Study of Non-Linear Optical Properties by using Z-scan Techniques

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Abstract

Z-Scan technique used for finding the nonlinear refractive index and absorption coefficient. In this method a nonlinear material is scanned through a laser beam and the intensity of the beam is measure. Due to nonlinear refractive index (depends on intensity) of the material, the beam diverges or converges through out the scan giving a pattern in the intensity measurement. In Z-Scan, we use open aperture method for finding the nonlinear absorption coefficients while close aperture method to measure nonlinear refractive index. We will use two samples CS_2 and Acetone for measuring these nonlinear properties

Chapter 1

Statement of Problem and Overview

1.1 Statement of Problem

The first laser built by *Theodore Maiman* on 16 May 1960 at the Hughes Research Laboratory in California, by shining a high-power flash lamp on a ruby rod with silver-coated surfaces. Nonlinear Optics is a branch of optics which deals with the behavior of light in nonlinear media(Polarization P related non-linearly with electric field). The non-linearity is typically observed only at very high light intensities (E=108 V/m) such as those provided by lasers. And hence by the laser invention, study of non-linear optical properties of materials becomes readily accessible.

However, High intensity Laser has a dark side. It can be harmful to human eyes and at times causing serious burns on direct contact, and can also damage some instruments. So there is a need of protection from laser. We are continuously looking for a materials which can transmit low intensity light but absorbs harmful high intensity light(laser radiation). These materials are known as optical limiters. While considering materials as optical limiter, two properties of interest are material's non-linear refractive index and its non-linear absorption coefficients as both changes the intensity of light non-linearly when light pass through a medium. Hence, by measuring these nonlinear properties we can identify materials which can be used for optical limiters.

1.2 Introduction

There are various techniques used for measuring non-linear absorption and non-linear refraction. The Z-Scan is the simplest and most sensitive of these techniques which is based on the principle of spatial beam distortion used for measuring the third order optical non-linearity and also allows computing the contributions of nonlinear absorption and nonlinear refraction towards the non-linearity. Z-Scan technique was first proposed by Sheik-Bahae in 1990 to measure the non-linear refractive index, n_2 and the non-linear absorption coefficient, β through the "closed" aperture and "open" aperture method respectively.

One of the key phenomenon that happens in Z-scan is Self focusing and defocusing. It is non linear optical process occurring due to change in refractive index of material when exposed to intense electromagnetic radiation. For Gaussian beam, maximum intensity is at the center and hence the maximum change of refractive index will be at the center. By keeping the overall power of beam constant, the sample is placed in the beam and translated through the focal region where the beam's intensity distribution changes because the beam size changes. As the sample is translated through the focus in z-direction, hence the name Z-scan given.

Aim of the experiment: Aim of our experiment was first to establishment of the Z-scan setup and then taking measurements of transmitted power for open and closed aperture by translating the material in z-direction. And by fitting these data with the appropriate formulas, we need to find nonlinear absorption coefficient and nonlinear refractive index of the medium.

Chapter 2

Theory

Nonlinear Optics is the branch of optics which studies the behavior of light in nonlinear medium.

2.1 Nonlinear Optical Media

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

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

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

$$\mathbf{P(E)} = \epsilon_0(\chi^{(1)}\mathbf{E} + \chi^{(2)}\mathbf{EE} + \chi^{(3)}\mathbf{EEE} + ...) \tag{2.2}$$

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.

2.1.1 Nonlinear wave equation

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

$$\nabla^2\mathbf{E} - \frac{1}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2} = \mu_0\frac{\partial^2\mathbf{P}}{\partial t^2}. \tag{2.3}$$

We can write $\mathbf{P}$ as sum of linear and nonlinear terms separately

$$\mathbf{P} = \epsilon_0 \chi^{(1)} \mathbf{E} + \mathbf{P}_{NL} \tag{2.4}$$

where $\mathbf{P}_{NL}$ is

$$\mathbf{P}_{NL} = \chi^{(2)} \mathbf{EE} + \chi^{(3)} \mathbf{EEE} + ... \tag{2.5}$$

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

$$\nabla^2 \mathbf{E} - \frac{1}{v^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2}. \tag{2.6}$$

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

2.2 Third-order Nonlinear Optics

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

$$\mathbf{P(E)} = \epsilon_0 \chi^{(1)} \mathbf{E} + 0 + \epsilon_0 \chi^{(3)} \mathbf{EEE} \tag{2.7}$$

where $\mathbf{E}$ is

$$\mathbf{E} = \frac{1}{2}(E_0 e^{ikz - i\omega t} + E_0^* e^{-ikz + i\omega t}) \tag{2.8}$$

Put this eq. in (2.7), we get

$$\mathbf{P(E)} = \epsilon_0 (\chi^{(1)} + \frac{3}{4} \chi^{(3)} |E_0|^2) \mathbf{E} \tag{2.9}$$

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

$$n^2 = 1 + \chi_{eff} = 1 + \chi^{(1)} + \frac{3}{4} \chi^{(3)} |E_0|^2 \tag{2.10}$$

$$n^2 = n_0^2 + \frac{3}{4} \chi^{(3)} |E_0|^2 = n_0^2 (1 + \frac{3\chi^{(3)} |E_0|^2}{4n_0^2}) \tag{2.11}$$

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

$$n = n_0(1 + \frac{3}{8n_0^2}\chi^{(3)}|E_0|^2) = n_0 + \frac{3}{8n_0}\chi^{(3)}|E_0|^2 \tag{2.12}$$

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

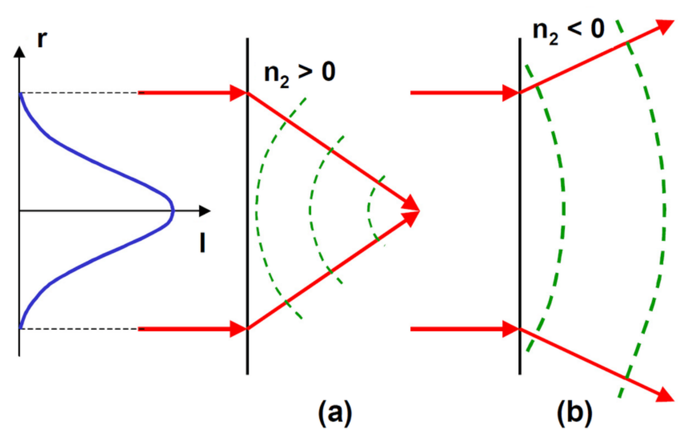
$$n = n_0 + \frac{3}{4}\chi^{(3)}(\frac{I}{\epsilon_0 n_0^2 c}) \tag{2.13}$$

$$\boxed{n = n_0 + n_2 I} \tag{2.14}$$

where $n_2 = \dfrac{3\chi^{(3)}}{4\epsilon_0 n_0^2 c}$ 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.

2.2.1 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).



2.3 Z-Scan Experiment

2.3.1 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 $n_2 < 0$.

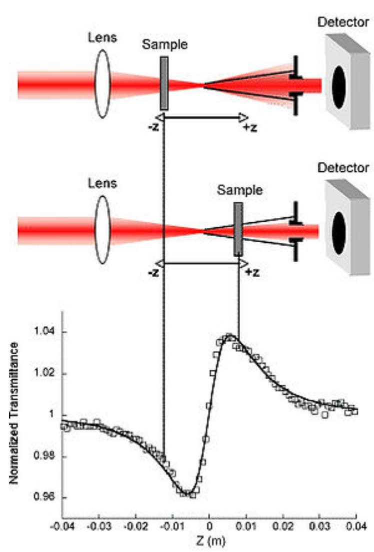


fig. 1a) Non-linear refraction

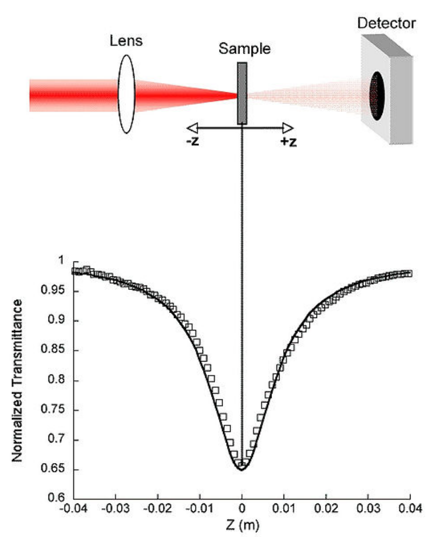


fig. 1b) Non-linear Absorption

If the material has a positive nonlinearity ($n_2 > 0$), 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)(fig. 2).

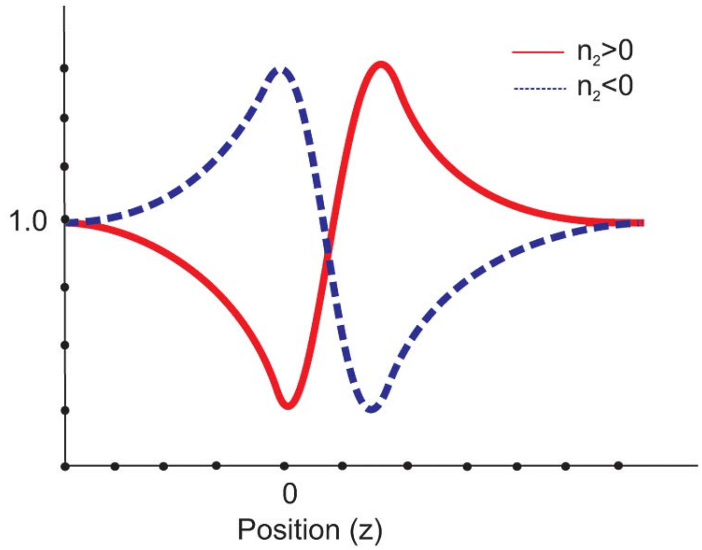
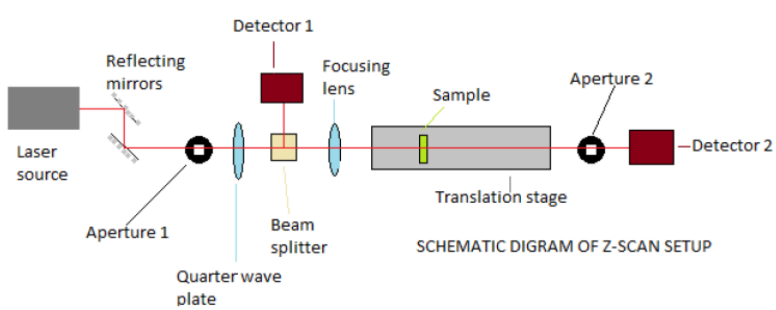


fig. 2. Determination of a sign of n_2 from transmittance graph

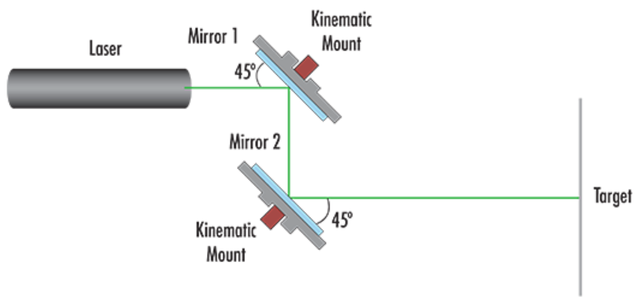
The aperture plays an important role in determining which values we are going to find. We use closed aperture for finding out the non-linear refractive index and open aperture to find the absorption coefficient.

2.3.2 Apparatus used and their Working

The experimental set up is given below



- 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)



Parallel (Z-Fold) Configuration

- 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)
- 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 electricallyand is measure of the absorbed energy.

- Photo detectors: A photo detector has a p–n junction that converts light photons into current. The absorbed photons make electron–hole pairs in the depletion region. Photodiodes and photo transistors are a few examples of photo detectors.

2.3.3 Formulas used to derive Nonlinear refraction and nonlinear absorption

We use Gaussian beam with

$$E(r,z,t) = E_0(t)\frac{w_0}{w(z)}exp[-\frac{r^2}{w^2(z)} - \frac{ikr^2}{2R(z)}]\ exp[-i\phi(z,t)] \tag{2.15}$$

where w(z) is the radius of the beam at z, E_0 is the electric field at the beam waist (z=0, r=0) and the last term contains all the radially uniform phase variations.

Open aperture-Nonlinear Absorption: As the sample is translated through the focal region of the beam, detector 1 measures the total transmitted intensity. Because only the irradiance at the sample is changing as the sample is translated, any deviation in the total transmitted intensity must be due to multi-photon absorption. In the limit where multi-photon effects are limited to two-photon absorption, the normalized change in transmitted intensity can be approximated by the following equation,

$$T(z) = 1 - \frac{q_0}{2^{3/2}(1+x^2)} \tag{2.16}$$

Here $q_0 = \beta I_0 L_{eff}$, where β is the nonlinear absorption coefficients, x=z/z_R where z_R is the Rayleigh range, I_0 is the intensity at focus

$$I_0 = \frac{P_{avg}}{(repitition\ rate)(pulse\ width)(\pi w_0^2)} \tag{2.17}$$

L_{eff}=(1-exp[-αL])/α is the effective propagation length inside sample. But in our case, L is very small compared to translation stage implies $L_{eff} \approx L$.

Close aperture-Nonlinear Refraction: The normalized transmission of closed aperture Z-scan is given by

$$T = [1 + \phi\frac{2x}{1+x^2} + \phi^2\frac{x}{1+x^2}]^{-1} \tag{2.18}$$

where ϕ is the nonlinear phase change. This equation can further be simplified to

$$T(z, S=1) = 1 - \frac{4\Delta\Phi_0 x}{(x^2+1)(x^2+9)} - \frac{2\Delta\psi_0}{(x^2+1)} \tag{2.19}$$

where $\Delta\Phi_0 = kn_2 I_0 L_{eff}$ and $\Delta\psi_0 = \beta I_0 L_{eff}/2^{5/2}$.

Chapter 3

Observation and Analysis

3.1 Alignment

In this z-scan experiment, we first aligned the whole setup using the laser beam of 532 nm with intensity at 40%. First we did the two mirror alignment by using two aperture. The first mirror was adjusted for centering the beam on the first aperture and second mirror was adjusted for centering the beam on the second aperture. After that half wave plate and polarizing beam splitter has been placed, followed by a lens of focal length 10 cm. The reflected beams were checked for perfect aligned, and ensured no back reflection directed into the source directly.

After the alignment process, a translation stage was placed in between the lens and the second aperture and laser should be at his full intensity. Detectors output and translation stage was connected to a desktop so that it can be controlled remotely. StarLab software was used to extract data from the detectors. We used two samples, CS_2 and Acetone to collect the data.

3.2 Data acquisition

We were to find nonlinear absorption coefficient and nonlinear refractive index for the above two materials. The StarLab software along with translation stage would give us the data points for a length of 10 cm. We use these data points and fit the open aperture data with eq. 2.16 formula and close aperture data with eq. 2.19. We use python for analyzing the data, plotting graphs, finding nonlinear refractive index and absorption coefficients along with errors.

3.3 Data Analysis

- Effective length of the sample, L_{eff}=1mm
- Repetition rate of Laser= 80 kHz
- Pulse width at a given repetition rate= 1.75 ns
- Beam waist of a laser beam = 35× 10^{-6} m

For CS_2 sample

1. Open Aperture

- RPM of motor= 100
- Average power= 8.765 mW
- q_0 =0.54538106 (fitting parameter)
- Non-linear absorption coefficient, $\beta = (3.3525 \pm 0.1275) \times 10^{-8}\ mW^{-1}$

2. Close Aperture

- RPM of motor= 400
- Average power= 205.399 μW
- $\Delta\Phi_0$ =0.48917417 (fitting parameter)
- Non-linear refractive index, $n_2 = (1.08645 \pm 0.02634) \times 10^{-13}\ m^2W^{-1}$

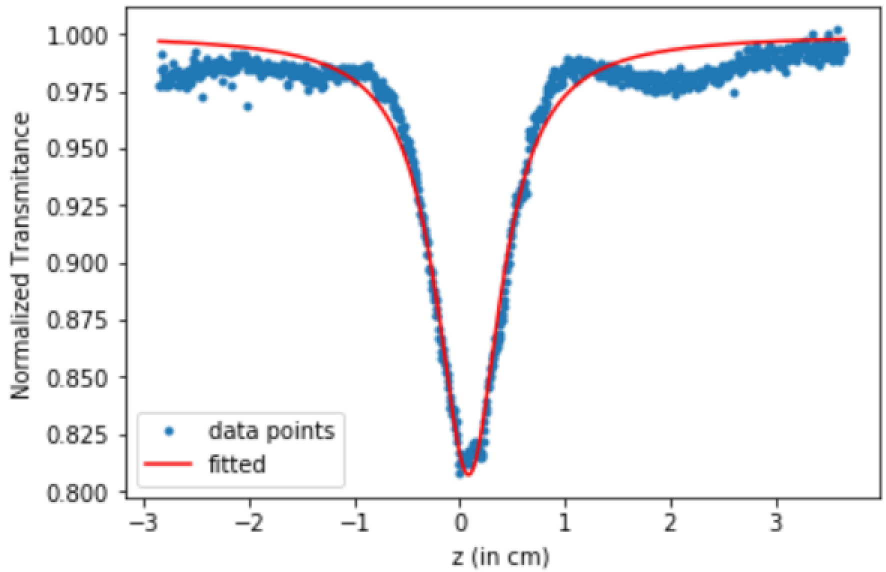


Figure 3.1: Open aperture data for CS_2

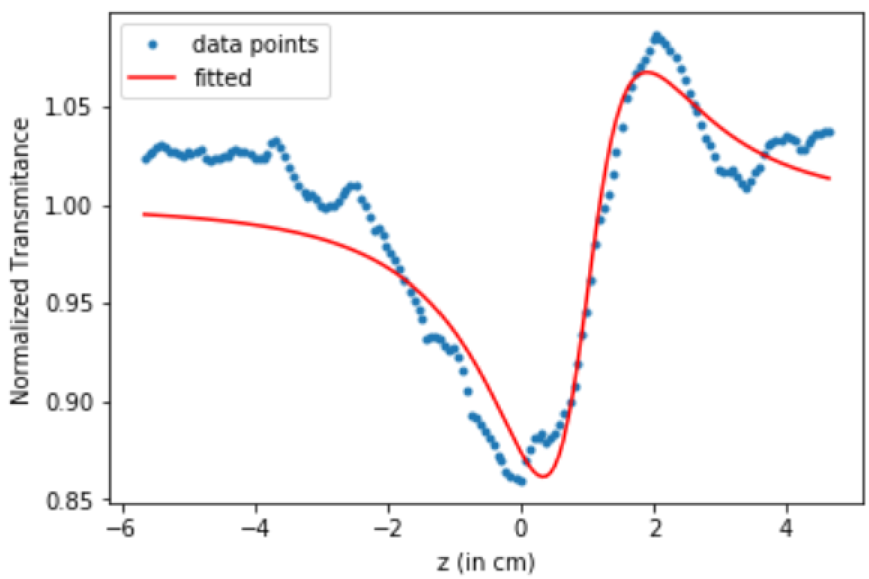


Figure 3.2: Close aperture data for CS_2

For Acetone sample

Open Aperture

- RPM of motor= 300
- Average power= 9.897 mW
- q_0 =0.1950707 (fitting parameter)
- Non-linear absorption coefficient, $\beta = (1.062 \pm 0.0378) \times 10^{-8}\ mW^{-1}$

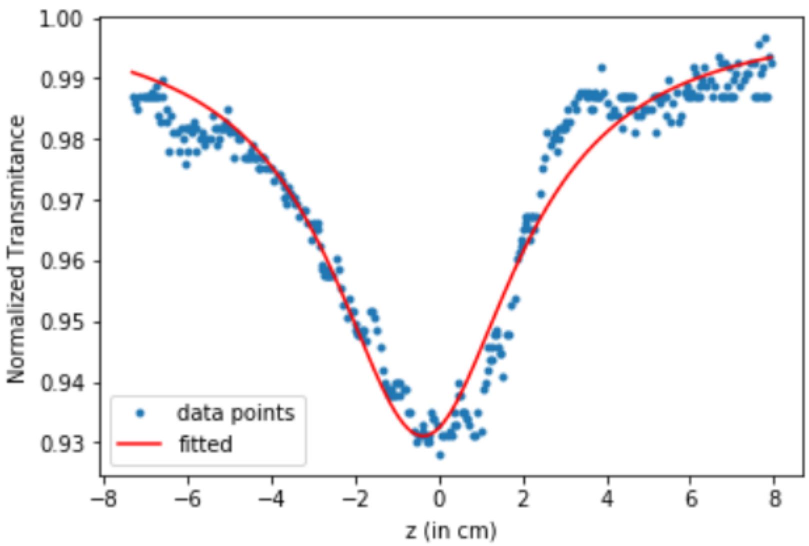


Figure 3.3: Open aperture data for Acetone

Close Aperture

- RPM of motor= 300
- Average power= 439.302 μW
- $\Delta\Phi_0$ =0.274286 (fitting parameter)
- Non-linear refractive index, $n_2 = (2.8483 \pm 0.5961) \times 10^{-14}\ m^2W^{-1}$

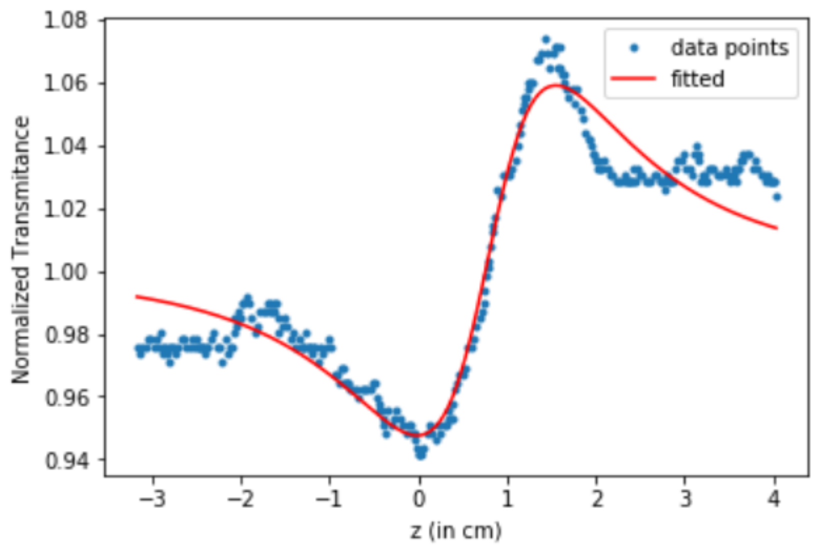


Figure 3.4: Close aperture data for Acetone

- For close aperture, Transmittance plots for both the samples shows valley is dominant over peak, that is the sign of nonlinear absorption. In case of pure refraction peak and valley should be of same size.
- CS_2 has large optical nonlinearities in this regime due to large nonlinear absorption and refraction. Due to these large nonlinear optical coefficients CS_2 acts as an ideal reference material.

3.4 Result

- Nonlinear refractive index found out to be $n_2 = (1.08645 \pm 0.02634) \times 10^{-13}\ m^2W^{-1}$ for CS_2 and $n_2 = (2.8483 \pm 0.5961) \times 10^{-14}\ m^2W^{-1}$ for Acetone.
- Nonlinear Absorption coefficient found out to be $\beta = (3.3525 \pm 0.1275) \times 10^{-8}\ mW^{-1}$ for CS_2 and $\beta = (1.062 \pm 0.0378) \times 10^{-8}\ mW^{-1}$ for Acetone.
- CS_2 shows more non-linearity than acetone.

3.4.1 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 nonlinearities 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.

Conclusion

We use Z-scan experimental configuration to obtain nonlinear refractive index and nonlinear absorption coefficients of a standard samples CS_2 and Acetone. The sign and magnitude of nonlinear refractive index of both the samples were measured. We have used the equations of normalized transmittance and fit them with the data points and turns out to be really good fitting for all the graphs.

Precautions and Sources of Error

- Proper alignment should be performed before taking data.
- 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 cuvette should be cleaned with acetone before use.
- The lasers should be turned off when not in use.

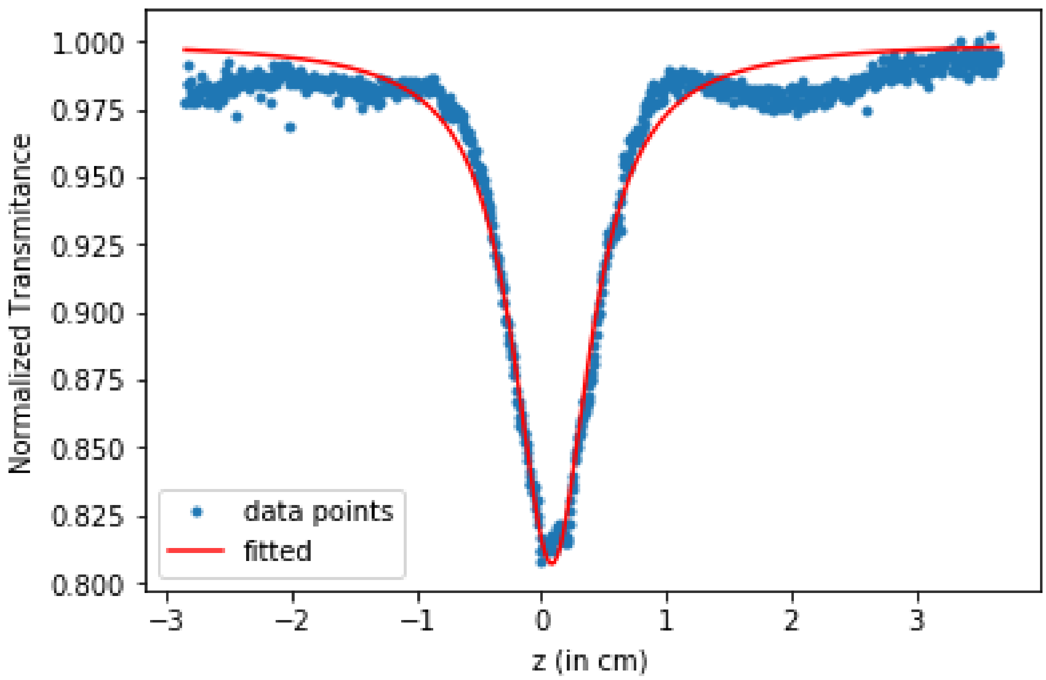
Appendix

October 2, 2019

In [42]:
```
################Open Aperture Fitting##################
import math
from scipy.optimize import curve_fit
import numpy as np
import matplotlib.pyplot as plt


s =np.loadtxt('open 100.txt ')
z =[]
power =[]
g=np.size(s,0)
for j in range(0,g):
    z.append(s[j][0])
    power.append(s[j][1])
#z=np.array(z)
#power=np.array(power)
print(((np.argmin(power))))
power=np.array(power)
z=(z-z[int((np.argmin(power)))])/10
power=power*110

def f(x,a,b,c):
    return 1-c/((2**(1.5))*((x/a-b)**2+1))

plt.plot(z,power,'.',label='data points')
u1,v1=curve_fit(f,z,power,maxfev=2000)
plt.plot(z,f(z,*u1),'r',label='fitted')
         #,label="a= %3.2f, b= %5.2f, c= %5.2f" %tuple(u1))
#plt.title('Open Aperature data for CS$_2$')
plt.xlabel('z (in cm)')
plt.ylabel('Normalized Transmitance')
plt.legend()
plt.show()
#gmodel=Model(f)
#result=gmodel.fit(power,x=z,c=1,a=1,b=0)
#print(tuple(u1))
s=np.sum(power)/(110*979)
```

```
print('average power=',s)
print(u1)
```

429

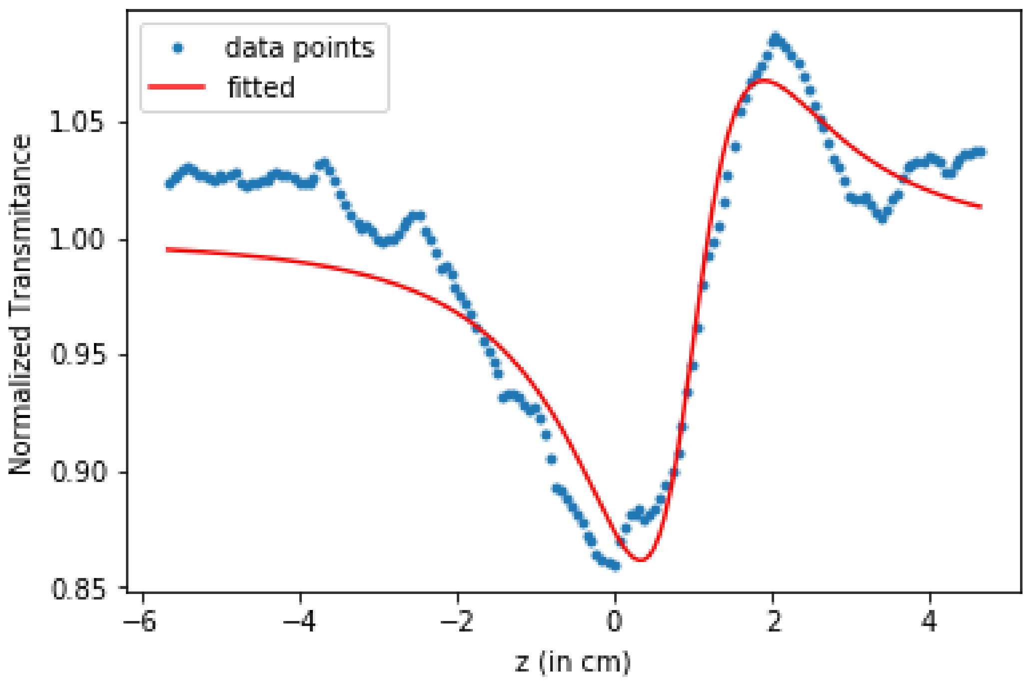



```
average power= 0.008764514811031665
[0.3702443  0.22124167 0.54538106]
```

In [41]:
```
################Close Aperture Fitting#################
import math
from scipy.optimize import curve_fit
import numpy as np
import matplotlib.pyplot as plt


s =np.loadtxt('close 400_cutted.txt ')
z =[]
power =[]
g=np.size(s,0)
for j in range(0,g):
    z.append(s[j][0])
    power.append(s[j][1])
```

```
print(((np.argmin(power))))
power=np.array(power)
z=z-z[int((np.argmin(power)+np.argmin(power))/2)]
power=power*4830

def f(x,h,s,a,b):
    return 1+((4*h*(x/a-b))/((((x/a-b)**2)+1)*(((x/a-b)**2)+9)))-(2*(((x/a-b)**2)+3)*s

u1,v1=curve_fit(f,z,power,maxfev=20000)
plt.plot(z,f(z,*u1),'r',label='fitted')
#,label="h= %3.2f,s= %3.2f,a= %3.2f,b= %3.2f" %tuple(u1))
#plt.title('Close Aperature data for CS$_2$')
plt.xlabel('z (in cm)')
plt.ylabel('Normalized Transmitance')
plt.legend()
plt.show()
s=np.sum(power)/(4830*156)
print('average power=',s)
print(u1)
```

```
85
```




```
average power= 0.00020539871794871795
[0.48917417 0.03097369 0.88407937 1.05766381]
```