CHAPTER II

INSTRUMENTATION

Abstract

In this chapter, a brief description of the main experimental techniques that have been used for the study of radiative and nonlinear processes in photonic materials are described, along with the specifications of the equipment used for it.
2.1 Introduction

The accuracy and range of the data gathered in an experiment depends mainly on the experimental technique and the detecting instruments used. The characterization of the sample under study is done by using the data thus obtained. In many situations, proper computer interfacing with necessary software support renders the data acquisition more accurate and less time consuming. In this chapter, the experimental details and the instruments used for nonlinear as well as radiative studies are given. The five major experiments that have been described in this thesis are (1) fluorescence measurements, (2) amplified spontaneous emission (ASE) measurements from dye impregnated polymer matrices, (3) photothermal phase shift spectroscopy, (4) Z-scan measurements and (5) transmission measurements.

2.2 Instrumentation for fluorescence emission studies

There are many ways by which a molecule excited to higher energy state can return to the ground state. One of them is the fluorescence phenomena, in which major part of the absorbed energy is released in the form of radiative emission.

Fig. 2.1 Schematic diagram of a fluorometer.
The successful application of fluorescence method requires an understanding of the instrumentation. There are two reasons for this. First, fluorescence is a highly sensitive technique and the gain of the instruments can be increased to obtain observable signals. However these signals may be due to the amplification of background noises and not from the fluorophore of our interest. Secondly, there is no ideal fluorometer which yields true emission spectra, because of the nonuniform spectral output of the light sources and the wavelength-dependent efficiency of the monochromators and photomultiplier tubes. In order to overcome these the entire system is calibrated in wavelength using a Xenon arc lamp with know spectral output. The polarisation of the emitted light can also affect the fluorescence intensities. To obtain reliable spectral data one should be aware of and control these numerous factors. The schematic diagram of a typical fluorometer is given in Fig.2.1. The three principal components of the fluorometer are the source of excitation, the sample holder, and the detector. Fluorescence is initiated by the absorption of a quantum of radiation. Hence an intense steady radiation at a known wavelength is required. Laser has been used as the excitation source in the present studies. The individual components of fluorescence measurement are described below.

2.2.1 Nd:YAG laser

A Q-switched, Nd:YAG laser (Spectra Physics, DCR-11) which has a fundamental infrared beam at 1.06 \( \mu \text{m} \) was used. The high peak power of the Q-switched pulses permit second harmonic generation at 532 nm by introducing the KD\(^+\)P crystal in the beam path. The pulse width (FWHM) is 10 ns and maximum energy per pulse available is 275 mJ at 1.06 \( \mu \text{m} \) with a power stability of \( \pm 4\% \). The pulse repetition frequency is variable from 1 to 16 Hz. The laser cavity is a diffraction coupled resonator giving a TEM\(_{01}\) mode with the beam diffraction limited to a diameter of 6.4 mm. 1.06 \( \mu \text{m} \) and 532 nm radiations are linearly polarised in mutually perpendicular directions and the beam divergence is 0.5 mrad. The laser beam has a line width of 1 cm\(^{-1}\) with 220 MHz spacing between the longitudinal modes.

Fig.2.2 shows the schematic of the Nd:YAG laser system used in the present studies and Fig.2.3 shows the energy level diagram of neodymium doped Yttrium Aluminium Garnet, the active medium of the laser system. The active medium is optically pumped.
by a flash lamp whose output matches with the principal absorption bands in the red and the infrared. Excited electrons quickly drop to the $F_{3/2}$ level where they remain for a relatively long time. The most probable lasing transition is to the $I_{11/2}$ state, emitting a photon at 1.06 $\mu$m. Since the electrons in that state quickly relax to the ground state, its population remains low. Hence it is easy to build a population inversion between the upper and lower laser levels. There are other competing laser transitions, but their lower gain and higher threshold than 1.06 $\mu$m and wavelength selective optics limit oscillation to 1.06 $\mu$m. To increase the peak pulse energy and shorten the pulse duration, Q-switch is used. An electro-optic Q-switch introduces high cavity loss to prevent oscillation. The Q-switch comprises a polarizer, a quarter-wave plate and a Pockels cell. The polarisation characteristic of the Pockels cell is varied by applying a high voltage. With no voltage applied, the Pockels cell doesn't affect the polarisation characteristics of the light passing through it. But when the voltage is applied, it cancels the polarisation retardation of the quarter wave plate and the light suffers less loss.

![Fig. 2.2 The schematic of the Nd:YAG laser](image-url)
The optical cavity resonator of the DCR-11 is an unstable one. In a stable resonator the ray travels close to the optical axis and is reflected toward the optical axis by its cavity mirrors, so it is always contained along the primary axis of the laser and hence can extract energy from only a small volume near the optical axis of the resonator.

Fig. 2.3 The transition scheme and energy level diagram of the neodymium-doped yttrium aluminium garnet (Nd:YAG).

But in an unstable resonator the ray is reflected away from the optical axis by one of the cavity mirrors. The output beam has large diameter and thus they can efficiently extract energy from active media whose cross sectional area is large. The output coupler in an unstable resonator is a small highly reflecting mirror mounted on a clear substrate which lies on the optical axis of the resonator. Energy escapes from the resonator by diffracting around this dot, which gives it the name diffraction coupled resonator (DCR). It delivers a doughnut shaped beam profile with a divergence of 0.5 mrad. This laser provides trigger outputs to synchronize the oscilloscope, boxcar averager etc.
2.2.2 Sample holder

The geometrical arrangement of the exciting beam and the direction of viewing of the fluorescence output light in relation to the specimen is one of the most controversial points in the design of the fluorometer. This is because of the interplay of concentration quenching and inner filter effect, which are geometry dependent phenomena. Fluorescence is collected in the front surface geometry to avoid the reabsorption effects in the sample. But in the case of ASE measurements perpendicular geometry is used because of the directional nature of ASE signal. Because of its freedom from the effects of large amounts of scattered and transmitted excitation, this is the most preferred arrangement.

2.2.3 Detector

The detector system of a fluorometer consists of a monochromator-photomultiplier combination interfaced to a PC, using a suitable software. In order to increase the signal to noise ratio a boxcar averager was also used.

2.2.3.1 Monochromator

The basic function of a monochromator is to isolate a narrow band of electromagnetic radiation of required wavelength. The specification of the performance of a monochromator includes the dispersion and the stray light levels. Generally, the dispersion efficiency is given in nm/mm, where the slit width is expressed in mm. Also it must have good light gathering power, minimum ambient light interference and good resolution. The monochromator has input and output slits of variable height and width. The light intensity which passes through a monochromator is proportional to the square of the slit width. Larger slit widths yield increased signal levels, therefore higher signal-to-noise ratios. The finer slit width provides better resolution at the expense of detected power of light. Grating monochromators may have planar or concave gratings. Concave gratings produced by holographic method are preferable in fluorescence studies, since imperfections are rare and stray light and ghost image interferences are absent.

A 1 meter long scanning spectrometer, Spex Model 1704, having a maximum resolution \( \approx 0.05 \) Å was used to conduct fluorescence measurements. The monochromator
covers a spectral range 350-950 nm, using a grating with 1200 grooves/mm blazed at 500 nm and spectral band pass 0.1 Å.

Fig. 2.4 Optical layout of the Spex monochromator

The start and end position of the scan and rate of scanning can be programmed using a microprocessor controlled compudrive arrangement. The output of the Spex monochromator is coupled to a thermoelectrically cooled photomultiplier tube (Thorn EMI, model KQB 9863, rise time 2 ns).

2.2.3.2 Photomultiplier tube (PMT)

The photomultiplier combines photocathode emission with multiple cascade stages of electron amplification to achieve a large amplification of primary photocurrent within the envelope of the phototube with the output current of the PMT remaining proportional to the input light intensity. The built-in amplifier system of the PMT containing a series of secondary electrodes (dynodes) may have up to 15 amplification stages, such that one photoelectron can give rise to as many as $10^8$ electrons reaching the anode. It is this anode which provides the signal current that is read out. The sensitivity of a
typical PMT can be varied by changing the voltage applied to the cathode and dynodes. The ultimate sensitivity is limited by the dark current which is caused by the ejection of electrons from the cathode by thermal activation or by traces of radioactivity in the surroundings causing luminescence of the envelope.

For a large number of applications, the PMT is the most practical sensitive detector available. The basic reason for the superiority of the PMT over other detectors is the secondary amplification which makes it uniquely sensitive among photosensitive devices currently used. The PMT has a photocathode in either a side-on or a head-on configuration. The side-on type receives incident light through the side of the glass bulb, while the head-on type has a semitransparent photocathode (transmission-mode photocathode) and it provides better uniformity than the side-on type having a reflection-mode photocathode. A schematic representation of a typical photomultiplier tube is given in the Fig.2.5.

The Spex monochromator was coupled to a thermoelectrically cooled (-50°C) Thorn EMI photon counting PMT with S-20 cathode with quantum efficiency of 22%. A high negative voltage bias of 1.7-2.1 kV was usually given to the cathode of the PMT. The spectral response of S-20 cathode is shown in Fig.2.6. To minimize the noise in the signal, the output of the PMT is fed to a boxcar averager.

2.2.3.3 Gated Integrator and Boxcar averager

During pulsed laser experiments, a form of gating and averaging is required so as to have a higher S/N ratio. The gated integrator and boxcar averager is used for analysing the noisy, transient, repetitive signals which are characteristics of experiments with pulsed lasers. We have been using a boxcar averager (Stanford Research Systems) for signal averaging and gating.

The main modules of the system used in the present studies are

(1) Gated integrator and boxcar averager module (SR250)
(2) Gate scanner module (SR200)
(3) System mainframe (SR280)
(4) Display module (SR275)
(5) Computer interface module (SR245)
(6) Data acquisition program (SR270)
Fig.2.5 Schematic representation of a typical photomultiplier tube.

Fig.2.6 Spectral response of S-20 cathode.
The Stanford Research Systems SR250 module consists of a gate generator, a fast gated integrator and exponential averaging circuitry. The gate generator, triggered by the laser pulse, provides an adjustable delay from a few ns to 100 ms, before it generates a continuously adjustable gate the width of which can be varied from 2 ns to 15 ms. The delay may be set by a front panel potentiometer. The signal at the gate is integrated by the fast gated integrator and is normalised by the gate width to provide a voltage which is proportional to the part of the input signal level at the gate. The sensitivity control of the boxcar averager provides further amplification of the signal. By fixing the delay and the gate width, the voltage from the part of the signal pulse alone is measured and improvement in the signal to noise (S/N) ratio of the detection is achieved. A moving exponential average of 1-10,000 samples are available at the averaged output. This traditional averaging technique is useful for pulling out small signals from noisy backgrounds. As one averages many noisy samples of a signal, the average will converge to the mean value of the signal and the noise will average to zero. Usually the signal was averaged for 10 successive pulses. Averaging over very large number of pulses increases the S/N ratio, but the time response of the system will suffer.

The SR200 gate scanner provides the signal needed to scan the SR250's delay multiplier (to scan the sample gate through the wave form) and to control the oscilloscope. Single or multiple scans may be done in the forward or reverse direction over any portion of the waveform. The X-axis output always ramps between 0 and 10 Vdc, regardless of the dial settings, providing a convenient interface to chart recorder.

The SR280 and SR275 provides the power necessary to the SRS modules and has three displays for monitoring the outputs. The SR245 computer interface is a versatile module capable of providing a variety of scanning, counting and communication functions. The SR270 is a software designed to acquire, display and manipulate data taken from the SR250 Boxcar integrator with the SR245 computer interface module. The system is connected to the personal computer through the serial port.

2.2.4. Experimental setup for fluorescence studies

The 532 nm beam from the pulsed Nd:YAG laser was used as the excitation source. For the measurement of the degree of polarisation, a polariser was kept in the emission
beam path. The samples were of size (13x10x1.5) mm³ and were mounted in such a way that fluorescence emission was collected from the front surface to avoid reabsorption of the emitted light. Using collimating optics, the emission was made to fall onto the entrance slit of the Spex monochromator to which a PMT is connected head-on. Proper filters were used to avoid the entry of the scattered laser radiation into the monochromator. The emission was wavelength scanned in the desired region and the optical intensity was detected by the Thorn EMI photomultiplier tube. The signal from the PMT is gated and averaged using the gated integrator and boxcar averager and its output is interfaced to the computer to obtain the fluorescence spectrum.

2.3 Instrumentation for gain spectroscopy

Interest in investigating stimulated emission from organic dyes has made it necessary to develop a system that will measure gain of the medium. The spontaneously emitted photons travelling along the path of the exciting pulse in dyes are amplified by stimulated emission. Using ASE, gain and other related properties of the laser active medium can be found. Gain spectroscopy can provide information which are not available from ordinary absorption and emission spectroscopy⁹. The experimental setup and the principal hardware components of the gain spectroscopy are similar to that of fluorescence studies, with the exception that the excitation beam is focused onto the sample using a cylindrical lens, so that larger area of the medium is excited. Also the emission is observed with a transverse geometry in which the detection system is kept in a direction perpendicular to the excitation beam.

The experimental setup consists of a source of excitation, the sample holder, and the detector. The discussion of these components are given in sections 2.2.1 - 2.2.3. The gain studies were done by varying the excitation intensity as well as the dye concentration in the sample. For measuring the laser energy during the experiment, a laser power energy meter was used. For temporal studies, a computer interfaced digital storage oscilloscope was used to monitor the PMT output.

2.3.1 Laser power energy meter

In order to measure the power output at 532 nm a laser power meter (Scientech model 362) was used. The detector is a disc calorimeter that employs a calibrated thermopile
which generates a voltage proportional to the heat that is liberated from the absorption of the input laser flux. Many thermoelectric junctions are arranged in series and sandwiched between the absorption surface which produces heat due to laser absorption. The heat flow is proportional to the power output of the laser beam and substantially independent of the laser beam spatial distribution. The thermopile output is a linear low impedance, dc signal of approximately 0.09 volts/Watts. It has flat spectral response in the region of 400 nm to 1200 nm, and can be used with CW and pulsed lasers for measuring from 0 to 10 Watts.

2.3.2 Digital storage oscilloscope

A digital storage oscilloscope from Tektronix, 100 MHz, (TDS 220) was used to monitor the signal from the PMT. It has a 1 GS/s sampling capability with storage and averaging facilities. The oscilloscope was synchronised with the laser pulse. RS232 communication port was used for transferring data to the personal computer for further processing.

2.3.3 Experimental setup for gain spectroscopy

![Schematic experimental setup for gain spectroscopy](image)

CL-Cylindrical lens, S-Shutter, PS-Polyacrylamide sample, PMT-Photomultiplier tube. DSO-Digital storage oscilloscope.

Fig.2.7 Schematic experimental setup for gain spectroscopy

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The schematic of the experimental setup is shown in Fig.2.7. The 532 nm, 10 ns pulses from an Nd:YAG laser (Quanta-Ray DCR-11) is used as the pump laser source. A cylindrical lens is used to focus the beam to a line onto the sample, thereby defining the gain region. By using an optical shutter the length of the pumped region can be varied from L to L/2 where L is the length of the sample. The ASE output was taken from the transverse direction and was allowed to fall onto the slit of a one meter Spex monochromator (1200 grooves/mm, 100 mm by 100 mm grating blazed at 500 nm). The output from the monochromator was passed through a thermoelectrically cooled Thorn EMI photomultiplier tube (PMT, model KQB 9863) which was coupled to a boxcar averager/gated integrator (Stanford Research Systems, SR250) and interfaced to a computer using SR270 data acquisition program. For temporal studies a computer interfaced Tektronix 100 MHz storage oscilloscope was used.

2.4 Photothermal phase shift spectroscopy

For the application of polymers as host-matrix, the resistance to laser damage is of vital importance. Photothermal phase shift spectroscopy (PTPS) was used to study the damage threshold of these matrices. The idea of PTPS is simple: a laser beam (pump beam) passing through the medium causes heating of the medium of interest. The heating modifies the refractive index of the laser irradiated region. The change in the refractive index of the medium is detected by a low power laser beam (probe beam).

The principal part of PTPS consists of a Michelson Interferometer (MI). For the construction of the MI, a 5 mW He-Ne laser was used as the light source along with two highly reflecting end mirrors equidistant from the beam splitter so that interference fringes are obtained. On one of its arm, close to the beam path, the sample whose damage threshold is to be measured is kept. A focused high power Nd:YAG beam is allowed to fall on the sample, which produces plasma when the laser intensity is above damage threshold. The presence of the plasma causes a refractive index variation in the path length of the probe beam, which can be detected as fringe shift in the MI. The shift in the interference fringe produced can be measured as intensity variation over an optical detector which is position sensitive. An optical fiber was used for this purpose. One end of the fiber acts as an aperture to sample the variation of...
probe beam intensity. The other end of it is introduced into the monochromator-PMT assembly whose output is connected to a digital storage oscilloscope and the fringe shift is measured as a voltage variation. The use of optical fiber introduces a certain amount of geometrical flexibility in the experimental setup.

The description of most of the instruments were given in the above sections. He-Ne laser which is used as a probe beam in PTPS and power meter to measure the 1.06 μm radiation incident on the sample while studying the damage threshold are described in the following sections.

2.4.1 He-Ne laser

As the probe beam in PTPS we have used 632.8 nm emission from a He-Ne laser (Model 105-1, Spectra Physics). It has TEM00 mode, with a power of > 5 mW. The beam divergence is 1 mrad\(^{13}\).

2.4.2 Power meter

In order to measure the energy of 1.06 μm radiation, pulsed laser energy monitor (Delta Developments) was used. This is an on-line energy meter and uses a polarisation compensated beam splitter to sample the beam, 85% of which is transmitted\(^{14}\). The sampled beam strikes a retroreflecting diffuser and reaches the photo-diode via a 'range plate' which attenuates the light appropriately for the range of energies being measured. The geometry of the energy meter is such that all positions on the diffuser give equal signals. Different range plates can be used for different energies and wavelengths being measured. The wavelength response of the energy meter can be varied from 200 nm to 1100 nm. Each range plate gives a factor of 30 in the energy giving full scale deflection. It has a usable range of 100:1. A switch allows readings to be referred to either the energy entering or leaving the instrument. BNC sockets provide pulse shape, pulse energy, trigger input, trigger output etc. The energy meter can be triggered externally with laser pulses or internally.

2.4.3 Experimental setup for phase shift spectroscopy

The basic element of PTPS is a Michelson Interferometer, with a 5 mW He-Ne laser beam as the light source (probe beam), as shown in Fig.2.8. The optical setup is aligned so as to get a well defined fringe pattern. The beam in one of the arms of MI

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passes parallel and very close to the target surface. High power laser radiation from a pulsed Nd:YAG laser (pump beam) at wavelengths 1.06 μm and 532 nm were focussed on to the target in order to produce plasma.

Fig.2.8 Experimental setup for phase shift spectroscopy

The samples used were discs of PMMA and dye doped polyacrylamide. The point of irradiation was shifted by mechanically rotating the target after each measurements so that fresh location is available for each pulse. The probe beam passes grazing the sample surface so that the length of the plasma near the target is taken as equal to the pump laser spot size. The shift in fringe pattern is measured as a voltage change using a PIN photodiode and displayed on the digital storage oscilloscope. The whole setup was properly vibration isolated by using a vibration isolation table. The schematic of the experimental setup for the PTPS is as shown in Fig.2.8.

2.5 Instrumentation for Z-scan technique

Using Z-scan technique which is a simple single beam method, the refractive and absorptive nonlinearity of certain nonlinear materials were studied. The Kerr as well
as the thermal nonlinearity can be studied using this method. This technique is based on the principle of spatial beam distortion and has a very high sensitivity.

In Z-scan technique a Gaussian laser beam is used, whose transmittance through a nonlinear medium as a function of sample position Z with respect to the focal plane is measured in the far field with an aperture (closed Z-scan) and without an aperture (open Z-scan) to measure refractive and absorptive nonlinearities respectively. The medium acts as a lens whose effective focal length varies with the incident intensity. When the medium is moved through the focal plane the varying intensity causes changes in the transmission of the beam. This change will be reflected in the intensity distribution at the aperture in the far field. The amount of energy transmitted by the aperture will depend on the sample location on the z-axis and on the sign of the nonlinearity. A Gaussian laser beam is used to induce the nonlinearity, a translator stage moves the sample through an intensity gradient obtained by a focusing lens and a detector to measure the transmittance of the sample are the different parts of the experimental setup. A general account of the equipment necessary for this technique is given below. As the exciting source Ar+ laser was mainly used. In the case of CW lasers for increasing the ratio of signal to noise Lock-in detection was made use of. Chopper, photodiode and power meter form other parts of the measuring system.

2.5.1 Ar+ laser

CW Ar+ laser was also used for Z-scan and emission studies. The Spectra Physics Model 171 CW Ar+ laser system gives high CW laser output power\(^6\). It consists of a laser head, power supply and a separate power meter to monitor the output power. The laser head houses the beryllium oxide plasma tube and optical resonator structure. The power meter continuously monitors the output power and the output laser power varies only within ±5% over periods of days. The major wavelengths of the laser output are 514.5, 496.5, 488, 476.5 and 457.9 nm. The laser has a water cooling system that cools the plasma tube.

2.5.2 Translator and sample holder

A motorised translator was used to move the sample along with the holder through the focal plane. It is graduated and the focusing lens is adjusted so that the focal point
comes at the middle of the translator. The height of the sample holder with cuvette mounted on it can be adjusted.

2.5.3 Detector

For the measurement of absorptive nonlinearity the total transmitted power through the sample was measured. For this Scientech power meter with large aperture was used. The details of the power meter was described in section 2.3.1. In order to measure the refractive nonlinearity in the far field the transmittance of the sample with an aperture was done. A silicon photodiode was used for the measurements.

2.5.3.1 Silicon photodiode

HP 4207 silicon planar PIN photodiodes are ultra-fast light detectors for visible and near infrared radiation. The speed of response of these detectors is less than one nanosecond. Laser pulses shorter than 0.1 ns may be observed. The frequency response extends from dc to 1 GHz. It has a sensitivity of NEP < -108 dBm.

2.5.4 Mechanical chopper

Mechanical choppers are designed to be used with a Lock-in amplifier for the purpose of synchronous detection. The optical chopper Model SR 540 C from Stanford Research systems was used in the present experiments. It is used to square wave modulate the intensity of optical signals. The chopping frequency can be varied from 4 Hz to 400 kHz with a frequency stability of 250 ppm. It also provides the reference signal to the Lock-in amplifier.

2.5.5 Lock-in amplifier

Lock-in amplifiers are used to detect and measure very small ac signals. Accurate measurements may be made even when the small signal is obscured by noise sources even thousand times larger. They use phase sensitive detection to single out the component of the signal at a specific reference frequency and phase. Lock in measurements require a frequency reference. Typically an experiment is conducted at an optimized frequency from an optical chopper. The lock-in detects the response from the experiment at the reference frequency. The Lock-in amplifier used was SR 850 DSP from Stanford Research Systems. It has a full scale sensitivity of 2 nV to 1 V with a CMRR of 90 dB.
The spectral data can be stored in a 3.5 in diskette or the instrument can be directly interfaced to a personal computer.

2.5.6 Experimental setup for Z-scan

The experimental setup, along with theoretical details for Z-scan is given in Chapter I. A cuvette of thickness 0.49 cm was moved along the intensity gradient produced by a focusing lens of focal length 18 cm. The absorptive nonlinearity was measured by monitoring the input and output laser powers using a power meter. Using Ar⁺ laser thermal nonlinearity produced has been measured. For detecting the absorptive nonlinearity, a powermeter was used, but for the refractive nonlinearity in the far field (with aperture) a silicon pin diode connected to a Lock-in amplifier for detection was used. Triggering of the lock-in amplifier was done using the output from a mechanical chopper. For studying the Kerr nonlinearity high power Nd:YAG laser was used.

2.6 Conclusions

The various aspects of the instrumentation used for the nonlinear and radiative studies are described in this chapter. Some variation in the instrumentation was necessary for further studies in nonlinearity which are described in the relevant sections.
References


[7] Instruction manual, Photomultiplier tubes, EMI.


[10] Instruction manual Scientech 362 power energy meter.


