

## **Diffraction patterns and nonlinear optical properties of Henna oil**

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### **Abstract**

The diffraction ring pattern and Z-scan techniques are used to estimate both the nonlinear refractive index,  $n_2$ , and nonlinear absorption coefficient,  $\beta$ , of Henna oil using low power continuous wave (CW) visible laser beam. The values of  $n_2$  and  $\beta$  are obtained of  $1.72 \times 10^{-7} \text{ cm}^2/\text{W}$  and  $1.21 \times 10^{-3} \text{ cm/W}$  respectively.

**Keywords:** Self-phase modulation, Diffraction ring pattern, Z-scan technique.

## نماذج حيود وصفات لاختية في زيت الحناء

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### الخلاصة

تم استخدام تقنيتين أنموذج حلقات الحيود والمسح بالاتجاه  $Z$  لتقدير قيمتي كل من معامل الانكسار اللاخطي  $(n_2)$  ومعامل الامتصاص اللاخطي  $(\beta)$  لزيت الحناء باستعمال حزمة ليزر مستمرة منخفضة القدرة . كانت قيمتي  $\beta$  و  $n_2$  على التوالي  $1.21 \times 10^{-3} \text{ cm/W}$  و  $1.72 \times 10^{-7} \text{ cm}^2/\text{W}$  .

## 1. Introduction

The search for nonlinear optical (NLO) materials with large optical nonlinearities and fast response is essential for potential applications in optical signal processing, all optical switching, optical limiting [Kovsl et al.(1999), Henariand and Cassidy (2012)], ultrafast optical communication, optical storage [Kurian et al.(2002), Zon et al.(2015)] etc. Though many kind of nonlinear optical materials have been extensively studied, searching for large nonlinear optical response is an going matter. The most forgettable, naturely occurring materials with potential applications are vegetable oils. These materials are available in local markets, cheap, available in liquid phase so they do not need solvents, etc. Generally vegetable oils have industrial and medical applications. The only available work on vegetable oils, to our believe, is that of Zamiri et al. (2012) on palm oil, and by Abed-Ali and Emshary (2017,2017) on paprika and pepper oils.

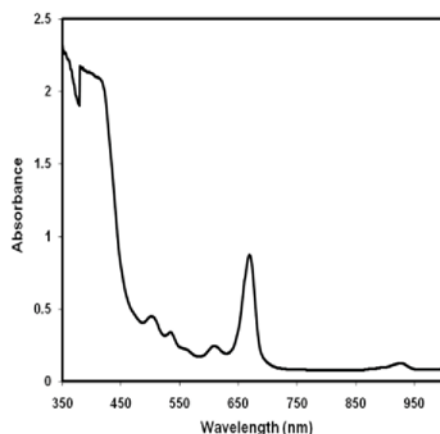
Numerous techniques are known for the measurement of nonlinear refraction in different materials. Among these techniques and the easiest are the diffraction ring patterns [Ogusu et al.(1996)] the Z-scan [Sheik-Bakae et al.(1990)] and thermal lens [Mahdieh and Jafarabadi (2012)]. Each one of these techniques required a laser with low variable CW power, single or two positive lenses, a semitransparent screen, a power meter and a cell contain the sample.

In this article we present the result of estimation of nonlinear index,  $n_2$ , the total change in refractive index,  $\Delta n$ , and the nonlinear absorption coefficient,  $\beta$ , via the use of the first two techniques and a visible, 473 nm, laser CW beam of low power.

## 2. Experiment

### 2.1 UV- visible spectroscopic study

The linear absorption spectra of Henna oil is shown in Fig. 1. The UV-vis (Ultraviolet-Visible) absorption spectra of the sample was recorded at room temperature using UV- visible spectrophotometer (Jenway-England-6800) in the spectral range (350 – 900 nm).



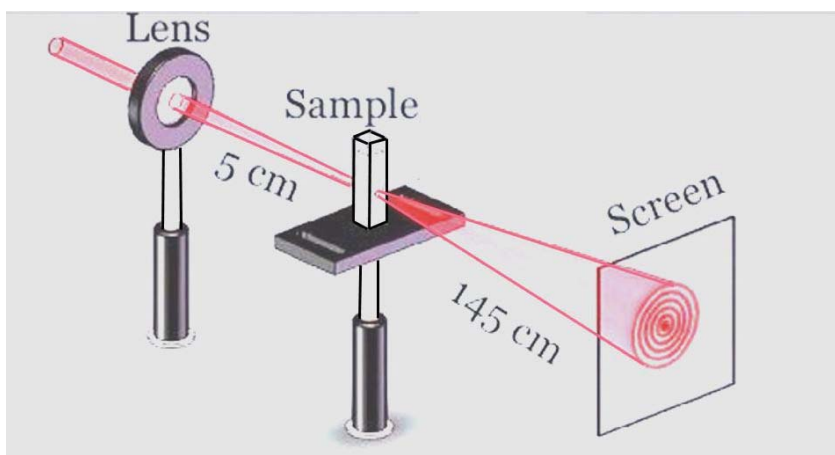
**Fig.1.** UV-vis absorption spectrum of Henna oil.

## 2.2 Diffraction rings technique

The experimental setup comprised a CW laser and a positive 5 cm focal length glass lens to focus the laser beam at the entrance of the sample cell of thickness 1 mm. A 30 x 30 cm semitransparent cell 100 cm from the cell to cast the diffraction ring patterns. The input power was measured using a digital multi-wavelength multi-range meter. The experimental arrangement is shown in Fig. 2.

## 2.3 Z-scan technique

The single beam Z-scan technique was used to measure the nonlinear susceptibility of the samples. This method allows the simultaneous measurement of both nonlinear refractive index and nonlinear absorption coefficient. Basically, the method consists of translating a sample through the focus of a Gaussian beam and monitoring the changes in the far field intensity pattern. When the intensity of the incident laser beam is sufficient enough to induce nonlinearity in the sample, it either converges the beam (self-



**Fig. 2.** Experimental set-up for diffraction rings technique.

focusing) or diverges (self-defocusing) depending on the nature of that nonlinearity. By moving the sample through the focus, the intensity dependent absorption is measured as a change of the transmittance through the sample (open aperture). The nonlinear refraction is determined by the spot size variation at the plane of a finite aperture detector combination (closed aperture), because the sample itself acts as a thin lens with varying focal length as it moves through the focal plane.

The Henna oil was placed in the 1 mm quartz cuvette, for nonlinear optical measurements. The nonlinear refraction and nonlinear absorption coefficient were measured with a CW laser light with 473 nm from a solid state laser (SDL). The spatial profile of the optical beam was Gaussian. The laser beam was focused by a lens of 5 cm focal length. The radius of the beam waist was measured to

be 22.19  $\mu\text{m}$  (half-width at  $1/e^2$  maximum). The incident and transmitted powers were measured simultaneously by photo detectors fed to digital power meters (Field Max II-To+OP-2 Vis Sensor). The NLO properties of the samples were manifested by moving the samples along the axis of the incident beam (z-direction) with respect to the focal point . An aperture of 2.5 mm in radius was placed in front of the detector to assist the measurement of the self-defocusing effect.

### 3. Results and discussion

#### 3.1 The absorption coefficient ( $\alpha$ )

The absorption coefficient ( $\alpha$ ) was obtained directly from the absorbance against wavelength curve using the relation [El-Fadl et al. (2005)]

$$\alpha = 2.303 \frac{A}{L} \quad (1)$$

where L is the sample thickness and A is the absorbance.

The values of absorption coefficient ( $\alpha$ ) at wavelength 473 nm for Henna oil have been calculated using Eq. 1 and the result are given in Table 2.

#### 3.2 Diffraction ring patterns measurement

For the experimental set mentioned above and from input powers (30 mW, 40 mW, 49 mW, 64 mW) diffraction ring patterns appeared instantaneously post irradiation where the number of rings in each pattern increases with the increase of input power as can be seen in Fig. 3. It was noted too that the diameter of outermost ring in each pattern increases with increase of input power.

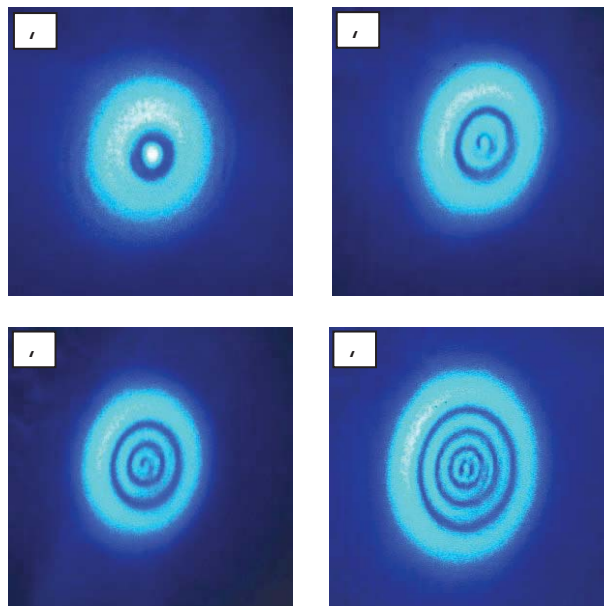


Fig. 3. Images of the far field diffraction rings at input laser power passing through the sample cell of (a) 30 mW, (b) 40 mW, (c) 49 mW, (d) 64 mW.

### 3.4 Estimation of the nonlinear refractive index

For each ring appeared in each pattern, the phase of the beam traversing the sample increase by  $(2\pi)$  so that for each pattern of N rings the total phase shift of the laser beam passing through that sample cell,  $\phi$ , can be written as [Ogusu et al.(1996)]

$$\phi=2\pi N \quad (2)$$

For a sample thickness, L, and total path length  $\Delta = \Delta nL$ , where  $\Delta n$  is the change in refractive index of the sample so that

$$\phi=k\Delta \quad (3)$$

$\Delta n$  can be written as  $n_2I$ , I is the laser beam input intensity ( $=2P/\pi\omega^2$ (P is the laser light power and  $\omega$  is the beam spot size at the entrance of the sample cell which is related the beam spot size ,  $\omega_0$ , of the laser beam falling on the positive lens by  $\omega=1.22f\lambda/\omega_0$ , f is the positive lens focal length  $f=5\text{cm}$ )). For  $\lambda=473 \text{ nm}$ ,  $\omega_0=0.15 \text{ cm}$ ,  $\omega=19.235\mu\text{m}$ . From equation (2) and (3) one can arrive at the following equations for  $\Delta n$  and  $n_2$  i.e

$$\Delta n=\lambda N/L \quad (4)$$

$$n_2=\Delta n/I \quad (5)$$

From the numerical given and input power of 30 mW, 40 mW, 49 mW and 64 mW, the magnitudes of  $\Delta n$  and  $n_2$  are given in Table (1)

Table (1) Change of refractive index,  $\Delta n$  , and nonlinear refractive index,  $n_2$ , for Henna oil using four input powers.

I (W/cm <sup>2</sup> )	Number of rings, N	$\Delta n \times 10^{-6}$	$n_2 \times 10^{-6}$ (cm <sup>2</sup> /W)
5161.9	1	473	0.916
6882.6	2	946	1.37
8431.6	3	1414	1.68
11012	4	1892	1.72

### 3.5 Simulating the ring patterns

The experimentally observed ring patterns are simulated theoretically using a well-known model based on the Fresnel-Kirchhoff diffraction [Chavez-Cerda et al. (2006)]. As a result of using a laser

beam with Gaussian distribution TEM<sub>00</sub>, the electric field of the laser beam at the entrance of the medium can be written as

$$E = E_0 \exp\left[-\frac{r^2}{w_0^2}\right] \exp\left[-ikz - i\frac{k}{2R}(z^2 - R^2)\right] \quad (6)$$

where  $r$  is the radial coordinate,  $z$  is the medium coordinate position,  $k$  is the free space wave vector,  $n_0$  is the air surrounding the medium refractive index,  $w_0$  is the beam waist at the medium entrance, and  $R$  is the radius of curvature of its wave-front in the position. So that the intensity of the beam passing through the medium and falling on a screen  $D=145$  cm, from the sample cell relative to the radial coordinate ( $\rho$ ) (see Fig. 4) can be written as

$$I(\rho) = I_0 \left(\frac{\rho}{D}\right)^2 \exp\left[-\frac{2k}{D} \left(\frac{\rho^2}{2} - R D\right)\right] \quad (7)$$

where  $J_0(x)$  is zero order Bessel function of the first kind,  $\theta$  is the far field diffraction angle,  $\rho$  is the radial coordinate in the far field plane of observation and the intensity  $I(\rho)$  can be written as [12]:

$$I(\rho) = I_0 \left(\frac{\rho}{D}\right)^2 \exp\left[-\frac{2k}{D} \left(\frac{\rho^2}{2} - R D\right)\right] \quad (8)$$

$D$  is related to the radial coordinate,  $\rho$  by  $\rho = D \theta$  as can be seen in Fig. 4.  $\alpha$  is the absorption coefficient of the sample at the wavelength  $\lambda$ .

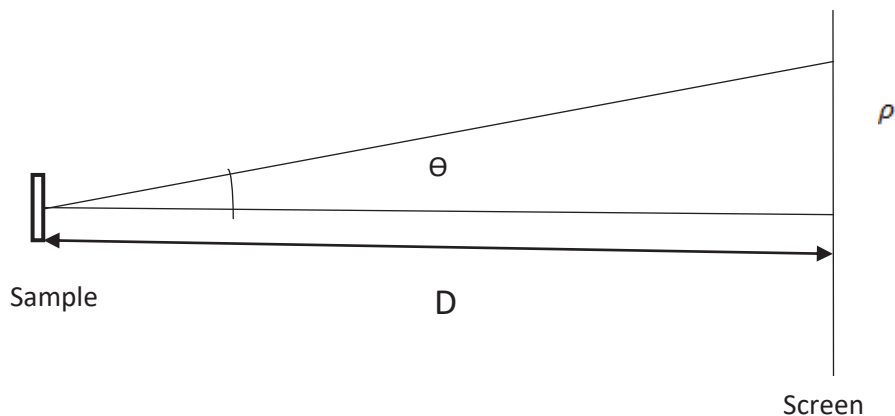
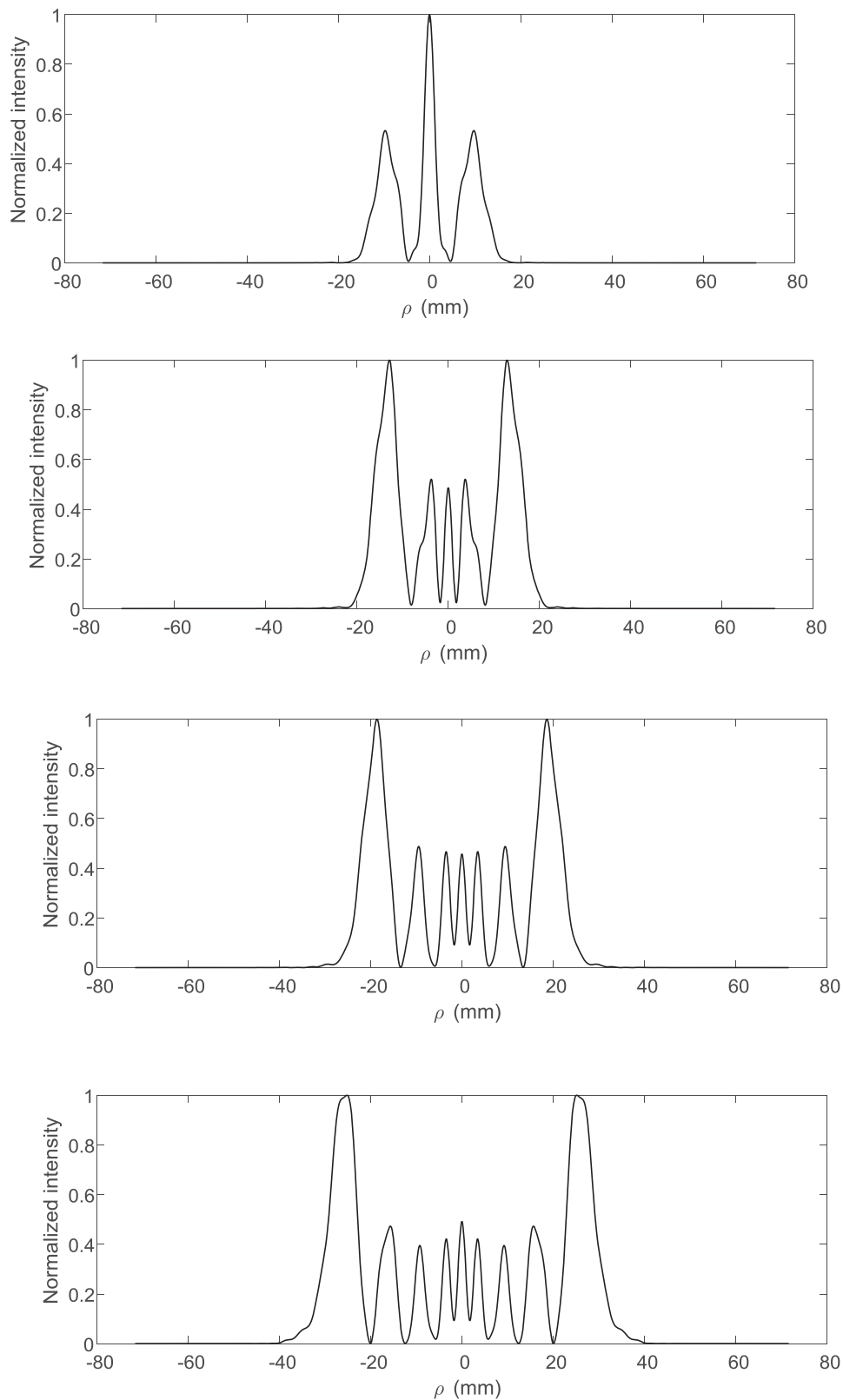


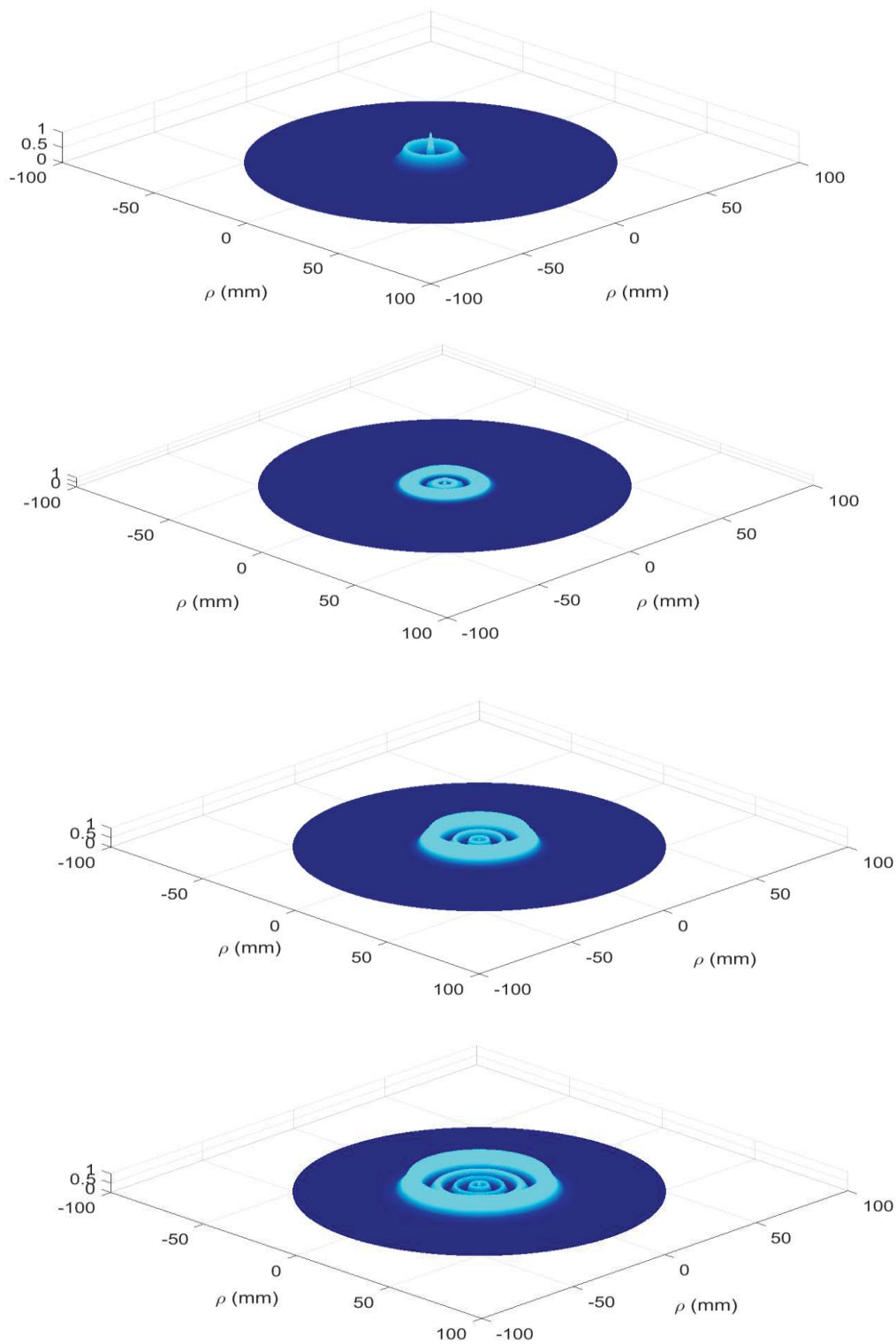
Fig .4. Definition of experimental  $\theta$ ,  $D$  and  $\rho$  used in the theoretical analysis of the diffraction patterns.



(a)

Fig.5: Calculated (a) 1D and (b) 3D intensity distribution in ring patterns in the far field.





(b)

The results are shown in Fig.5 for one (a) and three (b) dimensional where it can be seen the reasonable agreement between theory and experiment (Fig.5 and 3).

### 3.3 Z-scan

The nonlinear absorption coefficient,  $\beta$ , of the Henna oil is evaluated under an open aperture configuration. Fig. 6a shows normalized open aperture transmittance of the Henna oil. A symmetric bell shaped transmission is measured with a minimum at the focus ( $z = 0$ ). The minimum transmittance at the focus is due to the nonlinear absorption.

The nonlinear optical absorption data obtained under the conditions used in this study can be well described by Eq. (9), which describes a nonlinear optical absorptive process [Vinitha and Ramalingam (2008)]

$$\beta = \frac{2\sqrt{2}\Delta T}{I_o L_{eff}} \quad (9)$$

where  $\Delta T$  is the one-valley transmittance value in the open aperture Z-scan curve ,  $L_{eff} = [1 - \exp(-\alpha L)]/\alpha$  is the effective thickness of the sample.

In the Z-scan experiment, a focused Gaussian light beam is usually used. The medium with nonlinear refractive index causes an additional, depending on  $z$ , focusing (at positive  $n_2$ ) or defocusing (at negative  $n_2$ ) of the light beam. It changes the power of the beam passing through a small aperture placed in the far field when moving the medium along  $z$  (closed-aperture Z-scan). As a result, “peak-valley” (self-defocusing ) or “valley-peak” (self-focusing ) like dependences of the transmittance versus  $z$  are obtained from which it is possible to determine the sign and magnitude of the nonlinear refractive index [Sheik-Bakae et al.(1990)].

Closed-aperture Z-scan transmittance curve of the Henna oil is shown in Fig. 6b. The peak to-valley structure of the closed-aperture Z-scan curve in Fig. 6b obviously implies that the Henna oil possesses the negative nonlinear refraction property (defocusing). The peak and the valley are asymmetric (i.e., there is a suppressed peak and an enhanced valley) owing to the presence of the nonlinear absorption effect.

In Z-scan measurement, the transmittance of the sample measured without an aperture (open aperture scan) gives information on purely nonlinear absorption coefficient whereas the apertured scan (closed aperture) contains the information of both the nonlinear absorption coefficient and nonlinear refractive index nonlinearities. The ratio of the normalized closed aperture and open aperture scans generates a Z-scan due to the purely nonlinear refractive index [Sheik-Bakae et al.(1990)], and results are shown in Fig. 6c.

The peak to valley transmittances difference,  $\Delta T_{p-v}$ , is a measure of the nonlinear refractive index as it is linearly related to the spatial phase distortion  $|\Delta\phi_0|$  of the beam, which is due to the nonlinear refractive index. The relation is given by [Sheik-Bakae et al.(1990)]

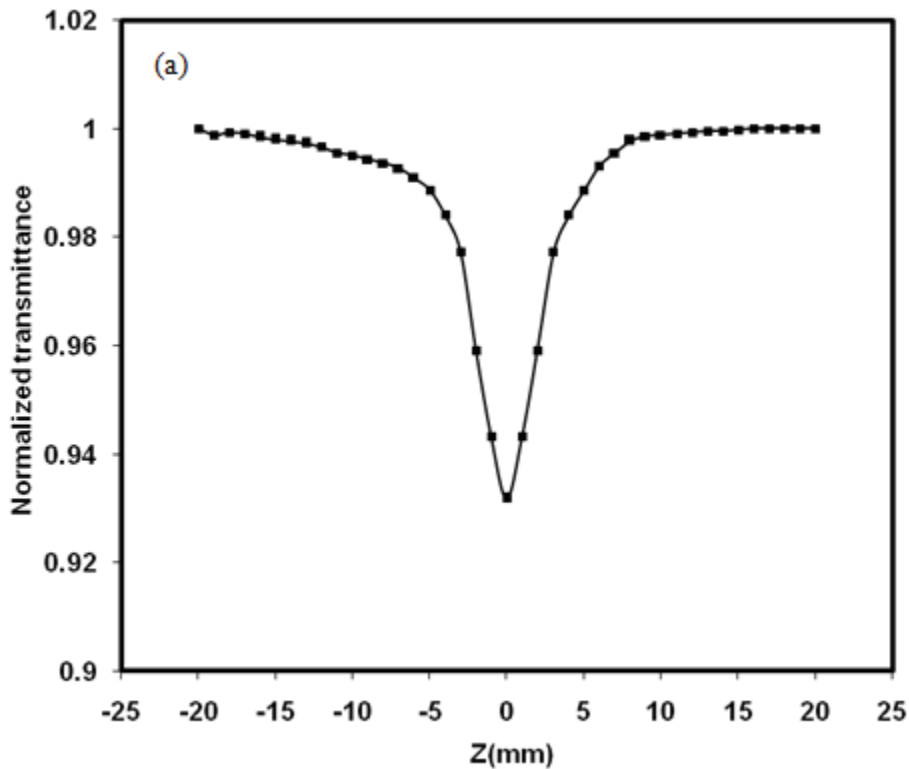
$$\Delta T_{p-v} = 0.406(1 - S)^{0.25} |\Delta\phi_0| \quad (10)$$

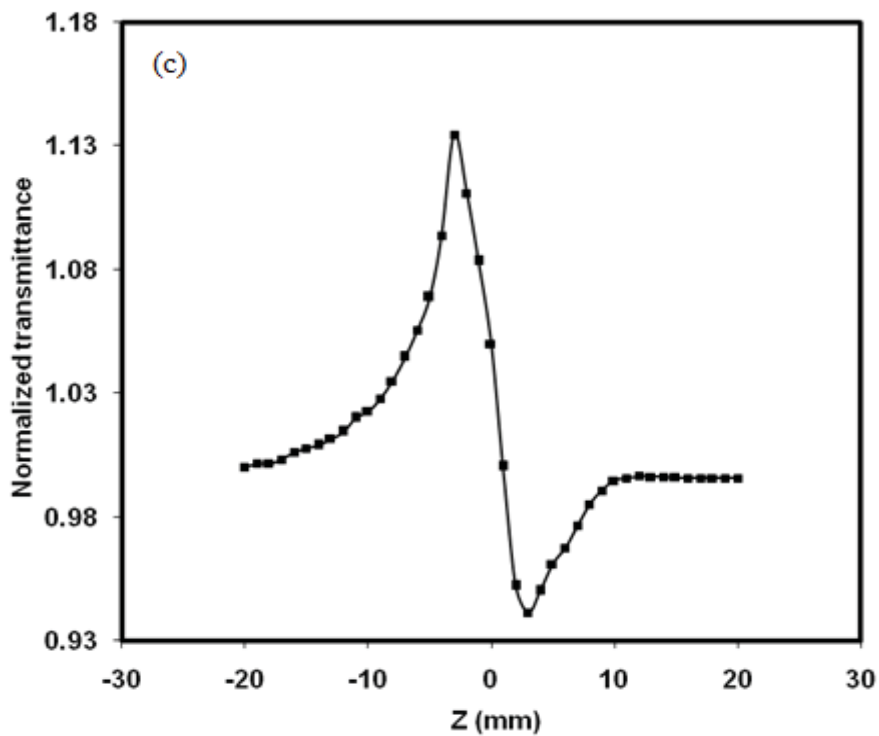
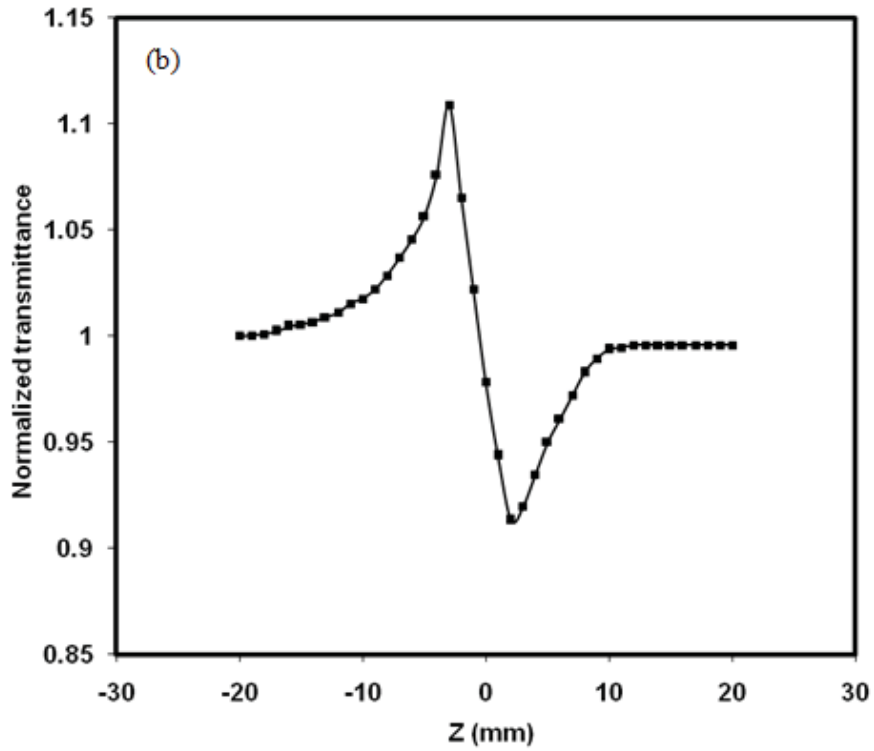
where  $|\Delta\phi_0|$  is the on-axis phase shift at the focus,  $S=1- \exp (-2r_a^2/\omega_a^2)$  is the aperture linear transmittance with  $r_a$  denoting the aperture radius and  $\omega_a$  denoting the beam radius at the aperture in the linear regime. The nonlinear refractive index is then given by

$$n_2 = \frac{\Delta\phi_0 \lambda}{2\pi L_{eff} I_0} \quad (11)$$

where  $\lambda$  is the laser wavelength and  $I_0$  is the incident intensity.

The nonlinear absorption coefficient,  $\beta$ , (cm/W) and nonlinear refractive index,  $n_2$ , (cm<sup>2</sup>/W) for Henna oil are calculated from the open and closed aperture normalized transmittances in Figs.6a and c respectively, their values are listed in Table 2.





**Fig. 6.** Z-scan data of Henna oil (a) open aperture scan (b) closed aperture scan and (c) the division of b by a, at 20 mW incident power.

Table (2) Nonlinear optical parameters for Henna oil using Z-scan techniques at 2587.1 W/cm<sup>2</sup> incident intensity.

Sample	$\alpha$ (cm <sup>-1</sup> )	Z-scan $\beta \times 10^{-3}$ (cm/W)	$n_2 \times 10^{-7}$ (cm <sup>2</sup> /W)
Henna oil	10.66	1.21	0.25

#### 4. Conclusion

In conclusion, we observed multiple diffraction rings of a CW laser light post passing through a Henna oil. We counted 4 rings for oil at an optical intensity of 64 mW. The number of diffraction rings linearly increased with intensity. We obtained large values of nonlinear refractive index  $n_2 = 1.72 \text{ cm}^2/\text{W}$  by self-diffraction technique. We also investigated the nonlinear refractive index and nonlinear absorption coefficients of Henna oil using the Z-scan technique at the wavelength of 473 nm. The results indicate that Henna oil exhibits self-defocusing nonlinearities. This large nonlinearity is attributed to a thermal effect resulting from linear absorption. These results show that Henna oil has potential applications in nonlinear optics.

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