National Institute of Science Education and Research

Semester Project

Z-Scan

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Chapter 1

Introduction

Z-scan has emerged as a widely accepted experiment in non-linear optics community,for determining the non-linear behaviour of a medium by measure both nonlinear absorption coefficient(α) and nonlinear refraction coefficient (n_2) in solids,liquids and liquid solutions.
The technique utilizes the change in the divergence of the beam after passing through the non-linear medium. As this is an intensity dependent refractive index, the beams radius of curvature changes as the intensity is increased or decreased. By measuring this change in divergence, it is possible to back calculate the non-linear refractive index. Keeping the overall power of beam constant, the sample is placed in the beam and translated through the focal region where the beams intensity distribution changes because the beam size changes. As the sample is translated through the focus in z-direction, hence its name z-scan.
We use a pinhole to mask off all the radiation of the laser, except the center in closed-aperture z-scan. When we translate the medium, divergence of the beam takes and hence the power at the center varies. This way by translating the sample along the focal region of the convex lens we can plot a graph between intensity and distance.

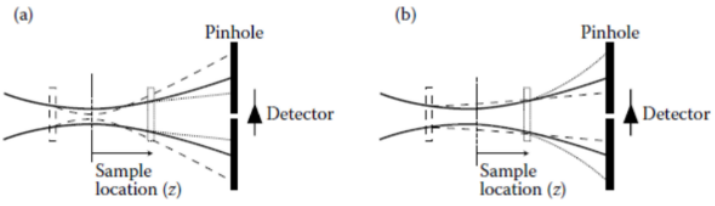


Fig 1:Beam divergence for various sample locations for (a) positive n_2 (b) negative n_2, z is measured with respect to original beam waist location

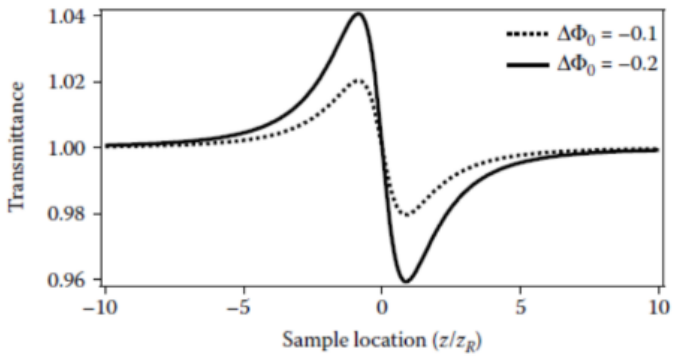


Fig 2:Transmittance as a function of sample position for a material with a negative nonlinearity. $\Delta\phi_0$ is the on-axis nonlinear phase shift at the focus

Let us consider a material with positive non-linear refractive index, n_2<0. The Gaussian beam has an intensity distribution that changes across its cross section and the presence of the sample is akin to placing a thin lens with a positive focal length at the sample location. As shown in Fig. 1a, on the side of focus where the beam is converging, the non-linear lens shortens the beams waist position to a negative z value. As the beam passes through the shifted focus, it diverges at a greater diffraction angle, so the beams power is spread over a wider area and the intensity of beam passing through the pinhole decreases. When the sample is on the positive z side, where the beam is diverging, the non-linear lens reduces the beams angle of divergence, thereby increasing the power passing through the pinhole. The effects are exactly opposite for n2 ¡ 0. The effective focal length generated depends on peak intensity so that when the sample is located far from focus non-linear effects are negligible. Fig. 2 shows the transmittance vs sample location plot of z-scan for a material with n_2<0.

Chapter 2

Theory

2.1 Origin of non-linearity

The origin of the non-linearity can be traced back to the interaction of the electrons of the host material with the light/energy. In the dielectric media of classical nature, the electrons are held in a potential well whose shape is quadratic in nature. When incident field strength increases, electron is pushed further from equilibrium, which corresponds to departure from pure quadratic. Classically, an electron experiences a restoring force and as force is defined as the grad of potential energy, we get a linear force.
Electrons held in quadratic potential thus experience a linear restoring force. This is identical to mass on spring harmonic oscillator. As higher order terms in the potential are added, the restoring force is modelled as a mass on a non-linear spring, the system is called an anharmonic oscillator. Classical anharmonic response is a perturbation to the linear harmonic oscillator solution.

2.2 Non-linear optics

In the conventional way the polarization(P) of a material depends on the electric field (E) in the following way

$$P = \epsilon_0 \chi E \tag{2.1}$$

Where χ is the linear susceptibility. But in non-linear optics the susceptibility is electric field dependent as follows.

$$\chi = \chi^{(1)} + \chi^{(2)} E + \chi^{(3)} E^2 .. \tag{2.2}$$

and

$$P = \epsilon_0 [\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 ..] \tag{2.3}$$

Where $\chi^{(i)}$ represents various orders of the susceptibility.

$$|\chi^{(3)}| << |\chi^{(2)}| << |\chi^{(2)}| \tag{2.4}$$

By writing all quantities in Eq.2.3 we impose a specific phase relationship between electric field and total polarization. Writing as a purely real number indicates that an electric field induces a polarization that is exactly in phase with it. Assuming susceptibilities in expansion are real is equivalent to assuming that the polarization response to the incident field is instantaneous.
In non-centrosymmetric media, second order displacement term dominates over the third order displacement term. So, we single out the third order term by considering centrosymmetric media.
Non-linear refractive index is commonly written as

$$n = n_0 + n_2 I, \quad n_2 = \frac{3}{2n_0^2 \epsilon_0 c} \chi^{(3)}$$

and I = $\frac{1}{2} n_0 \epsilon_0 c E^2$
We use Gaussian beam with,

$$E(r,z,t) = E_0(t) \frac{w_0}{w(z)} \exp\left[-\frac{r^2}{w^2(z)} - \frac{ikr^2}{2R(z)} \right] \exp[-i\phi(z,t)]$$

w is radius of beam at z, E_0 is electric field at beam waist.
For sample length, we consider there is no change in beam diameter within the sample. Phase shift $\Delta\phi$,

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

Now the phase shift is Gaussian function of transverse coordinate with respect to centre of the beam.

$$\Delta\phi_0 = k n_2 I_0(t) L_{eff}$$

$$I_0 = \frac{P_{avg}}{(\text{repition rate})(\text{pulse width})(\pi w_0^2)}$$

I_0 is the irradiance at focus, L_{eff} = (1 - exp[$-\alpha L$])/α is effective propagation length inside sample.

2.2.1 Multi photon absorption

Two photon absorption is absorption of two photons of identical frequencies in order to excite a molecule froom one state to a higher energy state. The energy difference between two state is is equal to the sum of the photon energies. of the two photons absorbed. It is a second order process, weaker than the linear absorption at low intensities. Hence, it is a non-linear optical process and can dominate over linear absorption at high intensities.
In these experiments we do not consider the nonlinear absorption coefficient of material.material. Z-scan method is also used to determine the coefficient of nonlinear absorption β. The whole absorption is defined

$$\alpha = \alpha_0 + \beta I$$

Where α_0 is the linear absorption coefficient and I is the intensity of the beam. In the above picture describing the Z-scan, multiphoton absorption suppresses the peak and enhances the valley, while saturation 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 2-scan will still be sensitive to nonlinear absorption. Nonlinear absorption coefficients can be extracted from such open aperture experiments.

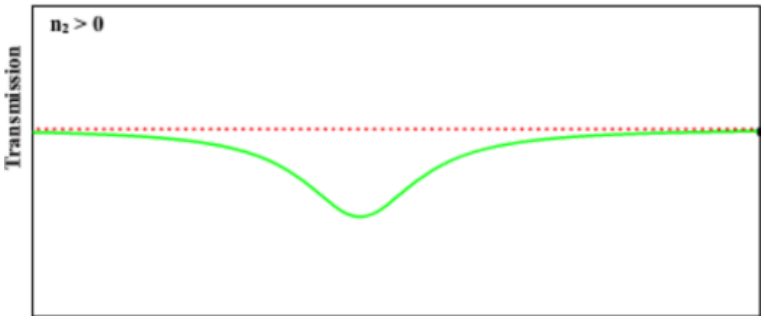


Fig 4:-Transmittance with respect to z measured using a open aperture detector.

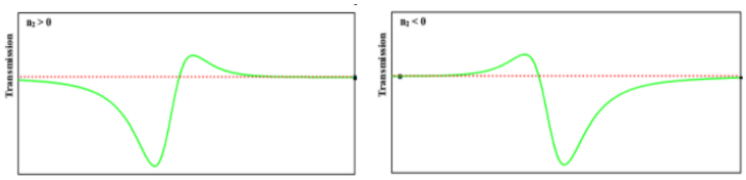


Fig 5:-Transmittance with respect to z measured using a closed aperture detector with non-linear absorption in mind. The high intensity peak to the right of z=0 has been suppressed due to high absorption at large intensity.

2.2.2 Theoretical Calculation

The beam exiting the sample has complex electric field which has phase distortion, we use Gaussian decomposition method in which the exiting Gaussian beam is decomposed through Taylor series expansion of non-linear phase term. At aperture we get electric field E_a(r,t) as a function of $\Delta\phi_0$. Spatially integrating $|E_a(r,t)|^2$ upto aperture radius r_a we get transmitted power $P_T(\Delta\phi_0(t))$. Normalized transmittance T(z)is given by

$$T(z) = \frac{\int_{-\infty}^{\infty} P_T(\Delta\phi_0(t))dt}{S\int_{-\infty}^{\infty} P_i(t)dt}$$

P_i(t) is instantaneous input power within the sample. $S = 1 - \exp(-2r_a^2/w_a^2)$. Most important parameter is ΔT_{p-v}, difference between the highest value and

lowest value of the transmittance. Based on numerical fitting we get a relation between T_{p-v} and $\Delta\phi$

$$\Delta T_{p-v} \simeq 0.406(1-S)^{0.25}|\Delta\phi_0| \tag{2.5}$$

We have formula to find the transmittance, without the use of S value.

For closed aperture,

$$T(z,\Delta\phi) = 1 + \frac{4\Delta\Phi_0 x}{(x^2+1)(x^2+9)} - \frac{2(x^2+3)\Delta\psi_0}{(x^2+1)(x^2+3)} \tag{2.6}$$

For very small opening, Open aperture,

$$T(z,S=1) = 1 - \frac{\beta L_0 L_{eff}}{2^{3/2}(1+x^2)} \tag{2.7}$$

$$x = \frac{z_0}{z}, \quad \Delta\Phi_0 = kn_2 I_0 L_{eff}, \Delta\psi_0 = \beta I_0 L_{eff}/2$$

2.3 Experimental set-up and procedure

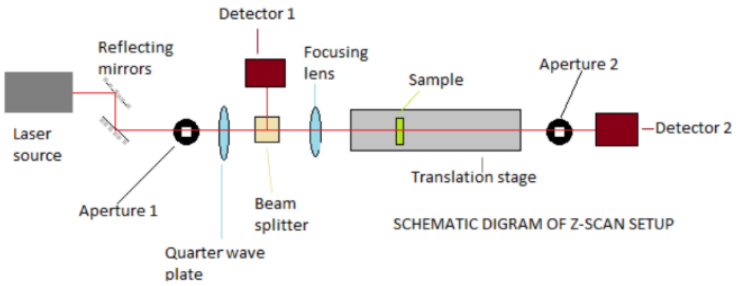


Fig 3:z-scan setup

Chapter 3

calculation and analysis

toluene

Here, we have
L_{eff}(width of the sample) = 1 mm,
Repetition rate = 80KHz,
pulse width 1.75 ns
wavelength = 532 nm

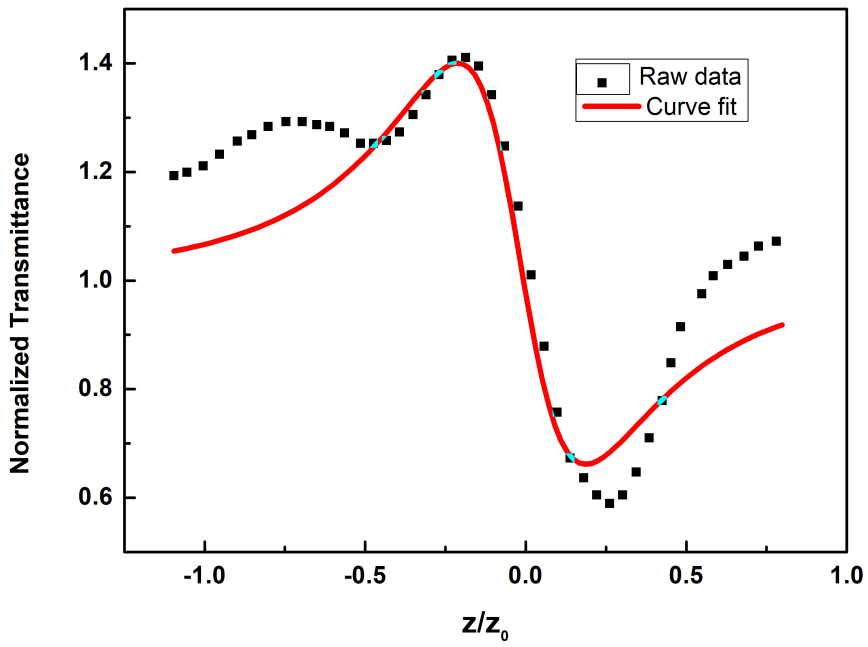
closed aperture

from the graph we can find $\Delta\Psi$ & $\Delta\Phi$ which further used to find the nonlinear refractive index(n_2) and the nonlinear absorption coefficient(β). for Average power, P_{avg} = 120 mW,
Refractive index,

$$n_2 = -2.732 * 10^{-15} \pm 1.925 * 10^{-17} m^2/W$$

Absorption coefficient,

$$\beta = 1.382 * 10^{-8} \pm 7.03 * 10^{-10} m/W$$

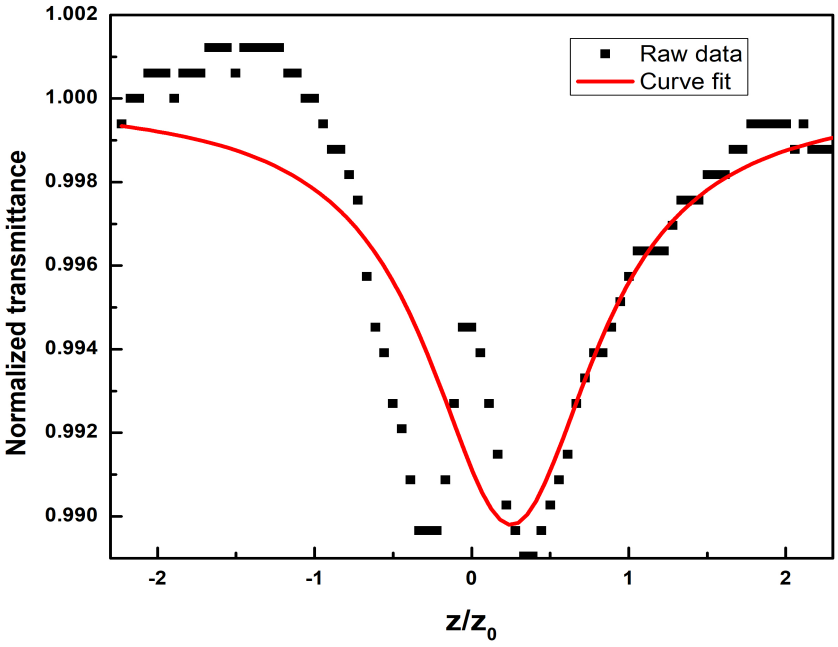


figure*Normalized transmittance plotted against* $\frac{z}{z_0}$ *of toluene sample for closed aperture configuration and its curve fit*

open aperture

open aperture set up can be used to find β alone, for Average power, P_{avg} =130 mW
Absorption coefficient,

$$\beta = 2.108 * 10^{-8} \pm 0.565 * 10^{-9} m/W$$



figure*Normalized transmittance plotted against* $\frac{z}{z_0}$ *of toluene sample for open aperture configuration and its curve fit*

Cs2

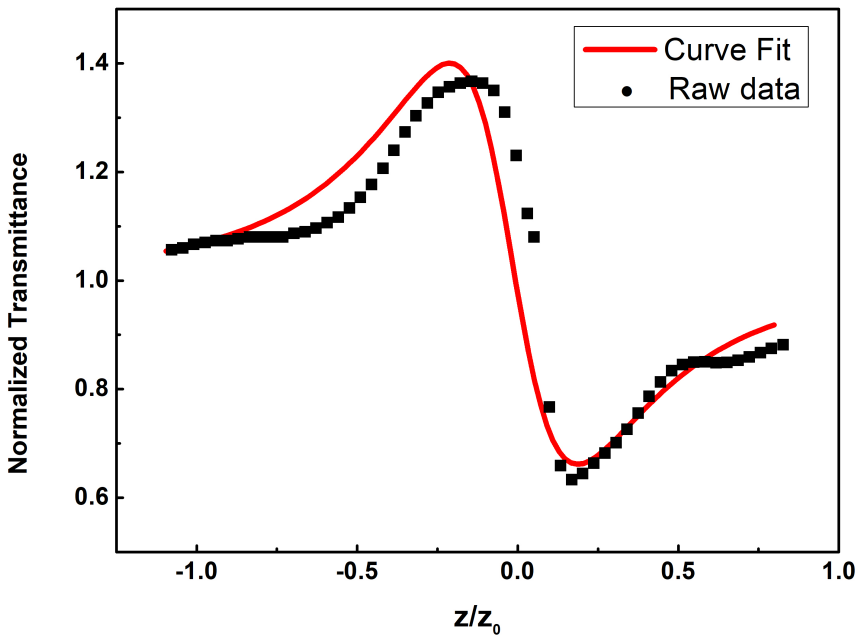
closed aperture

for Average power, $P_{avg} = 120$ mW,
Refractive index,

$$n_2 = -7.598 * 10^{-15} \pm 1.452 * 10^{-18} m^2/W$$

Absorption coefficient,

$$\beta = 9.53 * 10^{-8} \pm 7.03 * 10^{-9} m/W$$

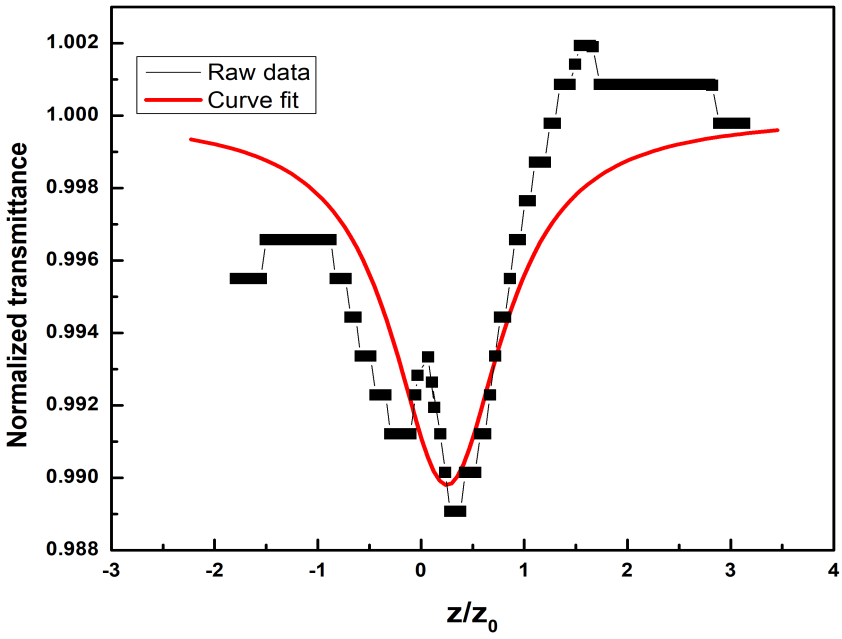


figure*Normalized transmittance plotted against* $\frac{z}{z_0}$ *of Cs2 sample for closed aperture configuration and its curve fit*

open aperture

for Average power, P_{avg} =130 mW
Absorption coefficient,

$$\beta = 9.837 * 10^{-8} \pm 2.56 * 10^{-8} m/W$$



figure*Normalized transmittance plotted against* $\frac{z}{z_0}$ *of Cs2 sample for open aperture configuration and its least square fit*

Chapter 4

Conclusion

We used the Z-scan experimental configuration to obtain nonlinear refractive index of a standard sample such as Toulene,CS2 and DMSO. The sign and magnitude of nonlinear refractive index of all the samples were measured and compared with literature value, which matches well with estimated value. We have used equations of normalized transmittance which doesnt involve the S value which corresponds to fraction of the aperture that is open. For analysis we have used the lmfit routine of the Python which gve a ood estimate of the values by performing least square fittings. Thus, we have shown that this method provides a reliable alternative to single-shot type measurements in which a large number of averages are needed to obtain data with little experimental noise.
In the table below we tabulate our results. Here n_2 is in (m^2/W) and β is in m/W.

4.1 Precautions

As we were working with high power lasers, the operation, if not done with proper precautions, can be dangerous. It was made sure by our instructors, not to bend down to the level of the laser which could damage the eyes severely, sometimes to the extent of being irreparable. We used protective glasses during our entire stay at the laboratory. Black Aluminum plates were used to block unwanted laser radiation from reflecting to surfaces and heating them up, at some point we were working with 1064nm laser, which was invisible to the naked eye, but could have caused a heating effect, we made sure that the source was

Sample name	P(avg)	n	Δn	β	$\Delta\beta$
Toluene	120mW	2.732 x 10^{-15}	1.925 x 10^{-17}	1.328 x 10^{-8}	7.03 x 10^{-10}
CS2	130mW	7.598 x 10^{-15}	1.452 x 10^{-18}	9.53 x 10^{-8}	7.03 x 10^{-9}

appropriately sheilded during the entire experiment. The cuvette was cleaned with acetone before using it. Similar procedure was done for lenses and mirrors, but their use was minimized so as not to damage the mirrors

4.2 Sources of Error

Even though the laser was aligned properly with the lens and detector,While translating the sample it was not properly following the z line.Deviation from the Z axis could give rise to error in the intensity data. And the detector used has a time resolution ,moving fast through the translation may not give the correct data.It should be taken step wise.

References

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