Measurement of nonlinear refractive index of organic materials by z-scan

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Abstract. The nonlinear effects characterization by using the Z-Scan transmission technique in many materials has generated great interest according to the technological necessities. The majority part of the nonlinear effects can be described by the classic electromagnetic theory, with the electrical susceptibility in the constitutive equation that relates the electrical polarization with the electrical field. In this work the sign and refractive index magnitude and the nonlinear absorption coefficient of the following organic substance were determined: methylene-blue, rodamine LD, vegetable powder and gentian violet a hundred percent pure dissolved in isopropyl alcohol, a laser Nd: YAG was used as a source excitation. The bunch of laser was focused with a lens of ten centimeters of focal length; by using a displacement system the sweeping of twenty centimeters was realized. The following results of the normalized curves of the transmittance in function of the z position were obtained applying the Sheik-Bahae theory: The nonlinear refractive index of the dye shows an increase in function of its concentration and the power of exciting of the laser with negative nonlinear sign in the majority of the sample.

1. Introduction
With the creation of the laser in 1960 by Theodore H. Maiman [1,2] was possible to produce luminous beams with high intensities and when they interact with the matter, they presented behaviours that were not described by the theories before known, Fraken and his collaborators observed that a crystal of quartz emits light with the double of the frequency of the incident light that is transmitted by a laser of ruby creating in this way one of the areas of the physics, The nonlinear optic. Giving beginning to the study of the phenomenon that happen as consequence to the modification of the optical properties of a material when is exposed to high densities of energy.

2. Z–Scan
Z–Scan technique proposed by Sheik Bahae [3,4,5] and his collaborators is based on the spatial distortion of one beam due to the self-modulation of phase of the sample. It is one of the most used techniques by scientists who study the phenomenon related with the nonlinear optic in the determination of the positive no linearity (self–focalization) and negative (self–defocusing) in organic samples such as the dyes (methylene-blue, vegetable powder, rodamine LD and gentian-violet) in its pure state and dissolved in isopropyl alcohol.

In the basic configuration of the Z–Scan technique (Figure 1). The nonlinear refractive index n₂ of a sample is measured through the simple relation between the variation of the transmittance of the potency observed in a distant field and the distortion of phase induced acquired when it passes through
the sample. The potency transmitted through the aperture with radius \( r \) is captured when one Gaussian beam crosses a sample that moves along its propagation axis, the sample moves around the focal position to receive different intensities generating in the material one of the nonlinear response known as effect Kerr.

![Figure 1. Schematic diagram of experimental arrangement for the Z-scan technique.](image)

The linear effects are eliminated when the potency transmitted to a sample in a position \( Z \) is divided by the potency transmitted when the simple is located far away of the focus.

\[
T(Z) = \frac{P(Z)}{P(\infty)}
\]  

(1)

When the beam passes through the nonlinear sample suffers a change in its phase, this difference can be determined through the equation:

\[
\Delta \Phi_o = k n \omega L_{eff}
\]  

(2)

Where \( I_o \) is the initial intensity of the laser beam, \( L_{eff} \) is the thickness of the sample, \( S \) is the diameter of the aperture, \( P \) is the potency of the incident beam and \( k \) is vector of wave.

\[
\Delta T_{pv} = 0.406(1 - S)^{0.25} |\Delta \Phi_o|
\]  

(3)

\( \Delta T_{pv} \) is the difference between the transmittance pick-valley or valley pick and this variation amplitude of transmittance pick-valley is proportional to the distortion of phase [6,7,8].

\[
\Delta T_{pv} = 0.406|\Delta \Phi_o|
\]  

(4)

Starting from the equation (3) it can be determined and if \( S \ll 1 \) is easy to show that the nonlinear refractive index \( (n_2) \) is given by the following expression [9,10]:

\[
\Delta Z_{pv} = 1.7z_o
\]  

(5)

\[
z_o = \frac{k\omega_0^2}{2}
\]  

(6)

3. Experimental arrangement

The experimental arrangement is similar to the (Figure 1) A laser of Nd: YAG of emission continuous with a longitude of wave of 532nm, a stable potency of 50mW and a \( L_{eff} = 1mm \) was used. The minimum radius of the laser \( \omega_o = 63.04\mu m \) and \( I_o = 800.973 \text{ W/cm}^2 \) was determined.

The beam was focalized with a lens of 10cm of focal distance, the wide area detector was put 20cm from the position of the lens on the axis of sweeping \( Z \) and previously an iris of 0.01mm of diameter.
was placed in order to capture only the intensity around the optical axis getting in that way a better sensibility in the measurements. Cells with 1mm of thickness were used to measure the nonlinear properties of the dyes: methylene blue, vegetable powder, gentian violet and rodamine LD. Several concentrations dissolved in isopropyl alcohol were prepared, and the nonlinear refractive index was determined for each one of them.

4. Results and experimental analysis

The study in four different sample of dyes was done. Methylene-blue, vegetable powder, gentian-violet and rodamine LD. During the measuring, a potency of excitation of the laser beam of $50\,\text{mW}$ was kept and sweepings of $20\,\text{cm}$ with rising of $0.21\,\text{mm}$ in a bank of line displacement were carried out, the sweepings were performed of $+z$ to $-z$.

The Figure 2 correspondent respectively to the normalized curves of transmittance for: methylene-blue, vegetable powder, gentian-violet and rodamine LD.

Using the equations (5) and (6) can be obtained the value of $w_0$ in an experimental way.

$$\omega_0^2 = \frac{\Delta Z_{\text{pp}}}{1.7k} \quad (6)$$

These values were replaced in the equation (1) and solving, in this way we obtained:

$$n_2 = \frac{\Delta r_{\text{pp}}\Delta Z_{\text{pp}}d^2}{2.7608\pi PL_{\text{eff}}} \quad (7)$$

Figure 2. shows normalized curves of the transmittance in function of distance $Z(\text{cm})$ for the dyes.

The methylene-blue by using a concentration of $0.02$ dissolved in $12\,\text{ml}$ of isopropyl alcohol with a value of the nonlinear refraction index $(n_2)$ of $6.43084\times10^{-8}$ and negative sign of $(n_2)$. For gentian-violet was used a concentration of $0.0002$ dissolved in $106\,\text{ml}$ of isopropyl alcohol with a refraction index of $5.7014\times10^{-7}$ and positive sign, in the rodamine LD was used $0.0058$ dissolved in $18\,\text{ml}$ of isopropyl alcohol, the refraction index was calculated $5.9495\times10^{-7}$ and the negative sign and finally the refraction index for $0.1308$ vegetable powder was calculated $2.99869\times10^{-7}$.

We studied the concentration of $0.03\,\text{M.B}$ with potency´s variations of laser, and we obtained this results: In $30\,\text{mW}$ we obtained a refraction´s index $1.54 \times 10^{-11}$, we increased the potency´s laser in 40
and 50 mW, obtaining this results respectly $8.79261 \times 10^{-8}$ and $8.84293 \times 10^{-8}$. This results shows a increase of nonlinear refractive index, as shown in Figure 3 and in Table 1.

![Figure 3. Variation of nonlinear refractive index with the incident beam intensity for the methylene-blue.](image)

| Dye                  | $\Delta T_{ps}$ | $\Delta Z_{ps}$(cm) | $n_2$(cm$^2$/W)   |
|----------------------|-----------------|---------------------|------------------|
| Blue of methylene    | 0.179839        | 5.46274             | $6.43084 \times 10^{-8}$ |
| Vegetable powder     | 0.665161        | 6.88702             | $2.99869 \times 10^{-7}$ |
| Rodamine LD          | 0.837419        | 10.8534             | $5.9495 \times 10^{-7}$ |
| Violet gentian       | 1.41396         | 6.15986             | $5.7014 \times 10^{-7}$ |

5. Conclusion

The nonlinear properties of the dye were characterized through the technique Z–Scan for $\lambda = 532 nm$. The normalized curve of transmittance for the nonlinear refractive index, magnitude and sign for different concentrations of the dye dissolved in ethyl alcohol were obtained showing negative nonlinear refractive index or positive according the cases. The obtained results demonstrate a variation in the nonlinear optical properties of the dyes.

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