Observation of Two-photon absorption using Z-Scan experiment

Subhajit Mishra
4th year, DPS, IISER-Kolkata
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We present a brief description of non-linear optical processes like, harmonic generation, self focusing, two-photon absorption etc. and determination of non-linear optical processes (non-linear absorption and refractive index) using Z-Scan technique. We have briefed about both open and closed aperture experiment. Lab-view programming language is used for the movement of motion controller. A short algorithm with a sample code is given.

I. INTRODUCTION

Nonlinear optics is a phenomenon, can be seen in certain materials when intense light is shown to those materials. It is the modification of natural optical properties by the presence of light. Typically, any intense laser light is enough to produce this phenomena. Nonlinear optical phenomena are "nonlinear" in the sense that, they occur when the response of a material to an applied optical field depends in a nonlinear manner on the strength of the applied field.

In case of linear optics, induced polarization depends linearly on applied electric field,

$$P(t) = \epsilon_0 \chi^{(1)} E \tag{1}$$

where $\chi^{(1)}$ is the linear susceptibility and ϵ_0 is permittivity in free space.

In nonlinear optics, polarization depends on field nonlinearly as below,

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

where $\chi^{(i)}$ is the i-th order susceptibility. $\chi^{(2)}$ can occur only in non-centro-symmetric crystals. Since, most of the materials posses centro-symmetry, $\chi^{(2)}$ is taken as 0.

II. NONLINEAR OPTICAL PROCESSES

In this section we will describe some nonlinear optical phenomenon.

A. Harmonic Generation

Here, we are considering second harmonic generation as nonlinear optical process. Consider a laser beam, whose intensity is given by,

$$\tilde{E}(t) = Ee^{-i\omega t} + E^* e^{i\omega t} \tag{3}$$

is incident upon a crystal whose $\chi^{(2)}$ is nonzero. For this case, the 2nd order nonlinear polarization is given by,

$$P^{(2)}(t) = \epsilon_0 \chi^{(2)} \tilde{E}(t) \tag{4}$$

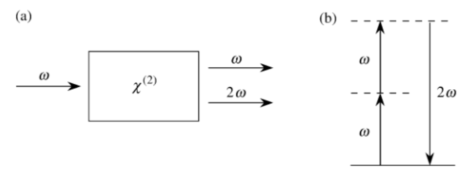

FIG. 1. (a) Geometry of second harmonic generation, (b) Energy-level diagram describing 2nd harmonic generation

after doing the squaring we get,

$$P^{(2)}(t) = 2\epsilon_0 \chi^{(2)}|E|^2 + E^2 \epsilon_0 \chi^{(2)} e^{-i2\omega t} + c.c \tag{5}$$

We can see that second order polarization has a frequency term which is twice of the incident frequency (2ω). This is second harmonic generation. Taking nth order polarization we can see that $n\omega$ frequency can be generated.

B. Intensity dependent Refractive index

From third harmonic generation we get,

$$P^{(3)}(t) = \frac{1}{4}\epsilon_0 \chi^{(3)} E^3 \cos 3\omega t + \frac{3}{4}\epsilon_0 \chi^{(3)} E^3 \cos \omega t \tag{6}$$

The first term of the above equation has nonlinear frequency 3ω of the incident wave of ω frequency. This gives rise to nonlinear effect of refractive index. Moreover, nonlinearity in refractive index can be written as,

$$n = n_0 + n_2 I \tag{7}$$

where n_0 is the linear refractive index, n_2 is the optical constant determined by the field intensity I.

Self-focusing or **Self De-focusing** can occur in this case due to non uniform refractive index, depending on n_2 is positive or negative. Similarly, **Self-Phase Modulation** can occur due to the optical path change of light in the material because of non uniform refractive index.

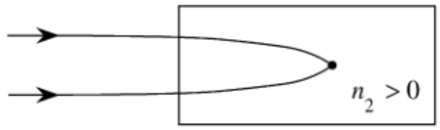

FIG. 2. Self focusing of light

III. NONLINEAR ABSORPTIONS

A. Saturable Absorption

Saturable absorption is the property of material where the absorption of light decreases with increasing light intensity. At sufficiently high intensity light, ground state atoms of a material can excite in such a rate that there is insufficient amount of time such that those excited atoms can de-excite. This process depletes the ground state, which in turn saturates the absorption. Absorption coefficient is given by,

$$\alpha = \frac{\alpha_0}{1 + \frac{I}{I_s}} \tag{8}$$

where I is the incident laser intensity and I_s is the saturable intensity.

B. Two-photon Absorption

In this process an atom makes a transition from ground state to excited state by absorbing two photons simultaneously. During the process a virtual state (shown by dotted line in the FIG.3) is created between the real ground state and excited state.

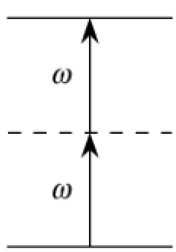

FIG. 3. Two-photon absorption

There is another absorption process called **Reverse Saturable Absorption**.

IV. Z-SCAN EXPERIMENT

Z-Scan measurement is a technique used for measuring non-linear refractive index and non-linear absorption coefficient. In this section I have briefly described the Z-Scan experiment.

A. Closed Aperture Z-Scan

Non linear refractive index can be measured using closed aperture z-scan. The basic set up is given below in the FIG.4:

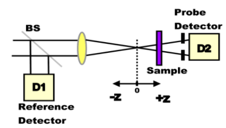

FIG. 4. Basic Z-Scan Setup *an aperture should be placed in front of the detector for closed aperture z-scan*

Since non-linear properties arises at high intensity, the material should be placed close to the focus of the beam *(to determine the focus for the Gaussian beam, we need to do **knife-edge experiment**)*.

Say, $z = 0$ is the focus and $z \to \pm\infty$ is considered to be far away from focus. Consider the non-linear sample having $n_2 > 0$.

- At $z \to -\infty$, the intensity is low. So, the non-linear effect is negligible and say transmitted intensity is $T_{-\infty}$.
- At $z \to 0^-$(i.e., when the sample is close to left side of the focus, moving from $-\infty$), there is sufficient intensity so that non-linear effect arises. Because of positive refractive index, the sample self focuses the beam. So, the focal point shifts to the left side resulting a larger cross section of beam falling on the detector. Since, an small aperture is present intensity of transmitted light decreases.
- At $z = 0$, minimum of the transmittance occurs. Now if we move towards $z \to \infty$ transmitted intensity rises, attains a maximum and becomes constant with $T_\infty = T_{-\infty}$.

The figure of transmittance as a function of z for both $n_2 > 0$ and < 0 is given below,

Thus by looking at the transmittance graph we can figure out the sign of non-linear refractive index.

B. Open Aperture Z-Scan

The non linear refractive index does not occur on its own, usually in conjunction with non linear absorption. To measure the non-linear absorption we need to remove the aperture by a=making it open so that all the transmitted intensity can fall into the detector.

We will consider the case of **two-photon absorption**, because it is relevant to our experiment.

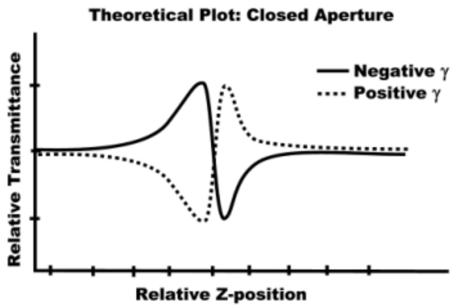

FIG. 5. Transmittance Intensity vs Relative Position(z). Dashed line is for $n_2 < 0$ and solid line is for $n_2 > 0$.

- At $z \to -\infty$, non linear effect is negligible. Say, transmittance is $T_{-\infty}$.
- At $z \to 0^-$, two-photon absorption occurs. So, transmitted intensity decreases.
- At $z = 0$, transmitted intensity attains minimum and when we move far away to $+\infty$ it again increases giving rise to FIG.6 with $T_\infty = T_{-\infty}$.

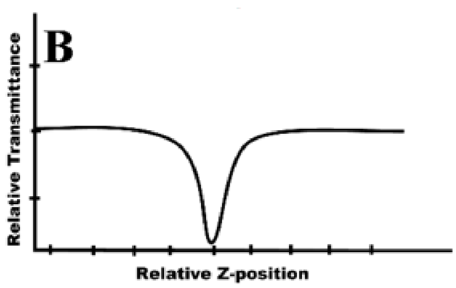

FIG. 6. Transmitted Intensity vs Position for Open Z-Scan

Thus by looking at this kind of graph one can find the two-photon absorption.

V. EXPERIMENT AND LAB-VIEW PROGRAMMING

To do the Z-scan measurement (We are only doing open z-scan. Since, we are only interested in observing two-photon absorption.) we used a movable stage on which the sample is fixed. Since we require fine movement of the sample near the focus we used motion controller, which allows us to move the stage precise via Lab-View code. A pulsed laser of 10 Watt with 100fps pulse length and 10ns frequency was used. To determine the focus precisely we planned to to knife-edge experiment. We wrote the Lab-View program which takes initial, final and position step as input, sends the command to motion controller to move the stage. After the sample reaches to

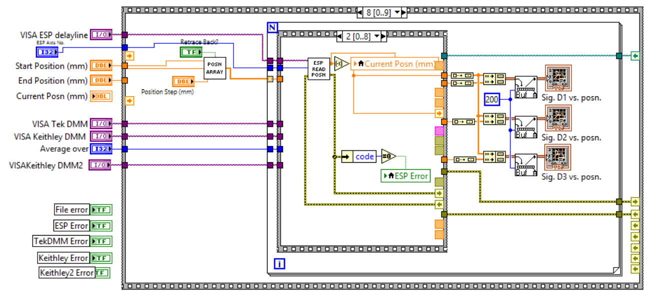

FIG. 7. Sample Code of Lab View code written for the experiment.

a position, the data is recorded and stored in a data file. A sample of code attached as figure.

For every optics experiment alignment is absolutely necessary. We devoted a significant amount of time to align the experimental set up.

VI. CONCLUSION

Z-scan measurement is one of the powerful techniques to determine the non-linear absorption and refractive index. We successfully implemented the labview program for motion controller and aligned the experimental set-up. But unfortunately, due to some unavoidable circumstances we could not complete our aim. However, we are planning to complete the experiment in near future.

VII. ACKNOWLEDGEMENT

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VIII. REFERENCES

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