarine) is within the range 20 Å - 90 Å, which shows that both pairs can be used as spectroscopic rulers.
- The lifetimes of fullerene C_{60} and fullerene C_{70} molecules do not change with change in the solute concentration. The optical gain is found to be concentration dependent and only fullerene C_{70} shows positive gain. Therefore, fullerene C_{70} can be used as lasing material.