Module 4 : Third order nonlinear optical processes

Lecture 26 : Third-order nonlinearity measurement techniques: Z-Scan

Objectives

In this lecture you will learn the following

- Theory of Z-scan technique for the measurement of third order nonlinear susceptibility $\chi^{(3)}$
- Its experimental details and
- Its merits and demerits.

Third-order nonlinearity measurement techniques: Z-Scan

Third order nonlinearity $\chi^{(3)}$ gives rise to multitude of four-wave-mixing phenomena which are of great technological interest. For example, $\chi^{(3)}(-\omega;\omega,-\omega,\omega)$ is responsible for intensity dependent refraction and intensity dependent absorption phenomena which play key roles in the development of photonic devices for all-optical signal processing. Therefore, the measurement of third order nonlinearity is of crucial importance. Further, the frequency dispersion study of the third-order susceptibility can shed light on the electronic structure of materials since it depends strongly on the internal atomic and molecular resonances.

The prescription for its measurement is contained in the very first sentence of lecture1. That is, to measure the response of a material system for a given strength of light-field stimulus in any of the nonlinear optical processes. However, the techniques based on some specific nonlinear optical processes enjoy greater merit and advantage in terms of the ease and the information that are available from the experiment. Some of the techniques that have gained greater popularity for the above said consideration are third harmonic generation (THG), degenerate four-wave mixing (DFWM), optical Kerr effect (OKE) and Z-scan. Being a parametric process, THG provides information purely on the electronic nonlinearity. However, errors can creep into the measurement if the nonlinear response generated at the interfaces is not handled carefully. In yet another technique, DFWM, measurement of $\chi^{(3)}$ is usually performed in comparative mode and is thus extremely fast and accurate. However, being a three-beam technique, it involves tedious spatial and temporal alignment of pulses. Further, it is difficult to measure real and imaginary parts of $\chi^{(3)}$ and discern the origins of nonlinearity.

Yet it is a very powerful technique to study the excitation dynamics and time resolved processes under resonant excitation. On the other hand, Z-scan being a single beam technique is extremely simple to implement and provides information on both the real and imaginary parts as well as sign of the nonlinearity. We will discuss this technique in detail.

Z-scan

Based on the principles of spatial beam distortions, Sheik-Bahae *et al*. proposed a single beam experimental technique, known as Z-scan, in 1989 [1]. The name Z-scan stems from the fact that in this technique, the sample is scanned along the direction of propagation of the incident laser beam, which is conventionally taken as Z-axis. The incident Gaussian laser beam is focused by a lens and the sample is scanned along the waist of the beam using a motorized translation stage. The transmittance of the nonlinear medium is measured through a finite aperture placed in the far-field as a function of the sample position (Z). As the sample is translated along the focused Gaussian beam, it experiences different incident field strength at different Z-positions. The nonlinear refraction in the sample arising from the third-order nonlinearity manifests itself as intensity dependent convergence or divergence of the transmitted beam (self-focusing/defocusing). Therefore, the light passing through an aperture placed in a far field will vary with the sample position. Consequently, the aperture transmittance is a function of the sample position Z. In this way, this technique is basically based on the transformation of phase distortion to amplitude distortion during beam propagation.

The sign of the nonlinear refraction is readily obtained from a Z-scan signature. An increase in transmittance in the pre-focal region followed by a decrease in the post-focal region (peak-valley) in the Z-scan signature denotes a negative nonlinear refraction, whereas a valley-peak configuration denotes a positive nonlinearity.

Removal of the aperture, i.e., collecting all the light on detector, which is referred to as an open-aperture Z-scan, will result in a flat response for a purely refractive nonlinearity. It is because the sensitivity to nonlinear refraction is entirely due to the aperture, and its removal completely eliminates the effect. However, if nonlinear absorption is also present, then it will be reflected as a transmission variation in the open aperture scan. Multi-photon absorption suppresses the peak and enhances the valley while saturation of absorption produces the opposite effect in the closed-aperture Z-scan. Thus, apart from the magnitude of the nonlinearity (both real and imaginary parts), this technique provides a direct measurement of the sign of nonlinearity also. The sign of the nonlinearity is an important parameter for the practical realization of optical signal processing devices. This information cannot be obtained by the other commonly used techniques such as DFWM and THG.

Theory:

When matter is subjected to an intense laser radiation, it exhibits intensity dependent index of refraction given by (Lecture 20)

$$n(I) = n_0 + \Delta n(I) \qquad (26.1)$$

where n_0 is the linear refractive index and $\Delta n(I) = n_2 I$ is the intensity dependent change in the refractive index. n_2 is called the refractive index intensity coefficient and is related to the real part of $\chi^{(3)}$ by

$$\mathrm{Re}\{\chi^{(3)}\}\,(esu) = \frac{n_0^2}{4\pi^2} 10^{-6}\, n_2 \qquad (26.2)$$

where is expressed in cm^2/GW.

The imaginary part of third-order nonlinear susceptibility $\chi^{(3)}$ is related to the nonlinear absorption coefficient β as

$$\mathrm{Im}\{\chi^{(3)}\}\,(esu) = \frac{n^2 c \lambda}{48\pi^3} \beta \qquad (26.3)$$

where β is in cm/GW.

Consequently, the phase change of the intense laser beam on propagation through a distance L through the nonlinear medium will be

$$\begin{aligned} \Phi &= k\, n(I)\, L \\ &= k\,(n_0 + \Delta n(I))\, L \\ &= \Phi^{L} + \Phi^{NL} \end{aligned} \qquad (26.4)$$

where k is the propagation constant in free space.
In Eq. (26.4), the first term is the linear phase shift. The second term is the nonlinear phase shift arising from nonlinear refraction and can be written as

$$\begin{aligned} \Phi^{NL} &= k\,(\Delta n)\, L \\ &= k\, n_2\, I\, L \end{aligned} \qquad (26.5)$$

It is thus clear that due to the n_2 effect, the beam passing through a nonlinear optical medium has an additional phase shift which is intensity dependent. Consequently, after passing through a nonlinear medium, the beam wavefront is modified due to intensity dependent phase changes. In the Z-scan technique, these nonlinear phase front distortions are translated into a transmission variation of an aperture placed in the far field. Hence the transmittance of the aperture is a measure of the nonlinear phase change or, n_2, and consequently, $\chi^{(3)}$. On the other hand, in the absence of any aperture, all the light transmitted by the sample is collected by the detector which is a measure of the nonlinear absorption exhibited by the sample.

We thus need to calculate the net intensity dependent changes and hence the modified wave front after propagation through the sample positioned at any location Z. This field can then be propagated to thee aperture plane and integrated over aperture radius to get the aperture transmission. These calculations can be performed by solving the nonlinear equations for propagation inside the sample and then those for the propagation of the field in free space from the exit face of the sample to the aperture.

If the sample length is less than the confocal beam parameters and if the phase changes in the due to nonlinear interaction are not transformed into amplitude changes within the sample] then the sample is considered thin. Considering a thin sample and using SVEA, we can separate the wave equation into an equation for phase and an equation for the irradiance

$$\frac{d\Delta\phi}{dz'} = \Delta n(I)k \tag{26.6}$$

$$\frac{dI}{dz'} = -\alpha(I)I$$

where k is the propagation constant in the free space, is the propagation distance inside the sample($z'=0$ for the input face and $z'=L$ is sample length to the exit face) and $\Delta\phi$ is the additional phase acquired due to nonlinear effect. $\alpha(I)$, in general, includes the linear and nonlinear absorption terms, the specifics form for which depends on the nature of the absorptive process as an example, in case of two-photon absorption $\alpha(I)=\alpha_0+\beta I$, where α_0 is the linear absorption coefficient and β is the two-photon absorption coefficient. Obviously, for purely refractive nonlinearity $\alpha(I)=\alpha_0$.

For a given sample position Z, the incident electric field of a TEM_{00} Gaussian beam having waist radius w_0 and travelling in the $+Z$ direction at $z'=0$ is

$$E(Z,r,t,z'=0) = E_0 \frac{w_0}{w(Z)} \exp\left(-\frac{r^2}{w^2(Z)}\left(1-i\frac{Z}{Z_0}\right)\right)\exp\left(-i\tan^{-1}\left(\frac{Z}{Z_0}\right)\right)F(t)$$

where E_0 is the peak on axis($r=0$) electric field amplitude,

$$w^2(Z) = w_0^2\left(1+\frac{Z^2}{Z_0^2}\right) \tag{26.09}$$

is the beam radius at Z, w_0 being beam waist; and

$$F(t) = \operatorname{sech}\left(\frac{1.76t}{\tau}\right) \tag{26.10}$$

is the Rayleigh range,
For a Gaussian temporal pulse

$$F(t) = \exp\left(\frac{-4(\ln 2)t^2}{\tau^2}\right) \tag{26.11}$$

And for sech pulse shape

$$F(t) = \operatorname{sech}\left(\frac{1.76t}{\tau}\right) \tag{26.12}$$

Where τ is the FWHM of the pulse.
A simultaneous solution of equations (26.6) and (26.7) will give the phase shift and amplitude of the beam after propagation through the sample i.e. at the exit face of the sample located at at position Z.

For purely refractive sample, the irradiance at the exit surface follows Beer's law and is given by

$$q_0 = \beta I_0 L_{eff} \tag{26.13}$$

where I_0 is the peak on axis input intensity.
Solving for the phase shift $\Delta\phi$ at the exit surface of the sample arising from cubic nonlinearity, which simply follows the radial variation of the incident irradiance at a given position of the sample Z:

$$\Delta\phi(Z,r,t,z'=L) = \Delta\phi_R(Z,t)\exp\left(-\frac{2r^2}{w^2(Z)}\right) \tag{26.14}$$

with

$$\Delta\phi_R(Z,t) = \frac{\Delta\phi_{R0}}{1+Z^2/Z_0^2} F^2(t) \qquad (26.15)$$

where $\Delta\phi_{R0}$ is the on-axis phase shift($r=0$) at focus for a purely refractive nonlinearity and is related to intensity as

$$\Delta\phi_{R0}(t) = kn_2 I_0 L_{eff} \qquad (26.16)$$

where

$$L_{eff} = \frac{1-\exp(-\alpha_0 L)}{\alpha_0} \qquad (26.17)$$

Using the irradiance and nonlinear phase shift expressions at the exit face of the sample, we can write the complex electric field at the exit surface as

$$E_e(Z,r,t,z'=L) = E(Z,r,t,z'=0)\exp(-\alpha_0 L/2)\exp\{-i\Delta\phi(Z,r,t,z'=L)\} \qquad (26.18)$$

Where $\Delta\phi$ is the nonlinear phase distortion.

We can determine the beam profile at the aperture plane E_a by propagating the field E_e by applying the Hygen-Fresnel principle through a Hankel transformation of E_e to yield

$$E_a(z,r,t) = \frac{-ik}{d-Z}\exp\left(\frac{ikr^2}{2(d-Z)}\right)\int_0^\infty E_e(Z,r',t,z'=L)J_0\left(\frac{krr'}{d-Z}\right)\exp\left(\frac{ikr'^2}{2(d-Z)}\right)r'dr'$$

Where d is the distance between the beamwaist and the aperture plane and J_0 is the Bessel function of the zeroth order.

The normalized transmittance $T(Z)$ of the aperture can be calculated as

$$T(Z) = \frac{\int_{-\infty}^{\infty} dt \int_0^{r_a} |E_a(\Delta\phi)|^2 \, 2\pi r dr}{\int_{-\infty}^{\infty} dt \int_0^{r_a} |E_a(\Delta\phi=0)|^2 \, 2\pi r dr} \qquad (26.20)$$

Numerator gives the nonlinear aperture transmission and the denominator is the aperture transmission in the linear regime i.e. corresponding to large Z and will reduce to

$$T_L = S\int_{-\infty}^{\infty} P_i(t)dt \qquad (26.21)$$

Where P_i is the input power and S is the aperture transmittance in linear regime.

$$S = 1-\exp(-2r_a^2/w_a^2) \qquad (26.22)$$

is the aperture linear transmittance, with w_a denoting the beam radius at the aperture in the linear regime.

Hence, by measuring the transmission T as afunction of sample position Z, one can deduce $\Delta\phi_{R0}$ and thus n_2 which is related to $\chi^{(3)}$.

In case, the sample exhibits nonlinear absorption, then that will be reflected in the open aperture scan while closed aperture scan records coupled refractive and absorptive parts. Therefore, one has to deduce the nonlinear absorption coefficient from open aperture scan, and then using closed aperture, the refractive and absorptive nonlinearity can be separated out. For example, if the nonlinear absorption is of two-photon in nature, then the absorption coefficient is given by

$$\alpha(I) = \alpha_0 + \beta I \qquad (26.23)$$

where β is the two-photon absorption coefficient. Now the coupled equations. (26.6) and (26.7) respectively yield the phase shift and irradiance distribution at the exit surface of the sample as

$$I_e(Z,r,t,z'=L) = \frac{I(Z,r,t,z'=0)e^{-\alpha_0 L}}{1+q(Z,r,tz'=0)} \qquad (26.24)$$

for the intensity and

$$\Delta\phi(Z,r,t,z'=L) = \frac{\Delta\phi_{R0}}{q_0}\ln[1+q(Z,r,t,z'=0)] \qquad (26.25)$$

as the phase change introduced due to the *n*2 effect in propagation through the sample.
In the above equation

$$q(Z,r,t,z'=0) = \frac{q_0}{1+(Z/Z_0)^2}\exp\left(\frac{-2r^2}{w^2(Z)}\right)F^2(t) \qquad (26.26)$$

where $q_0 = \beta I_0 L_{eff}$ is a parameter which is the measure of the strength of the two-photon absorption. Note that in equation(26.25), the refractive and absorptive parts are couple by $\frac{\Delta\phi_{R0}}{q_0}$. In the limit of negligible two-photon absorption, this equation reduces to the phase change for purely refractive nonlinearity.

Combining Eqs. (26.24) and (26.25), the complex electric field at the exit surface of the sample is given as

$$E_e(Z,r,t,z'=L) = E(Z,r,t,z'=L)e^{-\alpha_0 L/2}(1+q(Z,r,t,z'=L))^{(ikn_2/\beta-1/2)} \qquad (26.27)$$

Now that the field at the exit face of the sample is known, we can calculate the field at the aperture plane by using equation (26.19) and calculate the aperture transmittance.

As noted earlier that the absorptive and refractive nonlinearities are coupled. In order to separate these, one uses open aperture information, which records only the absorptive part. Since the beam is fully collected this time, integration over intensity profile spatially and temporally will give the nonlinear transmission of the open aperture. In case of two-photon absorption discussed above, the open aperture transmittance of, for a Gaussian-spatial and temporal input is

$$T(Z,S=1) = \frac{1}{\sqrt{\pi}q(Z,r=0,t=0)}\int_{-\infty}^{\infty}\ln\left[1+q\ (Z,r=0,t=0)e^{-\tau^2}\right]d\tau \qquad (26.28)$$

For $|q_0| < 1$, this energy transmittance can be expressed in terms of the peak irradiance in a summation form as
Thus, once an open aperture (S = 1) Z-scan is performed, the nonlinear absorption coefficient β can be estimated. With β known, the Z-scan with aperture in place (S < 1) can be used to extract the nonlinear refractive index coefficient n_2.

Similarly in case of other nonlinearities (e.g. saturation of absorption, excited state absorption, free carrier absorption etc.), we can deduce the refractive and absorptive contribution to nonlinearity by appropriately modifying the equations for phase shift and beam attenuation, and solving for nonlinear phase shift and field amplitude at the exit surface of the sample.

Sheik-Bahae *et al*. [1] have shown that the above tedious calculations can be simplified provided the phase change induced by the nonlinear medium is small. For $|\Delta\phi| < \pi$, the peak-valley difference in transmission in case of purely refractive nonlinearity is given by

$$T_{p-v} = 0.405(1-S)^{0.25}kn_2 I_0 L_{eff} \qquad (26.29)$$

which is valid within 3%. The peak-valley separation along Z-axis in this case is related to Rayleigh range through

$$Z_{p-v} = 1.7Z_0 \qquad (26.30)$$

Therefore, we can get quick estimate of nonlinearity by just measuring the peak-valley difference in transmission.

Z-scan simulations

Figure 26.1 (a) and 26.1 (b) show open aperture Z-scan transmission for positive and negative β respectively. In case of positive β (two-photon absorption, excited state absorption or free-carrier absorption), we get a valley at the beam waist position of the focused beam while negative β (saturation of absorption) results in a peak at the same position. Thus, the sign of absorptive nonlinearity is readily obtained from the open aperture Z-scan signature. If the sample does not exhibit any nonlinear absorption, then no transmittance variation will be recorded in the open-aperture scan.

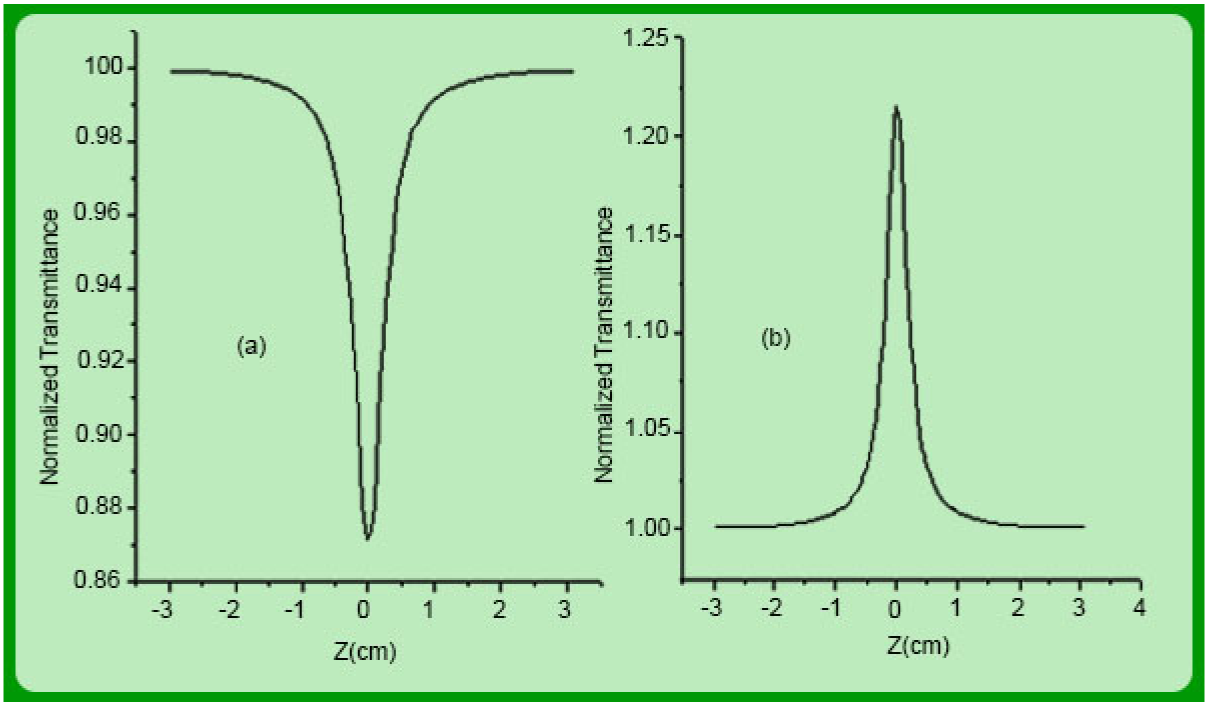

Figure 26.1: Open aperture simulation for (a) positive β and (b) negative

Figure 26.2 (a) shows Z-scan simulation for purely refractive positive and negative nonlinearities corresponding to a phase change $\Delta\phi=\pm 0.5$ and an aperture parameter $S=0.2$.

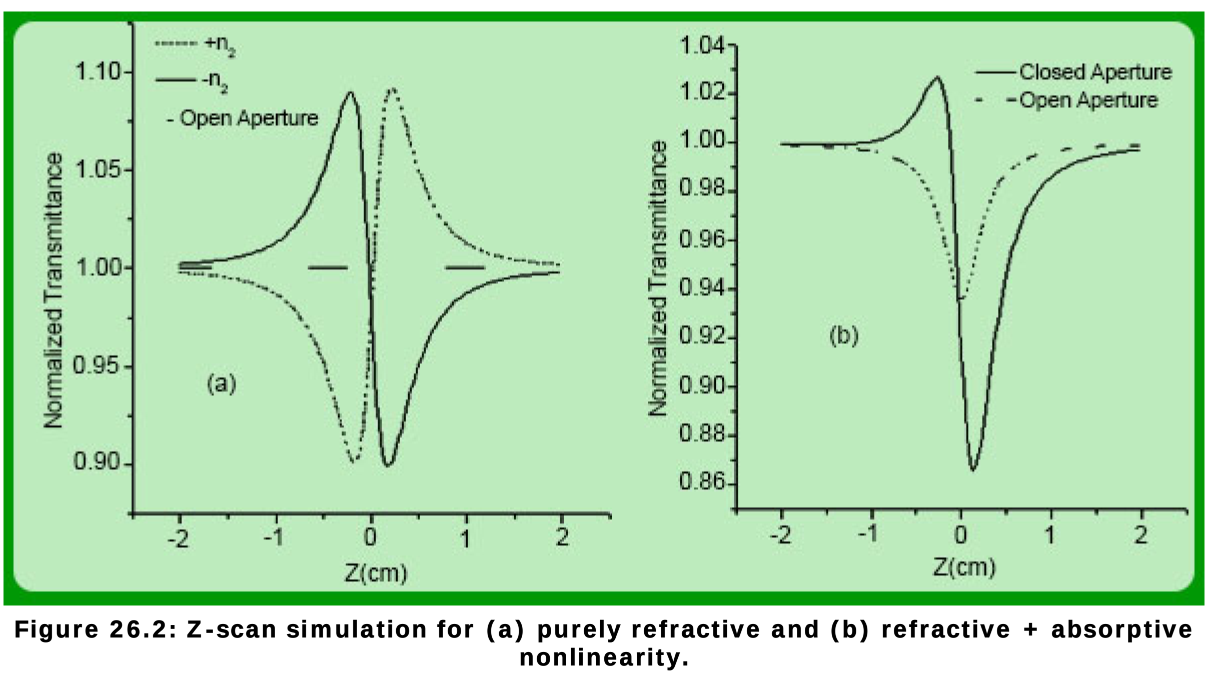

Figure 26.2: Z-scan simulation for (a) purely refractive and (b) refractive + absorptive nonlinearity.

As explained earlier, negative nonlinear refraction results in increase in transmittance in the pre-focal region followed by decrease in the post-focal region (peak-valley) while positive nonlinear refraction exhibits the opposite behavior. Also shown is the open aperture simulation in which there is no transmittance variation in case of purely refractive nonlinearity.

When nonlinear absorption is also present, then in the closed aperture signature, the peak gets suppressed while the valley gets enhanced for positive absorptive nonlinearity. The opposite behaviour will be exhibited in case of negative absorptive nonlinearity. Figure 26.2 (b) shows simulation for a negative refractive nonlinearity and positive absorptive nonlinearity for a phase change of $\Delta\phi = -0.5$ and two-photon absorption parameter $q_0 = 0.2$.

Experimental setup

The Z-scan experimental setup is shown in Figure 26.3. A TEM_{00} laser beam coming from Ti:sapphire oscillator was focused on to the sample using a lens of focal length, say, 10 cm to a beam waist of 25 μm. The sample is translated in the focal region of the lens by a stepper motor controlled translation stage along the direction of beam propagation. The transmitted beam from the sample is split into two arms by a beam splitter and collected at two photodiodes D_1 and D_2. The detector D_2 is partially closed with an aperture and serves as the closed aperture while D_1 acts as an open aperture detector. Outputs from the detectors are connected to data acquisition system. Typical open and closed aperture Z-scans of a sample are ashown in figure 26.4 along with theoretical fits.

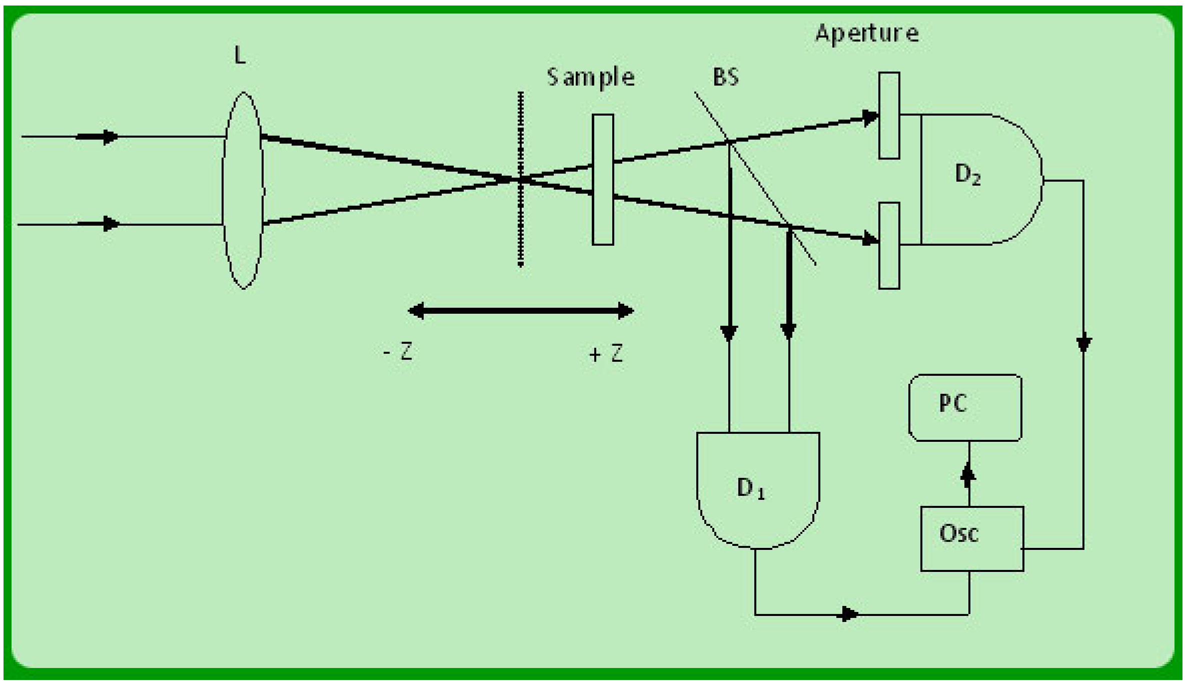

Figure 26.3: Z-scan experimental setup. D1, D2 – Detectors, L – Lens, BS – Beam Splitter, Osc – Digital Oscilloscope, PC – Microcomputer.

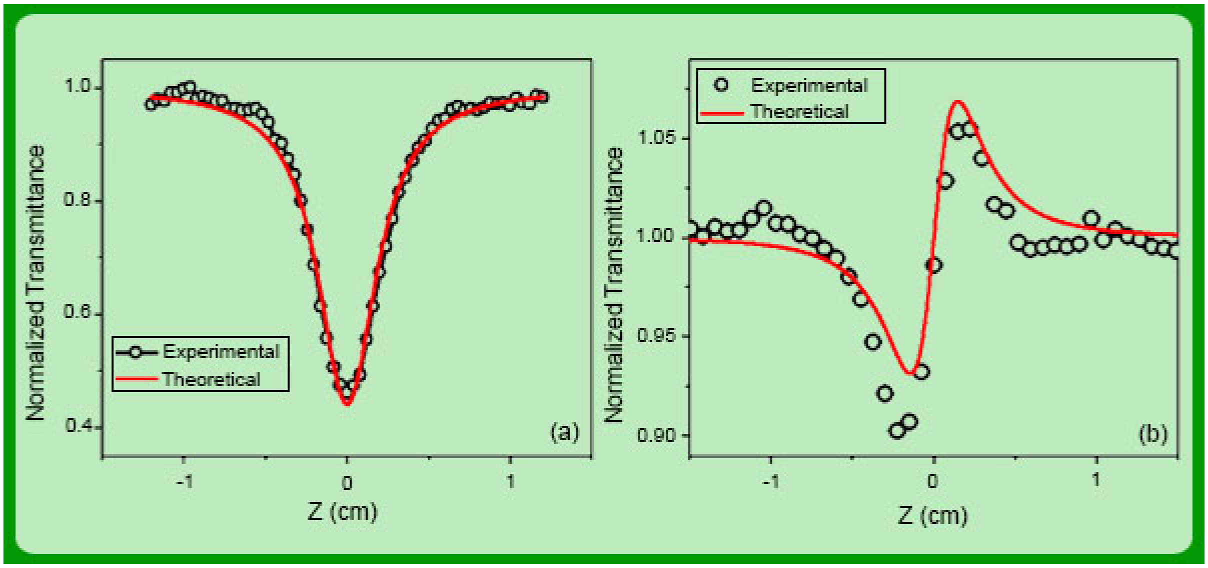

Figure 26.4:Typical Z-scan of a sample (a) Open aperture(b) Closed aperture.

Merits of Z-scan:

Following are the advantages of Z-scan technique:

a. A simple single-beam technique with no tedius alignment.
b. It yields magnitude as well as sign of $\chi^{(3)}$.
c. Allows separation of the real and imaginary parts of the.

Demerits of Z-scan:

a. Since the whole principle is based on the wavefront modification due to nonlinear effect, anomalies like sample scatter and surface inhomogeneity can cause artifact to Z-scan signatures. Therefore, good surface quality samples are required for doing experiments with this technique.
b. Can not differentiate between different mechanisms of nonlinearity.

Sources of errors and precautions:

Various sources of errors and precautions needed to be taken are described below:

a. Sample translation should be parallel to beam axis
b. The analysis presented above is valid only if the sample thickness is less than the Rayleigh range. Therefore, one needs to use appropriate focal length lens such that Rayleigh range is more than the sample thickness.
c. Beam spot size of the focused Gaussian beam should be measured carefully.
d. Aperture size should be chosen carefully. Too large an aperture lowers the sensitivity and too small an aperture lower the transmission below the detection with good S/N ratio by attenuating. As a rule of thumb, an aperture size of 10-20% (S = 0.1-0.2) is an optimum choice.

References:

1. Sheik-Bahae, A. A. Said and E. W. Van Stryland, Opt. Lett. **14**, 955 (1989).

Recap

In this lecture we have discussed

- Theory of Z-scan technique for the measurement of third order nonlinear susceptibility $\chi^{(3)}$.
- Corresponding experimental details.
- sources of errors and precautions and
- Its merits and demerits.