P443 P444: Study of Non-Linear Optical Properties using Z-Scan

Shriman Keshri
1811150

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Abstract

In this experiment, we started with learning non linear optics basics and its uses. Then we explored the Single Beam Z-Scan Technique and saw how Z-Scan traces for different materials can be obtained. We have analyzed these traces using Python and saw how the Non-Linear Refractive Index (n2) and the Non-Linear Absorption Coefficient ( ) for some materials can be calculated. We have also examined certain future directions that we could take this experiment in.

Contents

1 Objective

- To study the basics of Non-Linear Optical Properties by using Z-scan Techniques.
- Taking measurements of transmitted power for open and closed aperture by translating the material in the z-direction.
- By fitting these data with the appropriate formulas, we will find the medium's nonlinear absorption coefficient and nonlinear refractive index.

2 Overview

The non-linear optical properties of materials have found several applications in recent times, primarily in ultra-fast optical switching and signal processing applications. In such cases, the relevant phenomena are Non-Linear Refraction (NLR) and Non-Linear Absorption (NLA). The Single Beam Z-Scan technique, is one of the principal techniques to explore these non-linear material characteristics owing to its simplicity and high sensitivity.

This technique can rapidly provide a measure of both NLR and NLA in solids as well as liquids (including solutions), yielding the sign as well as the magnitude of the non-linear refractive index and the non-linear absorption coefficient. This technique is based on the principles of spatial beam distortion. It makes use of the fact that a spatially transverse variation in LASER intensity induces non-linear optical changes in the material, giving rise to a lens-like effect, which in turn affects the beam propagation. This leads to a self-focusing or defocusing effect (self-lensing effect). This in turn causes changes in the far-field diffraction pattern, that can be utilized to obtain the relevant non-linear optical properties of materials. "Single Beam", in the name, comes from the fact that a single Gaussian laser beam in a tight focus geometry, is used to induce the non-linear effects.

3 Theory

Nonlinear Optics is a branch of optics that deals with light behavior in nonlinear media(Polarization P relates non-linearly with electric field).
While considering nonlinear optical materials, two properties of interest are the material's nonlinear refractive index and its nonlinear absorption coefficients. Both change the intensity of light non-linearly when light passes

through a medium.
The Z-Scan is based on the principle of spatial beam distortion used to measure the third-order optical nonlinearity and allows computing the contributions of nonlinear absorption and nonlinear refraction.

3.1 Nonlinear Optical Media

In a linear dielectric medium, there is a linear relation between Electric field and induced electric polarization

$$\mathbf{P} = \epsilon_0 \chi \mathbf{E}$$

where ϵ_0 is the electric permittivity of vacuum and χ is the dielectric susceptibility of the medium. In a nonlinear dielectric medium, **P** and **E** are related non-linearly.

$$\mathbf{P}(\mathbf{E}) = \epsilon_0 \left(\chi^{(1)} \mathbf{E} + \chi^{(2)} \mathbf{EE} + \chi^{(3)} \mathbf{EEE} + \ldots\ldots \right)$$

where $\chi^{(n)}$ are the higher order susceptibilities which governs the nonlinear processes. When the intensity of the light will be sufficiently high, only then these higher order polarization terms will be significant.

3.2 Nonlinear wave equation

A wave equation for propagation of light in a nonlinear medium can be derived from maxwell equations

$$\nabla^2 \mathbf{E} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}}{\partial t^2}$$

We can write **P** as sum of linear and nonlinear terms separately where $\mathbf{P}_{NL}$ is $\mathbf{P}_{NL} = \chi^{(2)} \mathbf{EE} + \chi^{(3)} \mathbf{EEE} + \ldots\ldots$ equations, $n^2 = 1 + \chi, c = \frac{1}{n}$ and velocity of light in a Now, by using the equations, $n^2 = 1 + \chi, c = \frac{1}{n}$ and velocity of light in a medium of refractive index n, v $= \frac{c}{n}$, we can write wave equation in a nonlinear medium

$$\nabla^2 \mathbf{E} - \frac{1}{v^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2}$$

This is the basic equation in the theory of nonlinear optics.

3.3 Third-order Nonlinear Optics

We generally work with centrosymmetric media (the properties of the medium which are not altered by the transformation $\mathbf{r} \to -\mathbf{r}$). Hence $\chi^{(2)}, \chi^{(4)}, \chi^{(6)}, \ldots$ terms will vanishes. and hence third order term $\chi^{(3)}$ becomes more important. Equation (2.2) will become

$$\mathbf{P}(\mathbf{E}) = \epsilon_0 \chi^{(1)} \mathbf{E} + 0 + \epsilon_0 \chi^{(3)} \mathbf{EEE}$$

where **E** is

$$\mathbf{E} = \frac{1}{2} \left(E_0 e^{ikz - i\omega t} + E_0^* e^{-ikz + i\omega t} \right)$$

Put this eq. in (2.7), we get

$$\mathbf{P}(\mathbf{E}) = \epsilon_0 \left(\chi^{(1)} + \frac{3}{4} \chi^{(3)} |E_0|^2 \right) \mathbf{E}$$

Hence $\chi_{eff} = \chi^{(1)} + \frac{3}{4} \chi^{(3)} |E_0|^2$. We have relation between refractive index n and susceptibility χ_{eff}

$$n^2 = 1 + \chi_{\text{eff}} = 1 + \chi^{(1)} + \frac{3}{4} \chi^{(3)} |E_0|^2$$
$$n^2 = n_0^2 + \frac{3}{4} \chi^{(3)} |E_0|^2 = n_0^2 \left(1 + \frac{3\chi^{(3)} |E_0|^2}{4 n_0^2} \right)$$

where we have used $n_0^2 = 1 + \chi_0$ (where n_0 is the linear component of refractive index). Now take square root both side and use binomial expansion, we get

$$n = n_0 \left(1 + \frac{3}{8 n_0^2} \chi^{(3)} |E_0|^2 \right) = n_0 + \frac{3}{8 n_0} \chi^{(3)} |E_0|^2$$

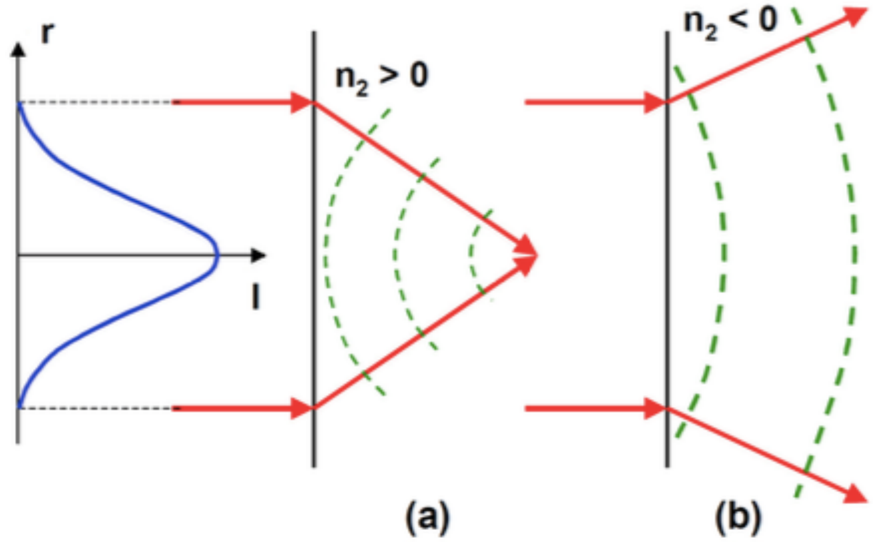
By using relation $I = \frac{1}{2} \epsilon_0 n_0 c |E_0|^2$

$$n = n_0 + \frac{3}{4} \chi^{(3)} \left(\frac{I}{\epsilon_0 n_0^2 c} \right)$$
$$n = n_0 + n_2 I$$

where $n_2 = \frac{3\chi^{(3)}}{4 \epsilon_0 n_0^2 c}$ is a nonlinear component of the refractive index. And hence we can increase the nonlinear effects by increasing the intensity of incident light. This effect is known as Optical Kerr effect, i.e. change of refractive index of a material in response to an applied electric field.

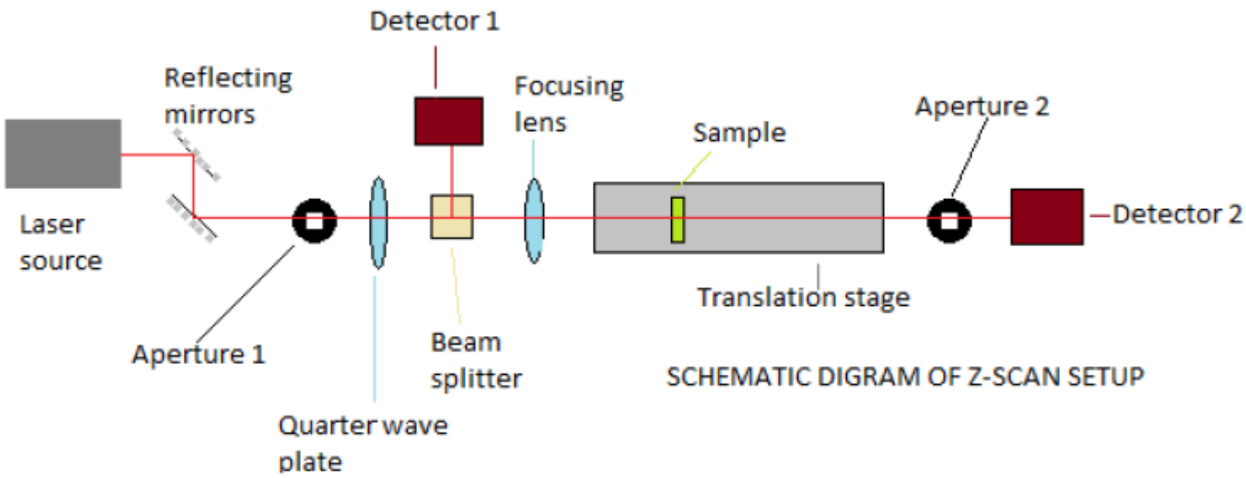
3.3.1 Self-focusing and defocusing

Self-focusing and defocusing is a non-linear optical processes induced by the change in refractive index of materials exposed to intense electromagnetic radiation. A medium whose refractive index increases with the electric field intensity acts as a focusing lens(fig a) for a Gaussian beam,while if it decreases with electric field intensity acts as defocusing lens(fig b).

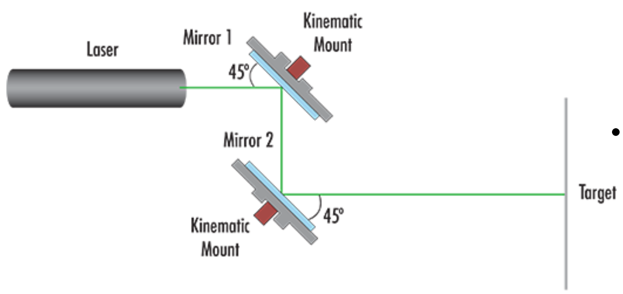


4 Z-Scan Experiment

4.1 Apparatus used and their Working



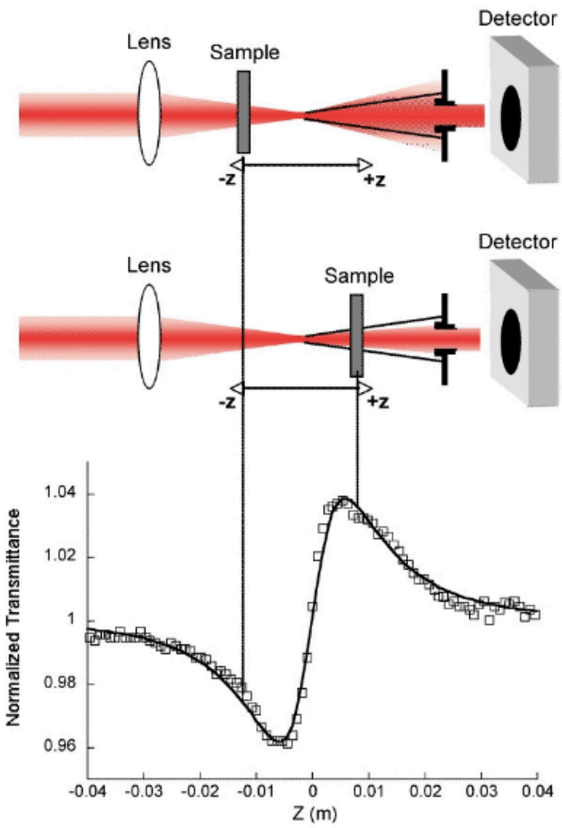
- Laser Source: We use the pulsed 532 nm laser.
- We used the two reflecting mirrors for aligning the beam by Parallel (Z-Fold) Configuration(as shown in below figure)



- Wave plate: Optical device which alter the polarization state of light wave traveling through it. There are two types of wave plate, first half wave plate which shift the polarization direction of linearly polarized light, second is quarter wave plate which converts linearly polarized light into circularly polarized light.
- Beam Splitter: Optical components used to split incident light at a designated ratio into two separate beams. There will be semi reflective coating on one of the prism. Ratio of reflection/transmission depends on coating material. There are two type of beam-splitter, first is polarizing beam splitter(divides incident unpolarized light into orthogonal polarized beams) and second is Non-polarized beam splitter(split by specific percentage that is independent of polarization).
- **Note:** We are using a combination of wave plate and beam-splitter to control the intensity of laser beam. Through half-waveplate we will bring a polarisation in the light and our beam splitter will split the beam intensity (will reflect some and pass some) as per polarisation.
- Thermal detectors: It is based on temperature change of the element through the absorption of EM radiation. Change in temperature causes change in temperature dependent property (resistance) of thermal detector, which is evaluated electrically and is measure of the absorbed energy.
- A Converging Lens (L0), with f = 15 cm, to focus the LASER light.
- A Translation Stage (TS) with wires and con- trol software. It has a stepper motor, which can run at up to 300 RPM. The end-to-end length covered by the translation stage, with the mounted sample, is 12.2 cm (˜ 12400 steps).
- A Beam Profiler to measure the Beam Waist.
- An Optical Table to assemble the setup.
- Allen Keys and bolts to hold the equipment in place.
- Multiple screens to avoid interference from other setups.

4.2 Principle

When a high intensity laser beam propagates through a material, induced refractive index changes leads to self-focusing or defocusing of the laser beam which enables to determine the third-order nonlinear optical properties of various materials. In this technique, the sample is translated along z direction through the beam waist of a focused beam by keeping the input power of beam constant. In our experiments a continuous pulse wave laser operating at 532 nm was used. The laser beam was focused using a 10 cm focal length lens. Let us consider a material with positive non-linear refractive index, $n_2 > 0$. As shown in Fig. 1a), on the side of focus where the beam is converging, the non-linear lens shortens the beam's waist position to a negative z value. As the beam passes through the shifted focus, it diverges at a greater diffraction angle, so the beam's 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 beam's angle of divergence, thereby increasing the power passing through the pinhole. The effects are exactly opposite for $n_2 < 0$.



Refraction.png

Non-Linear Refraction

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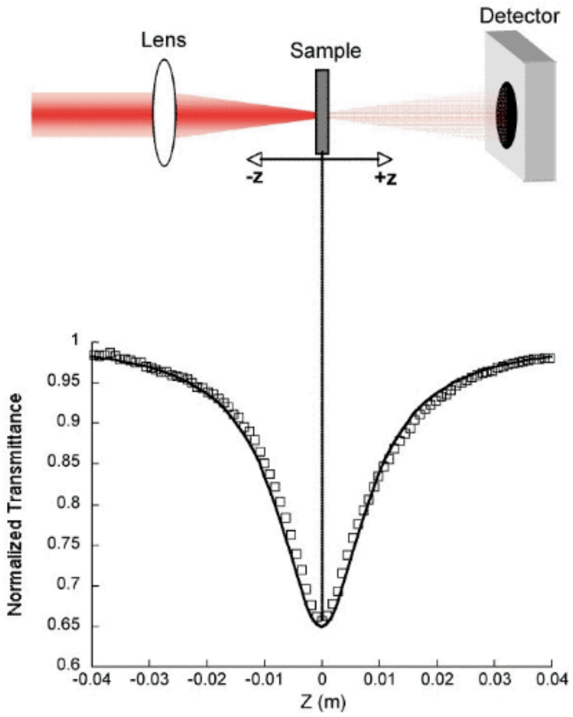
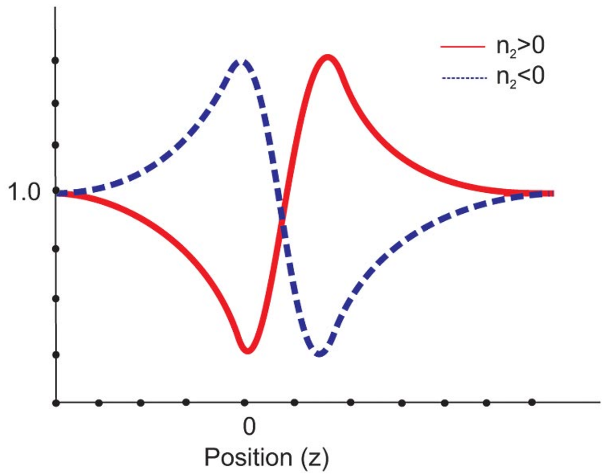


Absorption.png

Non-Linear Absorption

If the material has a positive non linearity ($n_2 > 0$), the transmittance graph has a valley first and then a peak. For the sample with $n_2 < 0$ the graph is exactly the opposite (first peak and then valley).



Determination of a sign of n_2 from transmittance graph

The aperture plays an important role in determining which values we are going to find. We use closed aperture for finding out the non-linear refractive index and open aperture to find the absorption coefficient.

4.3 Formulas to derive Nonlinear refraction and nonlinear absorption

Consider a Gaussian Beam, propagating along the z-direction with beam waist, w_0, wavelength, λ, and Rayleigh Length, $z_0 = \pi w_0^2/\lambda$:

$$E(r,z) = E_0(t)\frac{w_0}{w(z)}\exp\left(-\frac{r^2}{w(z)^2}\right)\exp\left(-ikz - ik\frac{r^2}{2R(z)}\right)\exp(-i\phi(z,t)) \tag{1}$$

Here, $k = 2\pi/\lambda$ is the Wave Number; $w(z) = w_0\left(1+x^2\right)$ represents the Beam Width, with $x = z/z_0$; and $R(z) = z\left(1+x^2\right)$ denotes the Radius of Curvature. The last term, $\exp(-i\phi(z,t))$, contains all the radially uniform phase variations. As we only wish to obtain the radial phase variations, $\Delta\phi(r)$, we can apply the Slowly Varying Envelope Approximation (SVEA) and ignore phase changes that are uniform in r. Now, let this beam be transmitted through a Kerr non-linear optical material of length, L. Then, considering a centrosymmetric medium, we can write the refractive index and absorption coefficient as:

$$n(I) = n_0 + n_2 I, \tag{2}$$

$$\alpha(I) = \alpha_0 + \beta I \tag{3}$$

where, n_0 is the linear refractive index, while n_2 denotes the non-linear refractive index and α_0 is the linear absorption coefficient, while β represents the non-linear absorption coefficient. I denotes the intensity of light. For a thin sample, the output field can be written as follows:

$$E_{\text{out}} = E(r,z)e^{-\frac{\alpha_0 L}{2}}(1+q)^{-\frac{ikn_2}{\beta}-\frac{1}{2}} \tag{4}$$

where, $q = \beta I L_{eff}$ with $L_{eff} = \left(1-e^{-\alpha_0 L}\right)/\alpha_0$, and $I_0 = |E_0|^2$ is the on-axis intensity at beam waist. For very thin samples, such as in this experiment, we can take $L_{eff} \approx 1$ (in proper units). Moreover, I_0 can be written as:

$$I_0 = \frac{P_{Avg}}{RR \times PW \times (\pi w_0^2)}, \tag{5}$$

where, P_{Avg} is the average beam power, RR is the LASER Repetition Rate, PW is the LASER Pulse Width and πw_0^2 is the beam area, at focus.

The intensity distribution and phase shift of the beam, at the exiting surface, must also fulfill these criteria [2]:

$$I_{\text{out}}(r,z) = I(r,z)\frac{\exp(-\alpha_0 L)}{1+q(r,z)} \tag{6}$$

and,

$$\Delta\phi = \frac{kn_2}{\beta}\ln(1+q(r,z)) \tag{7}$$

Now, the intensity, $I(r,z)$, can be expressed as the product of I_0 and a local Gaussian profile, G_{local} [2], that can be written as follows:

$$G_{local} = \frac{1}{1+x^2}\exp\left(-\frac{2r^2}{w^2(z)}\right) \tag{8}$$

When only NLR is present, we can write the non-linear phase change as:

$$\Delta\phi = \Delta\Phi_0 G_{\text{local}} \tag{9}$$

while for materials exhibiting pure NLA, q takes the following form:

$$q(r) = \Delta\Psi_0 G_{\text{local}} \tag{10}$$

Here, $\Delta\Phi_0$ and $\Delta\Psi_0$ are merely the results of the respective first order expansions of the expressions for $\Delta\phi$ (Eq. 7) and q, in the cases of pure NLR and pure NLA respectively. This gives us:

$$\Delta\Phi_0 = kn_2 I_0 L_{eff} \tag{11}$$

and,

$$\Delta\Psi_0 = 2^{-1.5}\beta I_0 L_{eff} \tag{12}$$

Substituting these quantities from Eq. 9 and Eq. 10 into Eq. 4 , we obtain a final expression for the outgoing field:

$$E_{\text{out}} = E(r,z)e^{-\frac{\alpha_0 L}{2}}\left(1+\Delta\Psi_0 G_{\text{local}}\right)^{-\frac{i\Delta\Phi_0}{\Delta\psi_0}-\frac{1}{2}} \tag{13}$$

Now, making use of SVEA and the Gaussian Decomposition method for local field changes and considering only the first two terms in the decomposition, we can obtain an analytical expression for the complex, on-axis far-field given as ([1], [2]):

$$E_{\text{out}} = E(r,z)\exp\left(-\frac{\alpha_0 L}{2}\right)\sum_{n=0}^{\infty}\left[\frac{[-i\Delta\phi_0(z)]^n}{n!}\prod_{n'=0}^{n}\left(1-i\left(2n'-1\right)\frac{\Delta\Psi_0}{\Delta\Phi_0}\right)\right]\exp\left(\frac{-2nr^2}{w^2(z)}\right). \tag{14}$$

Using Eq. 14 we can write the complex field at the aperture planes as follows:

$$E_a = E(r,z)\sum_{n=0}^{\infty}\frac{[-i\Delta\phi_0(z)]^n}{n!}\prod_{n'=0}^{n}\left(1-i(2n-1)\frac{\Delta\Psi_0}{\Delta\Phi_0}\right)\frac{w_{n0}}{w_n}\exp\left(-\frac{r^2}{w_n^2}-\frac{ikr^2}{2R_n}+i\theta_n\right) \tag{15}$$

Here, $\Delta\phi_0 = \Delta\Phi_0/\left(1+x^2\right)$

When only NLR is considered, $\Delta\Psi_0 \to 0$, and we can reduce the previous equation to obtain:

$$E_a = E(r,z)\sum_{n=0}^{\infty}\frac{[-i\Delta\phi_0(z)]^n}{n!}\frac{w_{n0}}{w_n}\exp\left(-\frac{r^2}{w_n^2}-\frac{ikr^2}{2R_n}+i\theta_n\right) \tag{16}$$

Taking d to be the propagation distance in free space, between the sample and the aperture, and $g = 1+d/R(z)$, we can represent the parameters in Eq. 15 and 16 as follows:

$$w_{n0}^2 = \frac{w^2(z)}{2n+1}$$

$$d_n = \frac{kw_{n0}^2}{2}$$

$$w_n^2 = w_{n0}^2\left(g^2+\frac{d^2}{d_n^2}\right)$$

$$R_n^2 = d\left(1-\frac{g}{g^2+d^2/d_n^2}\right)^{-1}$$

and,

$$\theta_n = \tan^{-1}\left(\frac{d}{gd_n}\right)$$

The total transmitted power through the aperture can then be obtained by spatially integrating $E_a(r,t)$, like so:

$$P_T\left(\Delta\Phi_0(t)\right) = \int_0^{r_a}\left|E_a(r,t)\right|^2 r\,\mathrm{d}r \tag{17}$$

This directly yields the normalized Z-Scan transmittance, $T(z)$, using the following expression:

$$T(z) = \frac{\int_{-\infty}^{\infty}P_T\left(\Delta\Phi_0(t)\right)\mathrm{d}t}{S\int_{-\infty}^{\infty}P_i(t)\mathrm{d}t} \tag{18}$$

where $P_i(t) = \pi w_0^2 I_0(t)/2$ is the instantaneous input power, within the sample, and $S = 1-\exp\left(-2r_a^2/w_a^2\right)$, where subscript - a denotes "aperture". This is where the aperture-dependence of the Z-Scan traces comes in.

For closed aperture traces, we can take $S \approx 0$ and for open aperture traces, $S \approx 1$. Now, the normalized transmittance for the on-axis electric field, at the aperture plane, can be written as:

$$T_2(z, \Delta\phi_0) = \frac{| E\left(r = 0, z, \Delta\phi_0\right|^2}{| E\left(r = 0, z, \Delta\phi_0 = 0\right|^2} \tag{19}$$

On substituting the expression for the field into Eq. 19 and simplifying, we obtain the final expression for $T(z)$, that considers the effect of both NLR and NLA, as given below:

$$T_2(z, \Delta\Phi_0, \Delta\Psi_0) = 1 + \frac{4\Delta\Phi_0 x}{(x^2+9)(x^2+1)} - \frac{\Delta\Psi_0}{x^2+1} \tag{20}$$

Eq. 20 provides a two-parameter fitting function for any third-order Z-Scan trace, the two parameters being $\Delta\Phi_0$ and $\Delta\Psi_0$, and we have used it to fit the closed aperture traces, in order to obtain the n_2 values in this experiment. On the other hand, for β values, we take the open aperture traces, that de-emphasize $\Delta\Phi_0$ or small intensity variations, as the entire signal is being measured. So, we can provide a simpler, one-parameter expression for $T(z)$ in this case:

$$T_2(z, \Delta\Phi_0 = 0, \Delta\Psi_0) = 1 - \frac{\Delta\Psi_0}{x^2+1} \tag{21}$$

Equations 20 and 21 are the working formulae for this experiment. Figure 4 shows variations in transmittance in Z-scan traces for materials exhibiting pure NLR, pure NLA and both NLR and NLA (mixed case), using various values for $\Delta\Phi_0$ and $\Delta\Psi_0$. It is clear from the plots that NLA suppresses peaks in the mixed Z-Scan traces, as compared to the pure NLR case. This also means that the symmetric nature of pure NLR curves is usually absent in most Z-Scan traces, where both NLR and NLA contribute. As such, we can utilize another interesting quantity, that is the Peak-Valley Transmittance Change, ΔT_{p-v}. It is expressed as follows in the closed aperture case (where we shall be using the two-parameter fitting function in Eq. 20):

$$\Delta T_{p-v} = 0.406\Delta\Phi_0 \tag{22}$$

This relation for ΔT_{p-v} should hold even for highly skewed Z-Scan traces. Therefore, it provides us with an excellent tool to verify the quality of our traces.

5 Conclusion

Now we can use Z-scan experimental configuration to obtain nonlinear refractive index and nonlinear absorption coefficients of any standard samples. The sign and magnitude of nonlinear refractive index of the samples can measured. We will then use the equations of normalized transmittance and fit them with the data points and will report the refractive index and nonlinear absorption coefficients.

6 Advantages and Disadvantages of Z-Scan

Advantages

- It is simple and very sensitive technique to measure the sign and magnitude of nonlinear refraction and absorption coefficients.
- It has no difficult alignment other than beam aligning on the aperture.
- Data analysis is quick and simple.
- It can determine both real and imaginary parts of $\chi^{(3)}$.
- Z-scan can also be modified to study non-linearities of higher order contributions.

Disadvantages

- It requires a high quality Gaussian TEM_{00} beam for absolute measurements.
- The analysis must be different if the beam is non-Gaussian.
- Sample distortions, tilting of sample during translation, can cause the beam to walk off the far field aperture.

7 Precautions and Sources of Error

- The polariser should be used to reduce the power of the laser in order to reduce the thermal effects of laser on the samples.
- Protective glasses must be worn while taking measurements and the laser beam shouldn't allowed to fall outside here and there by carefully blocking it with black aluminium plate.
- The lasers should be turned off when not in use.

8 References

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