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ARTICLE

INVESTIGATION OF THE NONLINEAR OPTICAL PROPERTIES OF THE SALEN-H_2 LIGAND USING Z-SCAN TECHNIQUE

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ABSTRACT

Background: *nonlinear optical properties of organic materials are widely used in optical switching and optical labeling.* ***Methods:*** *In this study, nonlinear optical properties of Salen-H2 (N, N′-ethylenebis (salicylideneimine)) ligand have been investigated. Linear optical responses of Salen-H2 have been studied using Uv-Vis spectroscopy. Non-linear refractive index and non-linear absorption coefficient of this ligand have been calculated using z scan. The scan method has been studied with the open and closed aperture.* ***Results:*** *Non-linear refractive index and absorption coefficient are related to the real and imaginary components of third order susceptibility coefficient, respectively.*

Conclusions: *Using Z scan method, the value of the nonlinear refractive index is calculated of the order of* $10^{-5,\frac{cm^2}{\omega}}$ *and is positive.*

KEY WORDS
Z-Scan; non-linear refractive index; non-linear absorption coefficient

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INTRODUCTION

In recent years, nonlinear optical properties of organic materials have been taken into consideration. These materials, with nonlinear optical properties are widely used in optical switching and optical labeling [1, 2]. The material used in the optical switching has high nonlinear refractive index and rapid response to radiant light intensities [3, 4].

Medium interaction with light is described in both linear and nonlinear optics and environmental loads caused by lighting are oscillated and this oscillation causes radiation of the electromagnetic waves. Phase-shift is between the resulting electric field and the radiant electric field [5]. Medium refractive index varies with charged particles environment. If the shifted refractive index is heavily dependent on the input spectrum, the refractive index is nonlinear and otherwise the medium has linear refractive index [5]. The nonlinear optical response of a material is often explained by polarization. Material polarization may be extended to the following formula:

(1)

$$p\,(t)=\chi^{((1))}E+\chi^{((2))}E+\chi^{((3))}E$$

Where $\chi^{((n))\,i}$ is Electric susceptibility of the material and E is the electric field. $\chi^{((2))}$ and $\chi^{((3))}$ are linear and nonlinear susceptibility, respectively, and for typical intensities of light passing, only the first extended statement is considered. For fields with high intensity, higher-order statements should be considered with extended series [6]. The principle of superposition is not true about the area of nonlinear optics and the frequency is not fixed in this medium. As well as the absorption coefficient and refractive index vary with changes in light intensity. $\chi^{((n))}$ is a complex quantity which its real and imaginary components express the nonlinear refractive index [7]. Non-linear refractive index dependence with heavy light intensity can be written as follows:

(2)

$$n = n_2 I + n_0$$

If n_2 is positive, the light beam while passing through the non-linear medium receives greater refractive index than around, therefore, it passes by lower speed than the previous medium and stay behind the edges. As a result, the initial wave front is changed too much. In this case, the medium acts like a convergent lens [8, 9].

However, if n_2 is negative, in this case the medium will be act like a divergent lens. In this case defocusing phenomenon occurs [10]. The easiest way to determine is the z- scan method which firstly evaluated by Sheikh Bahai. In this study, the method is used to determine the coefficients in $C_{16}H_{16}N_2O_2$.

MATERIALS AND METHODS

Synthesis of Salen

4.0 mmol of 2-hydroxybenzaldehyde in ethanol (50 ml) was added dropwise to an ethanolic solution (10 ml) of 2.0 mmol ethylenediamine. This mixture was refluxed for 2 h. Afterwards the mixture was cooled to room temperature, then cooled for 24 h at 5ºC. The yellow solid was filtered off, and recrystallized from ethanol [11]. Yield: 92.5%. m.p.: 125ºC .

Selected IR bands (KBr, cm^{-1}): 3450 (OH); 1635s (C=N); 1576m (C=C); 1371m (C-N); 1283m (C-O); UV–Vis (DMSO): λmax=410 (nm).
Schiff base ligand is shown in [Fig. 1]

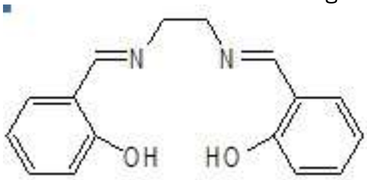

Fig. 1: Schiff base ligand structure (SalenH_2 or (N, N'-ethylenebis (salicylidene amine)

Optical absorption graph

Firstly, the absorption spectrum of the material was measured and according to [Fig. 2], we find that material is absorbed in the region of 200 to 450 nm.

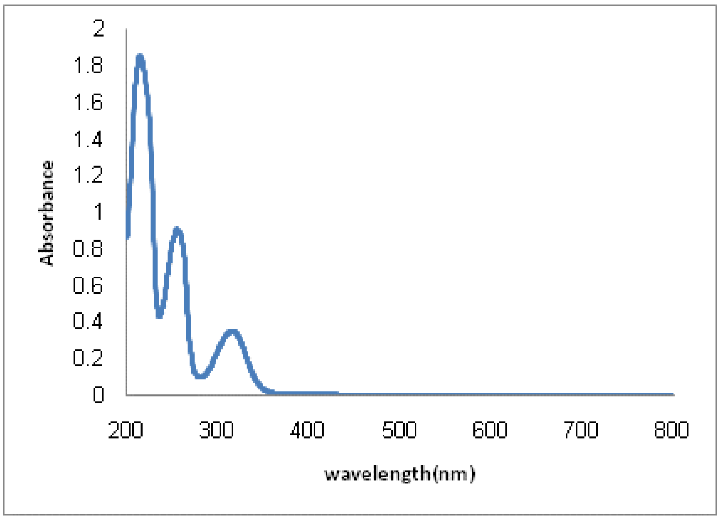

Fig. 2: Absorption graph versus wavelength

RESULTS AND DISCUSSION

Z-scan arrangement includes two open and closed apertures that determine β nonlinear absorption coefficient and n_2 non-linear refractive index respectively. In [Fig. 3], z-scan experiments layout has been shown [12, 13].

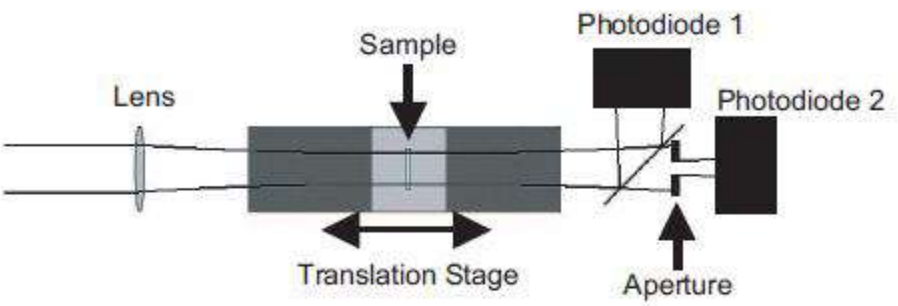

Fig. 3: Arrangement of Z-scan test

In this technique, the sample moves to the laser beam radiation towards to focusing beam point. This movement causes the light intensity to change the sample. During this motion, every evident step by the power meter is recorded.

CHEMISTRY

For open aperture, the intensity increases until they reach the center of the sample absorption. The lowest transition occurs at the center because of the high absorption and then is decreased with the passage of nonlinear absorption center and finally transition is increased to reach to the linear transition. The resulting graph will be the valley .As the sample is passed from a focal area of the beam, (1) detector measures the total transmitted intensity. Since in scanning only the radiant density from the sample is changed, any deviation in the intensity of the total transmission is caused by multi-photon absorption.
β is calculated using open aperture arrangement with nonlinear absorption coefficient. If the nonlinear absorption coefficient β is positive, crossing is increased by increasing the input power implying that the two-photon absorption occurs in this case. If β is negative, increasing input power decreases crossing and saturated absorption takes place in this case. The value β is obtained by fitting the [Fig. 4] and by the following approximation [14].

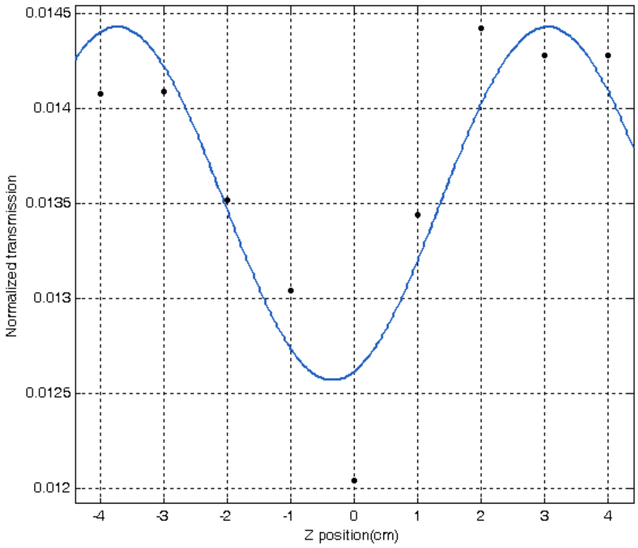

Fig. 4: Z-scan graph in the Open apertures

(3)

$$q_0 < 1$$

$$T(z,s=1) = \sum_{m=0}^{\infty} \frac{[-q_0(z)]^m}{(m+1)^{\frac{3}{2}}}$$

The $q_0(z)$ is obtained from the following equation
(4)

$$q_0(z) = \frac{I_0 L_{eff} \beta}{\left(1 + \left(\frac{z}{z_0}\right)^2\right)}$$

In the above equation z_0 is diffraction length and its value is [15]:
(5)

$$z_0 = \frac{K\omega_0^2}{2}$$

Z negative values indicate that the sample position between the lens and the focal point of the beam is narrow and positive values Z represent the position of the focal point between the beam and detector. Given the values of I_0, L_{eff}, q_0, and an estimated value for β, graph is plotted and empirical data curve on this graph has been plotted. By changing the value of β, good fit has been made between experimental and theoretical graph and finally the nonlinear absorption coefficient is calculated.
In the closed aperture, as the sample passes from a narrow focused area, photon detector measures the reduced intensity from the transmitted sample. The nonlinear refraction coefficient has been calculated by [Fig. 5] and the following relationship.

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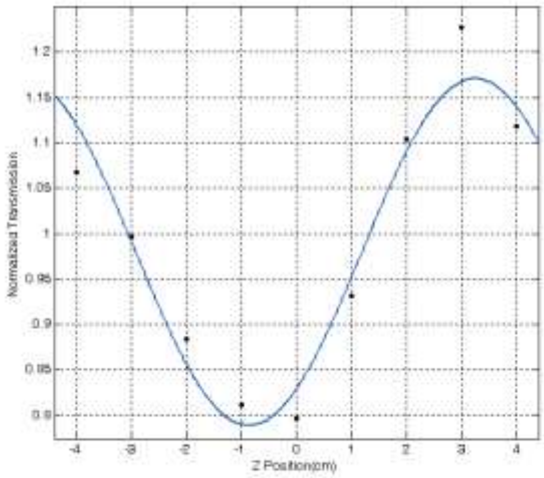

Fig. 5: Z-scan graph in the closed aperture

(6)

$$\Delta T_{p-v} = 0.406(1-S)^{0.25}\left(\frac{2\pi}{\lambda}\right)n_2 I_0 L_{eff}$$

ΔT_{p-v} is the distance between the valley and the passing graph from the aperture and I_0 is the intensity in the focused sample.

(7)

$$I_0 = \frac{2p}{\pi\omega^2}$$

Also S is the linear transition which is obtained by the following relationship.

(8)

$$S = 1 - exp\left(\frac{-2r_a^2}{\omega_a^2}\right)$$

Where r_a is the aperture beam and the beam radium in w_a aperture, L_{eff} is the effective length which was calculated through the following relationship [16]

(9)

$$L_{eff} = \frac{(1-e^{-\alpha L})}{\alpha}$$

Finally, α is the absorption coefficient in low laser powers and is calculated through the following relationship [16]

(10)

$$\alpha = -\frac{1}{L}\mathrm{Ln}\left(\frac{\mathrm{P}}{\mathrm{P}_0}\right)$$

Where P is the output power with sample and P_0 is the output power without the sample. The initial power of laser in without the sample is 45 mW. In this experiment, the laser beam intensity is $8 \times 10^3 \frac{K\omega}{m^2}$.

According to the figure 5, since a peak is observed in the curve after the valley, it can be inferred that the non-linear refractive index is positive and actually self-focusing phenomenon has been occurred here.

Here for the calculation of non-linear phase shift, the data obtained from the results of the closed aperture data is divided by open aperture data. The laser used in the experiment has nm532 wavelength and diffraction length Z_0 is 3.68 mm.

The results are shown in Table 1 for β and n_2.

Table 1: The nonlinear absorption coefficient and refractive index

$n_2 \frac{m^2}{\omega}$	$\beta \frac{m}{\omega}$	α_{mm}^{-1}
0.81×10^{-5}	0.407×10^{-3}	0.325

If the non-linear refractive index material is on the order of $10^{-7} - 10^{-5}\frac{cm^2}{\omega}$ and the sample is placed under a continuous laser solution, it could be argued that the origin of non-linear effects for the refractive index is thermal.

CONCLUSION

In this paper, using the z sweeping test, both nonlinear refractive index and two-photon absorption coefficient values were calculated. Thus, this graph of material has a positive nonlinear refractive index and self-focusing phenomenon has occurred in this case. In fact, when its thickness is smaller than the diffraction length, nonlinear refractive index of the sample is focused and makes it as a thin lens with a variable focal length. And also due to the value measured for nonlinear refractive index, it could be argued that the origin of nonlinear effects of the refractive index is thermal.

CONFLICT OF INTEREST
There is no any form of conflict of interest

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No financial sponsor in the form of person, institution or organization is involved in the present work.

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