CHAPTER 3

EXPERIMENTAL TECHNIQUES

3.1 INTRODUCTION

The growth of interest in using all-optical devices in modern technology such as telecommunications has been prominent, especially in the last few years. The substitution of electronic devices by the optical counterparts in communications technology, proved to be an impressive accelerator for processing, transport, and storage of data. In this respect, manipulation of amplitude, polarization, direction or phase of the optical beam gains significance. Such a control over light by light itself is possible through various NLO phenomena. In order to carry out these manipulations efficiently, an understanding of the underlying nonlinear optical phenomena is essential. The structural prerequisite for the verification of NLO phenomena in organic compounds is the presence of a network of delocalized \( \pi \)-conjugated electrons, which infer high polarizability and fast charge redistribution when the conjugated molecule interacts with rapidly varying intense electromagnetic fields like those of laser.

A greater understanding of the photo physical properties of the dye molecules is essential to understand the nonlinear optical properties of dye molecules to a better extent. Therefore, a study of the spectral characteristics of the laser dyes becomes essential. This chapter explains the details of spectrophotometer and spectrofluorometer used for recording absorption and fluorescence spectra, pico second time correlated single photon counting.
technique for lifetime measurements, the experimental set-up used for measuring the nonlinear refractive index by Z-scan and optical limiting studies of dyes in liquid and solid medium.

3.2 SPECTROPHOTOMETER

The basic block diagram of spectrophotometer (SL 159 UV – VIS spectrophotometer) is shown in Figure 3.1 (SL159 UV-VIS spectrophotometer). This consists of a light source (Deuterium (D₂) and Tungsten (W) halogen lamps), monochromator (Czerny-Turner type with 1200 lines/mm holographic grating), detector (wide range Photo diode), and computer. The light from the halogen lamp is passed through the monochromator to get the required wavelength. This is allowed to pass through the sample. The amount of light that is transmitted depends on the absorption of the solution. The transmitted light is allowed to fall on a detector, which is a measure of the transmittance, and from this value the instrument directly gives the value of absorption.

![Rotates to achieve scan](image)

**Figure 3.1 Block diagram of spectrophotometer**

3.2.1 Absorption Spectra

The absorption spectra of the dyes in liquid medium are recorded using the Perkin-Elmer Lamda 35 spectrophotometer. The concentration of the dye in liquid are kept at 0.01 mM. The spectral parameters of the dyes are calculated as follows:

The bandwidth \((\Delta \nu)_{\frac{1}{2}}\) in cm\(^{-1}\) of the absorption spectra is calculated by measuring the full width at half maximum of absorbance.
The molar extinction coefficient \( \varepsilon(\lambda) \) (in L M\(^{-1}\) cm\(^{-1}\)) at the peak wavelength of the absorption spectra is calculated using the relation,

\[
\varepsilon(\lambda) = \frac{OD}{LC}, \text{ in L mol}^{-1}\text{ cm}^{-1}
\]  

(3.1)

where OD is the absorbance at the peak wavelength of the absorption spectra, L is the path length of the dye solution medium (in cm) and C is the molar concentration of dye (in moles/liter).

The oscillator strength \( f \) was calculated by using the relation,

\[
f = 4.33 \times 10^{-33} \varepsilon(\lambda) ((\Delta \nu)^{1/2}) \text{ in L/mol cm}^{2}.
\]  

(3.2)

### 3.3 SPECTROFLUOROMETER

Spectrofluorometer is used to obtain the fluorescence or excitation spectrum of a liquid sample. This instrument is interfaced with a computer and the required excitation wavelength, emission scan range, scan speed etc., can be fed to the instrument through the computer. We can get the fluorescence spectrum excitation spectrum either on the CRT or on a dot matrix printer. The basic block diagram of spectrofluorometer is shown in Figure 3.2. Spectrofluorometer consists of a xenon lamp which excites the sample. The fluorescence is collected at right angles to the direction of excitation. The xenon lamp emits light in the entire spectrum including UV, visible and IR. Therefore, using monochromator, we have to select a particular wavelength out of this band to excite the sample, which is controlled by stepper motor. Similarly, the fluorescence of the sample is also broad band and this is also passed through the monochromator to get a spectrum. The output fluorescence is detected by a photo detector and this signal is amplified (variable gain amplifier) so that the output relative intensity can be calibrated to a fixed value. Fluoromax 2 spectrofluorometer
and Perkin Elmer Lambda LS 45 were used to record the fluorescence spectra of the dye solutions.

![Block diagram of spectrofluorometer](image)

**Figure 3.2 Block diagram of spectrofluorometer**

### 3.3.1 Fluorescence spectra

The fluorescence spectra of the dyes in liquid medium are recorded at low concentration of the dye (0.01 mM) and these spectra are corrected using quinine sulphate in 0.1N H$_2$SO$_4$ and fluorescein in 0.1N NaOH. The fluorescence spectral bandwidth (FWHM) is calculated by measuring the full width at half maximum of fluorescence intensity.

In order to understand the solute-media (solvent/polymer) interactions, stokes shift is calculated from the absorption and fluorescence spectra. The Stoke’s shift ($\nu_a-\nu_f$) of the absorption and fluorescence maxima of dyes in different media is calculated using the relation,

$$ (\nu_a-\nu_f) = (1/\lambda_a - 1/\lambda_f) \text{ in cm}^{-1} . $$

(3.3)
3.4 FLUORESCENCE LIFETIME

3.4.1 Introduction

The time correlated single photon counting technique is used for determining the fluorescence lifetime of dye molecules in liquid and in solid medium. Fluorescence lifetime is the decay time of the molecule and it can be defined in the time domain in terms of the rate of depopulation of the first excited singlet states following δ- function (i.e., impulse) optical excitation from the ground state. As the excited state population is proportional to the fluorescence quantum intensity, the fluorescence lifetime can be determined experimentally by measuring the time taken for the fluorescence intensity to fall to 1/e of its initial value following δ-function excitation. This observation forms the basis of time correlated single-photon counting technique whereby the quantum nature of the light enable the time distribution of individual photon within the decay profile to be recorded.

3.4.2 The Time Correlated Single Photon Counting Technique

The time-correlated single photon counting fluorometer consists of a pulse light source, usually a flash lamp or mode locked laser, which generates multi photon excitation pulses, which stimulates absorption in an assembly of sample molecules. At low levels of excitation power, each sample molecule absorbs one photon at the most, on a time scale which is instantaneous. The subsequent de-excitation of these molecules through the emission of fluorescence photon occurs with a distribution of the time delays which is normally exponential. The single photon timing technique records this distribution by measuring the time delays of the individual fluorescence photons with respect to the excitation pulse. When the excitation occurs, a synchronization pulse or ‘start’ timing pulse trigger the charging of a capacitor in the TAC (Time-to-Amplitude converter). The voltage on the
capacitor increases linearly until either a preselected time range is reached or a ‘stop’ timing pulse is detected. The latter is initiated by detection of a fluorescence photon and the ‘start-stop’ time interval generates a proportional voltage across the capacitor. This voltage pulse is stored according to amplitude using an analog-to-digital converter within a multichannel analyzer (MCA). On repeating the ‘start-stop’ cycle many times, a histogram representative of the fluorescence decay is acquired in the MCA memory. The data parameter can then be extracted using numerical and statistical procedures. Photomultipliers are the most widely used single-photon timing devices, and in order to minimize the registering of noise pulses and to ensure that the timing definition of the ‘start’ and ‘stop’ pulses is largely independent of the signal pulse height, discriminators are used. The shape provides the time definition for the pulses in the start and stop channel. The aim of decay time experiment is to study decay kinetics. This is an indirect process: The measurement system yields fluorescence decay curves, consisting of intensity values at some hundreds or thousands of consecutive short-time periods (channels) after the time of pulsed excitation. The parameters describing the kinetics are then obtained by reducing these data using statistical methods of data analysis (Lakowicz 1991).

3.4.3 Experimental set-up

The fluorescence decay measurements of the dyes studied were recorded using IBH time correlated single photon counting spectrometer (TCSPC), with micro channel plate photomultiplier tube (MCP-PMT) as detector and 375 nm LED as excitation source. TCSPC is a digital technique, counting the photons, which are time correlated with the excitation pulse. The heart of the method is a time-to-amplitude converter. The sample is repetitively excited using a pulsed light source. Each pulse was optically monitored by a high-speed photodiode, to produce a start signal, which is used to trigger, the voltage ramp of the TAC. The voltage ramp is stopped
when the first fluorescence photon from the sample is detected. The TAC provides an output pulse whose voltage is proportional to the time between start and stop signals. A multichannel analyzer converts this voltage to time channel using an analog-to-digital converter. The MCA builds up a probability histogram of a count versus channels by summing over many pulses. The counting continued until 10,000 counts were collected in the peak channel.

The light from LED source was focused on the sample and the fluorescence photons from the sample were collected at right angle to the excitation beam. The emitted photons were detected by a MCP-PMT (Hamamatsu R3809U) after passing through the monochromater (f/3). A schematic representation of picosecond time correlated single photon counting set-up is shown in Figure 3.3. The photon signal from the MCP-PMT was fed into the CFD and the NIM out from the CFD serves as a stop signal in the TAC. The MCP output was directly read on a rate meter. The TAC output is fed in the MCA Card (Oxford Corporation U.K) and data collection was carried out by the software (Data Station 2000) provided by IBH. Repetitive laser pulsing and emitted photon collection produces a histogram of voltage (time) against counts. The histogram represents the measured decay. For recording the lamp profile, a scatterer was placed instead of the sample and the same procedure was repeated. The response time of this instrument is around < 1 ns.

3.4.4 Fluorescence decay analysis

The measured fluorescence decay is the convolutions of true fluorescence decay, excitation function and the instrument response function. The fluorescence kinetic parameters (lifetime, amplitudes etc.) are obtained by deconvoluting the excitation and the instrument response function from the measured fluorescence decay.
Figure 3.3  Experimental setup of time correlated single photon counting technique
The data analysis was carried out by the software provided by IBH (DAS-6) which is based on reconvolution technique using iterative non-linear least square methods. The global analysis of fluorescence decays were carried out using the PTI global analysis software. The reconvolution is preceded by the series of iterations until a Chi-square is reduced. The quality of fit is normally identified by the reduced $\chi^2$, weighted residual and the auto correlation function of the residuals. The average life time ($\tau$) (for bi-exponential decay) was calculated from the relation

$$\tau = \frac{\tau_1 B_1 + \tau_2 B_2}{100}$$  \hfill (3.4)

The average life time ($\tau$) (for tri-exponential decay) was calculated from the relation

$$\tau = \frac{\tau_1 B_1 + \tau_2 B_2 + \tau_3 B_3}{100}$$  \hfill (3.5)

where $\tau_1, \tau_2, \tau_3$ is the life time and $B_1, B_2$ and $B_3$ are the relative amplitudes emitting species.

### 3.5 INTRODUCTION TO NONLINEAR OPTICS

The third-order nonlinearities are responsible for the variation of refractive and absorptive properties of media and the propagation of intense light through the materials. Numerous studies have been carried out on the analysis of these properties, due to the growing interest in the applications of nonlinear optical propagation effects for optoelectronics, various nonlinear optical devices, optical switching and limiting etc. The nonlinear optical studies of various materials fullerenes, dyes, metals, crystals, and liquids in different wavelength and intensity can be measured by Z-scan technique.
Z-scan technique (Sheik-Bahae et al 1989, 1990), based on the spatial distortion of a laser beam, passed through a nonlinear optical material, is widely used in material characterization because of their simplicity, high sensitivity and well-elaborated theory. The opportunity to conduct simultaneous measurements of various nonlinear optical parameters in one set of experiments also makes this technique attractive and applicable for different materials. This method yields both the sign and the magnitude of the nonlinearity, and the value of the nonlinear refractive index, $n_2$, may be easily extracted from experimental data with a minimum of analysis. Z-scan studies, also yield important information regarding the response time and the dynamics of the transient processes contributing to the nonlinear refractive index (Sucharita Sinha et al 2000).

3.5.1  Z-Scan Technique for Determining the Nonlinear Refractive Index

3.5.1.1  Principle of Z-scan Technique

The basic idea behind the Z-scan technique is self-focusing. Using a Gaussian laser beam, the transmittance of a nonlinear medium through a finite aperture placed in the far field as a function of the sample position (Z) with respect to the focal plane can be measured. The principle is to move the sample along the optical axis in the vicinity of the laser beam focused on an external lens. For each position of the sample around the focus, the induced lens inside the sample possesses different focal lengths. This focal length depends on the incident Gaussian shape. The experiment consists of the measurement of the irradiance through a small aperture in a far field for each position versus the focus position. When the sample has negative refraction, it can be regarded as a concave lens near the focal position. As the sample position becomes close from the negative position of z to the focal position, the sample shifts the focal position to the positive $z$- direction. Therefore the
beam at the aperture plane is suppressed and the intensity of the detectable beam becomes larger than the intensity without the sample at the negative side. As the sample position becomes close from the positive position of z to the focal position, the sample spreads the beam at the aperture plane and detectable intensity is larger at the positive side. Any nonlinear absorption present in the sample can be found in a measurement where the aperture is removed (open aperture Z-scan) and is replaced by a lens to collect the entire transmitted laser beam.

A pre-focal transmittance maximum (peak) followed by a post-focal minimum (valley) determines the refractive nonlinearity is negative. On the other hand, for positive refractive nonlinearity, there will be a valley followed by a peak (Figure 3.4). An extremely useful feature of this technique is that the sign of the nonlinear index is immediately known from the Z-scan traces.

![Characteristic curves depicting both positive and negative nonlinear refraction as measured by Z-scan](image)

Figure 3.4 Characteristic curves depicting both positive and negative nonlinear refraction as measured by Z-scan
For the present study the Z-scan technique is performed on the dyes in solvents and dye doped polymer films in order to determine third-order optical nonlinearities using the theory proposed by Sheik-Bahae et al (1990).

3.5.1.2 Theory

In general, for cubic nonlinearity, the index of refraction $n$ is expressed in terms of $n_2$ (esu or m$^2$/W).

$$n_0 + \frac{n_2}{2} |E|^2 = n_0 + n_2 I$$  \hspace{0.5cm} (3.5)

where $n_0$ is the linear refractive index, $E$ is the peak electric field (cgs) and $I$ is the incident light intensity (MKS) within the sample. The relation between esu and m$^2$/W is given by

$$n_2 \text{(esu)} = \left( \frac{cn_2}{40\pi} \right) n_2 \left( \frac{m^2}{W} \right)$$  \hspace{0.5cm} (3.6)

where $c$ (m/s) is the velocity of light in vacuum.

Assuming a TEM$_{00}$ Gaussian beam of waist radius $\omega_0$ traveling in the $+Z$ direction, the magnitude of the electric field $E$ can be written as,

$$|E(r, Z, t)| = |E_0(t)| \left| \frac{\omega_0}{\omega(Z)} \right| \exp \left( \frac{-r^2}{\omega^2(Z)} \right)$$  \hspace{0.5cm} (3.7)

where $\omega^2(Z) = \omega_0^2 \left( 1 + Z^2 / Z_0^2 \right)$ is the beam radius at $Z$, $Z_0 = k\omega_0^2/2$ is the diffraction length of the beam, $k = 2\pi/\lambda$ is the wave vector and $\lambda$ is the wavelength of light. $E_0$ denotes the radiation electric field at the focus and contains the temporal envelope of the laser beam.
If the sample length is small enough such that changes in the beam diameter within the sample due to either diffraction or nonlinear refraction can be neglected, the medium is regarded as thin. In this case, the amplitude and nonlinear phase change $\Delta \phi$ of the electric field within the sample is expressed by

$$\frac{d\Delta \phi}{dz} = (2\pi/\lambda) \Delta n$$

(3.8a)

and

$$\frac{d|E|}{dz} = -(\alpha/2)|E|$$

(3.8b)

where $\alpha$ is the linear absorption coefficient and $z$ is the depth within the sample. Equations (3.8a) and (3.8b) are solved to give the phase shift $\Delta \phi$ at the exit surface of the sample, which simply follows the radial variation of the incident irradiance at a given position of the sample $Z$.

$$\Delta \phi(r, Z, t) = \frac{\Delta \Phi_0}{1 + Z^2 / Z_0^2} \exp\left(-\frac{2r^2}{\omega^2(Z)}\right)$$

(3.9)

The on-axis phase shift at the focus is defined as

$$\Delta \Phi_0(t) = k\Delta n_0(t)L_{eff}$$

(3.10)

where, $L_{eff} = (1-e^{-\alpha L})/\alpha$, with $L$ the sample length and $\alpha$ is the linear absorption coefficient. Here $\Delta n_0 = n_2 I_0(t)$ with $I_0(t)$ being the on-axis irradiance at focus (i.e. $Z = 0$). One can ignore the Fresnel reflection losses such that $I_0(t)$ is the irradiance within the sample.
The electric field at the surface of the sample now contains the nonlinear phase distortion.

\[ E'(r, Z, t) = E(r, Z, t) \exp(-\alpha L/2) \exp(i\Delta \phi(r, Z, t)) \]  

(3.11)

By virtue of Huygens’s principle, the far field pattern of the beam at the aperture plane can be obtained through Henkel transformation of \( E' \).

Having calculated the electric field at the aperture, one can obtain the normalized instantaneous \( Z \)-scan power transmittance as,

\[ T(z,t) = \frac{\int_{0}^{r_{a}} |E_{a}(\Delta \Phi_{0}, r, Z, t)|^2 rdr}{S \int_{0}^{\infty} |E_{a}(0, r, Z, t)|^2 rdr} \]  

(3.12)

where \( r_{a} \) is the aperture radius and \( S \) is the aperture transmittance in the linear regime.

The measurable quantity \( \Delta T_{p,v} \) can be defined as the difference between the normalized peak and valley transmittances, \( T_{p} - T_{v} \). The variation of this quantity as a function of \( |\Delta \phi_{0}| \) is given by (Xia et al 1994).

\[ \Delta T_{p,v} = 0.406(1-S)^{0.25} |\Delta \phi_{0}| \]  

(3.13)

where \( S \) is the aperture linear transmittance (0.4), \( \Delta \phi_{0} \) is the on-axis phase shift.

The on-axis phase shift is related to the third order nonlinear refractive index by,

\[ |\Delta \phi_{0}| = kn_{2} L_{eff} I_{0} \]  

(3.14)
where \( k = 2\pi/\lambda \), \( L_{eff} = [1 - \exp(-\alpha L)]/\alpha \) is the effective thickness of the sample, \( \alpha \) is the linear absorption coefficient, \( L \) is the thickness of the sample, \( I_0 \) is the on-axis irradiance at focus and \( n_2 \) is the third order nonlinear refractive index.

The nonlinear absorption coefficient \( \beta \) can be estimated from the open aperture Z–scan data. The normalized transmittance for the open aperture condition (Sheik-Bahae et al 1990) is given by,

\[
T(z, S = 1) = \sum_{m=0}^{\infty} \frac{[-q_o(z)]^m}{(m+1)^{3/2}}
\]

(3.15)

for \( q_o(0) < 1 \), where \( q_o(z) = \beta I_o L_{eff}/(1 + z^2/z_R^2) \), \( z_R = k\omega_o^2/2 \) is the diffraction length of the beam and \( \omega_o \) is the beam waist radius at the focal point and \( k = 2\pi/\lambda \) is the wave vector.

The experimental measurements of \( n_2 \) and \( \beta \) allow one to determine the real and imaginary parts of the third-order nonlinear optical susceptibility \( \chi^{(3)} \) according to the following relations (Cassano et al. 2001),

\[
\text{Re}\chi^{(3)} \text{ (esu)} = 10^{-4} \varepsilon_0 c^2 n_0^2 n_2 (\text{cm}^2/\text{W}) / \pi
\]

(3.16)

\[
\text{Im}\chi^{(3)} \text{ (esu)} = 10^{-2} \varepsilon_0 c^2 n_0^2 \beta (\text{cm}/\text{W}) / 4\pi^2
\]

(3.17)

where \( \varepsilon_0 \) is the vacuum permittivity, and \( c \) is the light velocity in vacuum, \( n_0 \) is the linear refractive index of the material and \( \lambda \) is the laser wavelength. The absolute value of \( \chi^{(3)} \) was calculated from the equation,

\[
|\chi^{(3)}| = \sqrt{\text{Re}(\chi^{(3)})^2 + \text{Im}(\chi^{(3)})^2}
\]

(3.18)
3.5.1.3 Experimental set-up

The transmittance of the sample through the aperture is monitored in the far field as a function of the position $Z$, of the nonlinear sample in the vicinity of the linear optics focal position. The required scan range in an experiment depends on the beam parameters and the sample thickness $L$. A critical parameter is the diffraction length, $Z_0$, of the focused beam defined as $\pi w_0^2/\lambda$ for a Gaussian beam where $w_0$ is the focal spot size (half-width at the $1/e^2$ maximum in the irradiance). For “thin” samples (i.e. $L \leq Z_0$), although all the information is theoretically contained within a scan range of $\pm Z_0$, it is preferable to scan the sample for $\approx \pm 5Z_0$ or more. This requirement, simplifies data interpretation when the sample’s surface roughness or optical beam imperfections introduce background “noise” into the measurement system. In many practical cases where considerable laser power fluctuations may occur during the scan, a reference detector can be used to monitor and normalize the transmittance. To eliminate the possible noise due to spatial beam fluctuations, this reference arm can be further modified to include a lens and an aperture identical to those in the nonlinear arm. The position of the aperture is rather arbitrary as long as its distance from the focus, $d >> Z_0$. Typical values range from $20Z_0$ to $100Z_0$. The size of the aperture is signified by its transmittance, $S$, in the linear regime, i.e. when the sample has been placed far away from the focus.

![Schematic diagram of the experimental arrangement for the Z-scan](image)

Figure 3.5 Schematic diagram of the experimental arrangement for the Z-scan
There are two different geometries in Z-scan technique. The geometry in which a finite aperture is kept before the detector is known as a closed aperture Z-scan, and (ii) the geometry in which the aperture is replaced by a convex lens to focus all the transmitted light into the detector is referred to as an open aperture Z-scan. A closed aperture Z-scan experiment is used to estimate the refractive nonlinearity while an open-aperture Z-scan experiment gives the estimate of the absorptive nonlinearity of a sample. The absorptive nonlinearity can be due to either (i) saturable absorption (SA), in which the absorption coefficient decreases resulting in the transmittance increase with increase in the input laser intensity, and (ii) reverse saturable absorption (RSA), in which the absorption coefficient increases resulting in the transmittance decrease with increase in the input laser intensity. Excited state absorption (ESA) and two-photon absorption (TPA) phenomena fall under the mechanism of RSA. The transmittance in the closed aperture is affected by both nonlinear refraction and absorption. The peak-to-valley normalized transmittance for the closed aperture z-scan data after dividing it by the corresponding open aperture z-scan data (to account for the nonlinear absorption in the medium), gives the contribution of pure refractive nonlinearity.

In Figure 3.4, describing the Z-scan, a purely refractive nonlinearity was considered assuming that no absorptive nonlinearities (such as multiphoton or saturation of absorption) are present. Qualitatively, multiphoton absorption suppresses the peak and enhances the valley, while saturation of absorption produces the opposite effect. The sensitivity to nonlinear refraction is entirely due to the aperture and removal of the aperture completely eliminates the effect. However, in this case, the Z-scan will still be sensitive to nonlinear absorption. Nonlinear absorption coefficients can be extracted from such open aperture experiments. S=1 case corresponds to
collecting all the transmitted light and therefore is insensitive to any nonlinear beam distortion due to nonlinear refraction.

One of the attractive features of the Z-scan technique is the ease and simplicity by which the nonlinear optical coefficients can be determined with a high degree of accuracy. Accurate determinations of the nonlinear coefficients such as \( n_2 \) or \( \beta \) depends on the laser source are characterized in terms of its temporal and spatial profiles, power or energy content and stability.

Once a specific type of nonlinearity is assumed (e.g. an ultra fast \( \chi^{(3)} \) response), a Z-scan can be rigorously modeled for any beam shape and sample thickness by solving the appropriate Maxwell’s equations. However, a number of valid assumptions and approximations will lead to simple analytical expressions, making data analysis easy yet precise. Beside from the usual SVEA (slowly varying envelope approximation), a major simplification results when we assume that the nonlinear sample is “thin” so that neither diffraction nor nonlinear refraction cause any change of beam profile within the nonlinear sample. This implies that \( L \ll Z_0 \) and \( L \ll Z_0/\Delta \Phi_0 \) respectively where \( \Delta \Phi_0 \) is the maximum nonlinearly-induced phase distortion. The latter requirement assures “external self-action” and simply states that the effective focal length of the induced nonlinear lens in the sample should be much smaller than the sample thickness itself. In most experiments using the Z-scan technique we find that this second criterion is automatically met since \( \Delta \Phi_0 \) is small.

In the present study, the Z-scan technique is used to evaluate the nonlinear refractive index \( n_2 \), nonlinear absorption coefficient \( \beta \) and third-order nonlinear optical susceptibility \( \chi^{(3)} \) for the chosen organic dyes.
Z-scan measurements are carried out using a diode-pumped Nd:YAG laser (532nm) of power 50mW (Coherent Compass™ 215M-50) and He-Ne (Research Electro Optics 3995) laser of wavelength 633nm (Research Electro Optics 3995) and power 17mW depending on the absorption wavelength of the organic dye used.

3.5.2 Optical Limiting

Optical limiting is a nonlinear optical process in which the transmittance of a material decreases with increased incident light intensity. Optical limiters are one of the most important types of devices used to control the amplitude of high intensity optical pulses. These device works due to intrinsic properties of the material used for their fabrication. An ideal optical limiter has a high linear transmittance at low input intensity, but above the threshold intensity its transmittance becomes constant.

3.5.2.1 Experimental set-up for Optical Limiting

The limiting effects of the dyes were studied by using a diode-pumped 50mW Nd:YAG CW laser at 532 nm and 17 mW He-Ne laser at 632 nm. The experimental set-up for the demonstration of optical limiting is shown in Figure 3.6. A 1mm quartz cuvette containing nonlinear material (dye solution) is kept at the position where the transmitted intensity shows a valley in the closed aperture Z-scan curve. A variable beam splitter (VBS) was used to vary the input power. An aperture A of variable diameter is used to control the cross-section of the beam coming out of the sample cuvette. This beam is then made to fall on the photo-detector (PD).
Figure 3.6 Experimental setup for measuring limiting effect.

The input laser intensity is varied systematically and the corresponding output intensity values were measured by the photodetector. The transmitted output intensity is found to vary linearly with the incident input intensity at very low input intensities, but starts to deviate at high incident intensities. After a certain threshold value the samples start defocusing the beam, resulting in a greater part of the beam cross-section being cut off by the aperture. Thus the transmittance recorded by the photodetector remained reasonably constant showing a plateau region.