Rafidain Journal of Science
Vol. 30, No. 4, pp. 32-38, 2021

DOI: 10.33899/rjs.2021.169975

https://rsci.mosuljournals.com

ISSN: 1608-9391
e-ISSN: 2664-2786

Received:5/6/2021
Accepted:30/7/2021

Nonlinear Optical Properties of ZnO Thin Film at Low Laser Intensity Using Z-Scan Technique

Tharaa J. Polus
Department of Physics/ College of Education for Pure Sciences/ University of Mosul
E-mail: tharaasony23@gmail.com
Thoalfiqar A. Zaker
Department of Physics/ College of Education/ University of Al-Hamdaniya/ Nineveh
E-mail: thoalfiqar.physics@uohamdaniya.edu.iq
Ammar T. Zakar** * Hala N. Mohammed**
Department of Physics/ College of Education for Pure Sciences/ University of Mosul
***E-mail: Ammar_za24@uomosul.edu.iq**
****E-mail: Hala.n.m@uomosul.edu.iq**

ABSTRACT

In this work, a highly sensitive well-known z-scan technique was employed to study the nonlinear optical properties of Zinc Oxide thin films as a function of low laser fluencies. The transmissions of the continues-wave red laser diode with wavelength of (650 nm) were measured from the ZnO thin film sample with thickness of (425nm). The thin film used in this study was deposited on the glass substrates based on atmospheric pressure chemical vapor deposition (APCVD) technique. The measurements were obtained at low laser powers ranging from (1.9-2.5) mW. The results indicated that the nonlinear absorption coefficient, refractive index and the third-order nonlinear optical susceptibility increase with increasing the laser intensity. The obtained curves of the closed aperture showed a positive sign of the nonlinear refractive index which in turn attributed to the self-defocusing of the material. The optical parameters obtained in this work are relatively comparable with that obtained elsewhere. The results also confirm the reliability of the z-scan approach even at low laser intensities.

Keywords: Z-scan technique, CW laser, ZnO thin film, nonlinear absorption, nonlinear refraction.

INTRODECTION

The nonlinear optical (NLO) materials have an important role in nonlinear optics (Arivuoli *et al*., 2015) as it changes the optical behavior of the material (De Araújo *et al*., 2016). Such materials have long been recognized to interact with light to induce a nonlinear property (Suresh *et al*., 2012). The nonlinear optical characteristics of semiconductors are the most important properties in terms of having a wide bandgap, that makes them a good choice for using in NLO-based devices (rimpan *et al*., 2008). Specifically, ZnO nanoparticles have gained the researchers interest over the years (Walden *et al*., 2020). This material has a direct bandgap of 3.37 eV and sturdy binding energy of 60 meV at room temperature (Walden *et al*., 2020; Abed *et al*., 2015; Hussain, 2008). Further, it is considered as a proper candidate in the short wavelength optoelectronic applications (Shanshool *et al*., 2016). The ZnO's optical transitions use different configurations such as optical reflection, absorption, spectroscopy, transmission.

A numerous technique has been employed to study the optical characteristics of the semiconductor materials. Some of these techniques have been used previously to determine the nonlinear refraction (NLR), that is also known as the nonlinear Kerr effect (Walden *et al*., 2020; Bundulis *et al*., 2019) and the nonlinear absorption (NLA). The optical properties associated with NLA include multi-photon, absorption, multi-photon filament formation, saturable absorber, Q-switching and optical limiting (Garmire, 2013). The Z-scan method is one of them, the most sensitive results can be obtained from it (Jabbar *et al*., 2018; Sheik-Bahae *et al.*, 1990). There are wide forms of z-scan approach including "EZ-Scan", "Excite-Probe z-scan", "White Light z-scan" and many more (Garmire, 2013). This technique has been further improved by Mansoor Sheik-Bahae *et al*. (Sheik-Bahae *et al*., 1990). This approach has attracted a wide and rapid interest as it is considered as one of the established techniques used to study the nonlinear properties of the semiconductor and the semiconductor derivative materials. Based on the proper configuration, whether it is open or closed aperture, the related optical properties of the material can be extracted. In the case of the closed aperture, the incident beam produces a phase distortion inside the sample leading to a distortion in the beam's amplitude due to the sample nonlinearity, while in the case of open aperture, the change in the laser intensity is due to the TPA process (Neethling, 2005).

The purpose of this study is to investigate the nonlinear optical properties of ZnO thin film based on the widely used z-scan approach. The transmission emerging from the thin film was studied at different laser fluencies for both aperture configurations. Since the z-scan technique is extremely sensitive, the measurements were obtained at low laser intensities to overcome the heating contribution. Based on the open and closed aperture configurations, the nonlinear properties including refractive index as well as the TPA were obtained at different laser flounces.

EXPERIMENT SETUP

The widely known z-scan approach is used to extract the nonlinear optical parameters of the ZnO material based on open aperture and closed aperture. Zinc Oxide thin films with one concentration prepared as follow, the precursor solution (concentration = 0.5 mol/L) was prepared via dissolving high purity Zinc acetate dihydrate ($Zn\ (CH_3COO)_2.2H_2O$) as the source of Zn metal. The thin films were deposited on the glass substrates using the chemical vapor deposition technique at atmospheric pressure, which is named also as the atmospheric pressure chemical vapor deposition (APCVD) technique. Substrates were initially cleaned by acetone, ethanol and distilled water for 15 min in each solution to eliminate all the contaminants. Then, these substrates were dried in a stream of hot air. These substrates were deposited for 20 min at temperature of 500 °C, by APCVD technique. Further, the experimental details can be found elsewhere (Mohammed, 2019). The as-grown ZnO thin films were light rainbow in appearance. The thickness of grown film was estimated to be 425 nm, using the Balance method.

To investigate the nonlinear properties of the sample, the linear absorption was measured from the material absorbance at different wavelength with the use of an EMC-11D-V spectrophotometer, where its wavelength range is 400-800 nm and wavelength accuracy of ±2 nm. From the following equation we can obtain the linear absorption coefficient (Borhani Zarandi and Amrollahi Bioki, 2017):

$$\alpha_{\circ} = 2.303\frac{A}{L} \qquad \text{.............................(1)}$$

Where, $\alpha_{\circ}$ is linear absorption coefficient; A is the material's absorbance and L is the sample's thickness.

We used a red laser diode of CLASS IIIA, a continues-wave (CW) laser with wavelength of (650nm). A CW laser means that the laser continuously emits a steady beam of light with a constant power (Tkachenko, 2006). The I-P characteristics of the laser diode is also measured as shown in the Fig. (1). The maximum power of the laser was around 3 mW allowing its use in the low power investigations. The figure also shows that the threshold current is approximately 30 mA.

The traditional z-scan geometry shown in the Fig. (3) which is used to measure the transmission. In order to focus the laser beam on the sample, a collimating lens with focal length of (30 mm) was used. The beam waist radius ($w_{\circ}$) is measured using edge knife technique and found to be ~27 μm. The Rayleigh length of Z_R= (3.52 mm) is also calculated based on the following equation (2):

$$Z_R = \frac{\pi w_{\circ}^2}{\lambda} \qquad \text{..............................(2)}$$

Where, Z_R is the Rayleigh length, $w_{\circ}$ is the beam waist radius at focus, and λ is the laser wavelength.

In order to measure the transmission as at each sample position, the sample was mounted on the Arduino controlled rotational stage. The power of the transmitted beam is measured using a Si photo detector for both open and closed aperture. The aperture diameter in the case of the closed aperture was fixed around 0.5 mm. The power of the laser beam was changed using the current controller with accuracy of ±0.01 mA. Also, the temperature controller with resolution of about ±0.05 °C was used to stabilize the temperature of the laser allowing a temperature-controlled operation of the laser diode.

RESULTS AND DISCUSSION

The measurements of both linear absorption coefficient and the material's absorbance were taken at different wavelength, see Fig. (2). They both exhibit a decreasing in their values with increasing the wavelength.

To obtain the experimental data, the setup shown in the Fig. (3) was employed. In the case of open aperture configuration, the transmission of the laser beam was measured at different sample positions and different laser intensities as shown in the Fig. (4). It can be seen from the figure that the transmission reaches the minimum when the sample reaches the focus. The reduction of laser intensity could be attributed to the increases in the laser fluence at the focus. Thus, any deviation in the intensity of the transmitted radiation is due the multi-photon-absorption process which is in our case the (TPA). To obtain the nonlinear absorption coefficient β, the results shown in the Fig. (4) were fitted using the expression below (Sheik-Bahae *et al.*, 1990).

$$T(z) = [1 + q_{\circ}(z)]/q_{\circ}(z) \qquad \text{.................(3)}$$

Here:

$$q_{\circ} = \frac{\beta I_{\circ}\left[\frac{1-\exp(-\alpha_{\circ}L)}{\alpha_{\circ}}\right]}{1+(\frac{Z}{Z_{\circ}})^2} \qquad \text{.................(4)}$$

Where, $I_{\circ}$ is the laser intensity at the focus which can be calculated according to the expression $I_{\circ} = \frac{2P}{\pi w_{\circ}^2}$.

The open aperture exhibited a valley shape, this indicates a positive value of the nonlinear absorption coefficient β. This change is attributed to the TPA process, in which β increases with increasing the optical intensity of the laser beam, see (Table 1).

Fig. (5), shows the closed aperture measurements. The nonlinear refractive index n_2 is extracted by fitting the transmission data to the equation (5) (Sheik-Bahae *et al.*, 1990).

$$T(z) = 1 + \frac{4\Delta\phi(I_\circ)_\circ^{(3)} \cdot \frac{z}{z_\circ}}{\left[\left(\frac{z}{z_\circ}\right)^2 + 9\right] \cdot \left[\left(\frac{z}{z_\circ}\right)^2 + 1\right]} \quad \ldots\ldots\ldots\ldots\ldots(5)$$

Here:

$$\Delta\phi(I_\circ)_\circ^{(3)} = K\Delta n \left[\frac{1-\exp(-\alpha_\circ L)}{\alpha_\circ}\right] \quad \ldots\ldots\ldots\ldots\ldots(6)$$

Where, $\Delta\phi(I_\circ)_\circ^{(3)}$ is the phase shift introduced by the third-order nonlinear optical effects, $K = 2\pi/\lambda$ is the wave number, and $\Delta n = n_2 I_\circ$ refers to the nonlinear change in the refractive index.

The closed aperture showed a valley-to-peak behavior indicating a positive nonlinear refractive index n_2, ($n_2 > 0$). This in turn attributed to the self-focusing action which is a nonlinear process induced by the change in nonlinear refractive index. In such processes the nonlinear refractive index increases with increasing the optical intensity of the laser beam as can be noticed in the (Table 1). The observed noise in the data could be also due to low laser power (signal to noise ratio). It can be also observed that the data of the closed aperture contain traces for both nonlinear absorption coefficient and nonlinear refractive index respectively. This provides the possibility of determining the pure nonlinear refractive index by simply dividing the data of closed aperture on the open aperture counterpart.

The measurements for both the imaginary and real part of nonlinear susceptibility were obtained using the equations (7) and (8), respectively (Zidan and Allahham, 2015).

$$Im(\chi)^{(3)} = \left(\frac{10^{-2} n_\circ^2 \varepsilon_\circ c^2 \lambda}{4\pi^2}\right)\beta \quad \ldots\ldots\ldots\ldots\ldots(7)$$

$$Re(\chi)^{(3)} = \left(\frac{10^{-4} n_\circ^2 \varepsilon_\circ c^2}{\pi}\right) n_2 \quad \ldots\ldots\ldots\ldots\ldots(8)$$

Where, $n_\circ$ refers to the linear refractive index of the material, $\varepsilon_\circ$ is the permittivity of the vacuum, c is the speed of light in vacuum.

The total third-order nonlinear optical susceptibility ($\chi^{(3)}$) can be also estimated according to the following expression:

$$|\chi^{(3)}| = \left[\left(Re(\chi^{(3)})\right)^2 + \left(Im(\chi^{(3)})\right)^2\right]^{\frac{1}{2}} \quad \ldots\ldots(9)$$

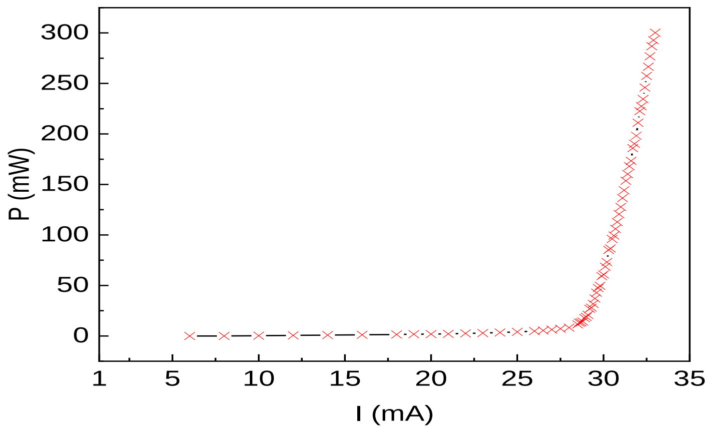

Fig. 1: I-P characteristics for the laser diode.

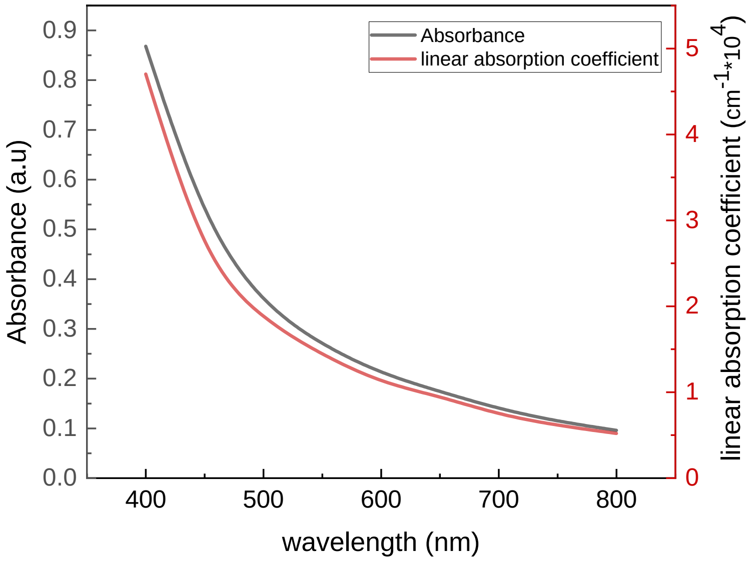

Fig. 2: Illustrated the absorption spectra and the linear absorption coefficient of ZnO thin films.

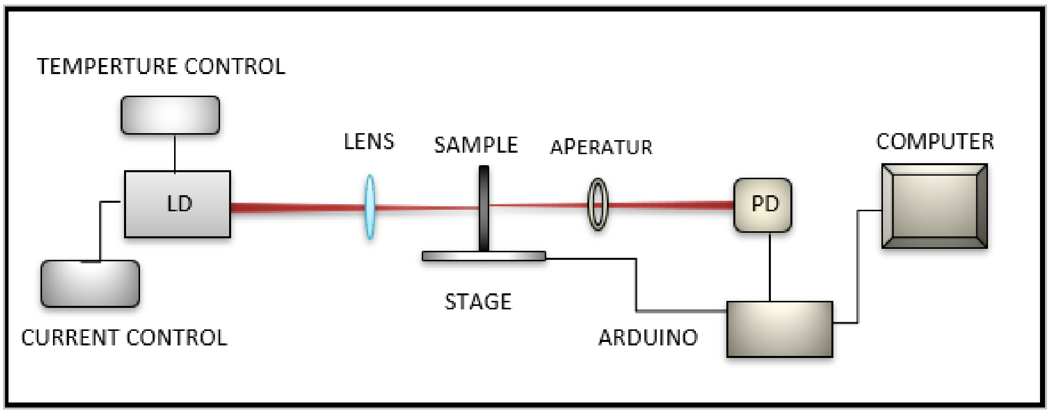

Fig. 3: Shows the experimental setup for z-scan. The rotational stage is used to move the sample on the axis.

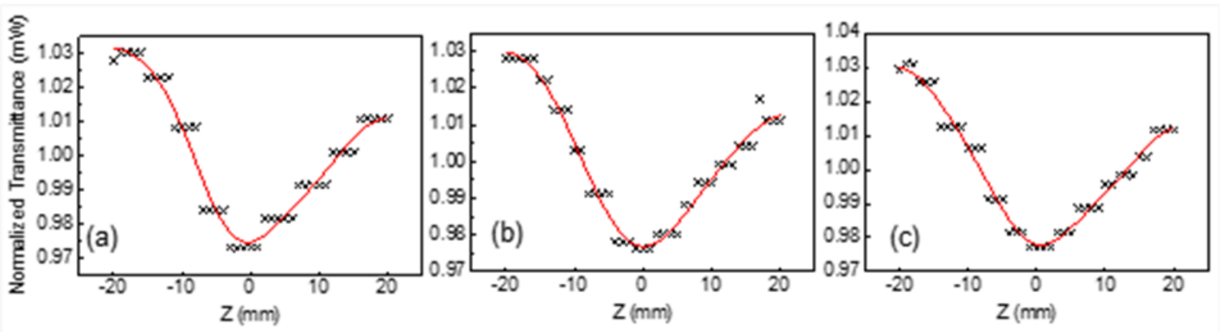

Fig. 4: Illustrates the normalized transmittance curves at open-aperture z-scan for ZnO thin films at different powers. (a) $P = 1.9$ mW; (b) $P = 2.2$ mW; (c) $P = 2.5$ mW.

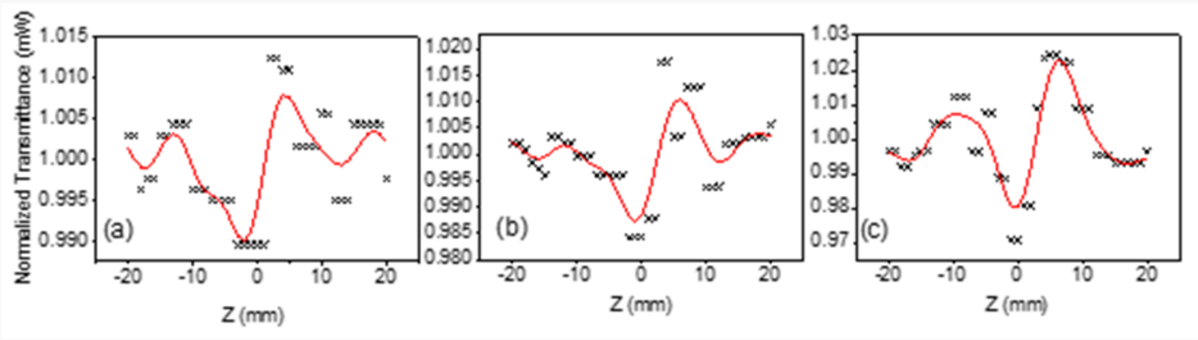

Fig. 5: Illustrates the normalized transmittance curves at closed-aperture z-scan for ZnO thin films at different powers. (a) $P = 1.9$ mW; (b) $P = 2.2$ mW; (c) $P = 2.5$ mW.

Table 1: indicates the extracted values of the nonlinear optical parameters of the ZnO thin film.

Powers of laser P mW	Laser intensity at each power I_o W/cm^2	Nonlinear absorption coefficient β cm/W	Nonlinear refractive index n_2 cm^2/W	Imaginary part of nonlinear susceptibility Im $(\chi^{(3)})$ esu	Real part of nonlinear susceptibility Re $(\chi^{(3)})$ esu	nonlinear optical susceptibility $(\chi^{(3)})$ esu
1.9	166.01	9.2039	0.101E-3	0.486E-2	10.33E-3	1.141E-2
2.2	192.22	10.391	0.126E-3	0.546E-2	12.887E-3	1.399E-2
2.5	218.83	10.5511	0.18E-3	0.558E-2	18.41E-3	2.152E-2

CONCLUSIONS

A simple z-scan technique was performed to measure the value of the nonlinear absorption and the value and sign of the nonlinear refractive separately. We have extracted the data at different low powers of laser operating at wavelength (650 nm) as an excitation source. The results indicates that ZnO thin films exhibits positive sign for the nonlinear refraction and two-photon absorption, which is referred to occurring the self-focusing effect and TPA process, respectively.

REFERENCES

Abed, S.; Bouchouit, K.; Aida, M.S.; Taboukhat, S.; Sofiani, Z., Kulyk, B.; Figa, V. (2016). Nonlinear optical properties of zinc oxide doped bismuth thin films using Z-scan technique. *Optical Materials.*, **56**, 40-44.

Arivuoli, D. (2001). Fundamentals of nonlinear optical materials. *Pramana.*, **57**(5), 871-883.

Borhani Zarandi, M.; Amrollahi Bioki, H. (2017). Effects of cobalt doping on optical properties of ZnO thin films deposited by sol–gel spin coating technique. *J. Optoelectronical Nanostructures.*, **2**(4), 33-44.

Bundulis, A.; Mihailovs, I.; Rutkis, M. (2020). Origin of the Kerr effect: investigation of solutions by polarization-dependent Z-scan. *JOSA B.*, **37**(6), 1806-1811.

De Araújo, C.B.; Gomes, A.S.; Boudebs, G. (2016). Techniques for nonlinear optical characterization of materials: a review. *Reports on Progress in Physics.*, **79**(3), 036401.

Garmire, E. (2013). Nonlinear optics in daily life. *Optics Express.*, **21**(25), 30532-30544.

Hussain, S. (2008). Investigation of structural and optical properties of nanocrystalline ZnO.

Irimpan, L.; Nampoori, V.P.N.; Radhakrishnan, P.; Krishnan, B.; Deepthy, A. (2008). Size-dependent enhancement of nonlinear optical properties in nanocolloids of ZnO. *J. Applied Physics.*, **103**(3), 033105.

Jabbar, A.S.; Abbas, S.I.; Al-zubaidy, M.H. (2018). Studying the linear and nonlinear optical properties by using Z-Scan technique for CdS thin films prepared by CBD technique. *Appl. Phys. Research.*, **10**(3), 7-13.

Mohammed, Y. H. (2019). Fabrication of n-MgZnO/p-Si heterojunction diode: Role of magnesium doping. *Superlatt. and Microstruct.*, **131,** 104-116.

Neethling, P.H. (2005). "Determining Non-Linear Optical Properties Using The Z-Scan Technique". Doctoral dissertation, Stellenbosch: University of Stellenbosch.

Shanshool, H.M.; Yahaya, M.; Yunus, W.M.M.; Abdullah, I.Y. (2016). Using z-scan technique to measure the nonlinear optical properties of PMMA/ZNO nanocomposites. *J. Teknologi.*, **78**(3).

Sheik-Bahae, M.; Said, A.A.; Wei, T.H.; Wu, Y.Y.; Hagan, D.J.; Soileau, M.J.; Van Stryland, E.W. (1990). Z-scan: a simple and sensitive technique for nonlinear refraction measurements. In *Nonlinear Optical Properties of Materials.*, **1148**, 41-51. International Society for Optics and Photonics.

Sheik-Bahae, M.; Said, A.A.; Wei, T.H.; Hagan, D.J.; Van Stryland, E.W. (1990). Sensitive measurement of optical nonlinearities using a single beam. *IEEE J. Quantum Electronics.*, **26**(4), 760-769.

Suresh, S.; Ramanand, A.; Jayaraman, D.; Mani, P. (2012). Review on theoretical aspect of nonlinear optics. *Rev. Adv. Mater. Sci*., **30**(2), 175-183.

Tkachenko, N.V. (2006). ″Optical Spectroscopy: Methods and Instrumentations″. Nikolai V. Tkachenko, Finland, Elsevier., pp. 39-59.

Walden, S.L.; Fernando, J.F.; Shortell, M.P.; Jaatinen, E.A. (2020). Accurate determination of nonlinear refraction in ZnO and Au composite nanostructures. *Optical Materials Express.*, **10**(2), 653-661.

Zidan, M.D.; Allahham, A. (2015). Z-Scan measurements of the third-order optical nonlinearity of a C60 doped poly (dimethylacetylendicarboxylate). *Acta. Physica. Polonica. A.*, **7**(1), 128.

الخصائص البصرية اللاخطية لغشاء ZnO عند الشدات الواطئة لليزر باستخدام تقنية Z-Scan

الملخص

باستخدام تقنية عالية الحساسية وهي المسح على المحور (Z-scan) تم قياس الخصائص الغير الخطية لرقائق أوكسيد الخارصين (ZnO) كدالة للشدات الواطئة. باستخدام ليزر ذو طول موجي مستمر عند (650 nm) تم حساب النفاذية للرقاقة عند سمك قدره (425 nm). تم تحضير العينة من خلال ترسيبها على طبقة من الزجاج باستخدام تقنية ترسيب البخار الكيميائي عند الضغط الجوي (APCVD). القياسات تم اخذها عند الطاقات القليلة ضمن المدى (1.9–2.5 mW). النتائج أظهرت زيادة كل من معامل الامتصاص والانكسار غير الخطيين والحساسية الغير الخطية من الرتبة الثالثة مع زيادة شدة الضوء المسلط. كل من معاملات الانكسار والامتصاص غير الخطيين امتلكا قيم بإشارات موجبة وهذه القيم الموجبة تشير الى ظهور كل من تأثير التركيز البؤري البصري اللاخطي (self-focusing) بالنسبة لمعامل الانكسار غير الخطي والى ظهور عملية الامتصاص ثنائية الفوتون (TPA) بالنسبة لمعامل الامتصاص غير الخطي. هذه النتائج قابلة للمقارنة بشكل نسبي مع تلك التي تم التوصل اليها في بحوث أخرى. النتائج اكدت ايضا دقة وموثوقية منظومة Z-scan التي تم استخدامها بالرغم من الشدات الواطئة لليزر المستخدم.

الكلمات الدالة: تقنية المسح على المحور، ليزر الموجة المستمرة، رقائق أوكسيد الخارصين، الامتصاص غير الخطي، الانكسار غير الخطي.