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# Investigating the Nonlinear Optical Response of Pepper Oil under Low Power Irradiation with Continuous Wave Visible Laser Beam

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***Abstract* - The nonlinear optical properties of pepper oil are studied by diffraction ring patterns and Z-scan techniques with continuous wave beam from solid state laser at 473 nm wavelength. The nonlinear refractive index of the sample is calculated by both techniques. The sample show high nonlinear refractive index. Based on Fresnel-Kirchhoff diffraction integral, the far-field intensity distributions of ring patterns have been calculated. It is found that the experimental results are in good agreement with the theoretical results. Also the optical limiting property of pepper oil is reported. The results obtained in this study prove that the pepper oil has applications in nonlinear optical devices.**

***Keywords*- Vegetable oil, Nonlinear refractive index, Diffraction ring patterns, Z-scan, Fresnel-Kirchhoff diffraction integral.**

## I. Introduction

The varieties of organic materials nonlinear optical properties have been studied extensively for the possible use in various photonic applications. Two properties viz., high nonlinear refractive index and fast response time these medium must have when considering them to be used in optoelectronic and photonic devices (Dneprovskii *et al.*, 1988; Ali *et al.*, 2006; Ali and Palanisamy, 2007; Manickasundaram *et al.*, 2008,2008,2011; Abdulkader *et al.*, 2018; Al-Mudhaffer *et al.*, 2016).

Numbers of physical mechanisms are responsible for the change of refractive index of materials in liquids reorientation of anisotropic molecules can occur under strong laser light beam irradiation that leads to changes of refractive index. In isotropic liquids, molecular redistribution can occur that leads to nonlinearities together with the electronic polarizability of electronic clouds. Resonance effects viz., two-photon absorption and saturation can also lead to changes of refractive index. Thermal effects can also leads to nonlinear refractive index (Zhang *et al.*, 2013).

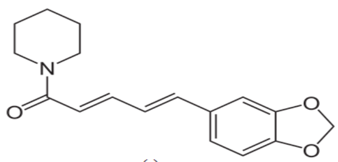
Among the many organic materials that were tested for their nonlinear properties, vegetable oils were almost forgotten by the nonlinear optics society, only one attempt published by (Zamiri *et al.*, 2012) in 2012. In the last two years extensive works were directed towards the study of nonlinearities in vegetable oils where in excess of 20 vegetable oils shown to behave nonlinearly under low power irradiation with visible, continuous wave, (cw), single mode laser beams (Sultan *et al.*, 2018; Hassan, 2017; Abed-Ali and Emshary, 2017; Hassan *et al.*, 2017). Vegetable oils are cheap, worldwide available, and do not need solvents, shows high nonlinear refractive index (up to $10^{-6}$ $cm^2/W$).

The present work aims to finding a material that have high nonlinear properties. So in this article, the total change in refractive index and the nonlinear refractive index of pepper oil are evaluated using diffraction ring pattern technique (Gordon *et al.*, 1965) together with Z-scan technique (Sheik-Bahae *et al.*, 1990). The optical limiting property is studied too. All these properties were obtained using visible, 473 nm, low power, single mode laser beam. It is_also compared the nonlinear refractive index value obtained by the Z-scan method of pepper oil with the value of the material known to have a high nonlinear refractive index.

## II. Experimental

### 1. Sample

The pepper (Black pepper) oil used in this work is available in the local markets, its chemical structure and chemical formula are shown in Fig.(1). Table 1 shows some




(a)

(b) $C_{17}H_{19}NO_3$

Fig.(1). Chemical structure (a) and chemical formula (b) of physical properties of the pepper oil.

Table (1): Some physical properties of pepper oil.

| | |
|---|---|
| Polarity | Nonpolar |
| Density | 0.873 gm/ml |
| Linear refractive index | 1.379 -1.488 at 20C° |

**2.Spectroscopic Study of Pepper Oil**

The UV-visible spectroscopy of the pepper oil was conducted using Jenway, England 6800 spectrophotometer at room temperature where the oil was in a 1 mm thickness quartz cell, in the spectral range (400-900 nm). The spectrum is shown in Fig.(2). The absorption coefficient, α, of the pepper oil is evaluated using the Fig. 2, at 473 nm and the formula (El-Fadl et al., 2005)

$$\alpha = 2.303\frac{A}{d} \qquad (1)$$

Where A and d are the pepper oil absorbance at 473 nm and its thickness respectively. A value of α = 55.3 $cm^{-1}$ is obtained.

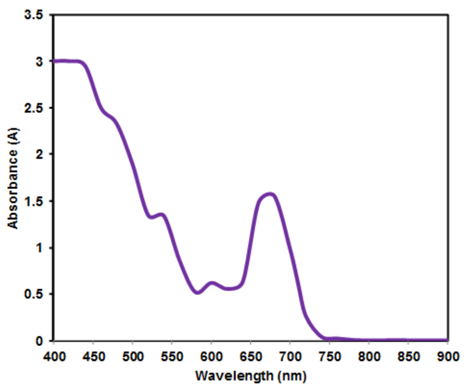



Fig. 2. The absorbance spectrum of pepper oil.

**2.Experimental set-up**

The experimental set-up used to produce diffraction ring patterns comprised of a solid state type SDL-473-050T laser device emits a blue 473 nm laser beam with Gaussian, $TEM_{00}$, intensity distribution. A sample cell made of glass of 1 mm thickness, a power meter type SDL-PM002 to measure the input power falling on the sample and a 5 cm focal length glass convergent lens to focus the laser beam on the sample cell were used. A 30 x 30 cm semitransparent screen used to cast the diffraction ring patterns obtained and a digital camera type Sony DSC- T99-8700-82-25 mm was used to register each pattern.

The Z-scan measurements were conducted using the same set-up which was used in diffraction pattern technique except replacing the screen with a power meter covered with a 2 mm diameter circular aperture. The sample was fixed on a translation stage to sweep the sample cell across the 5 cm focal point of the lens (±z). This technique is called closed-aperture Z-scan. For the open aperture Z-scan measurements the circular aperture usually replaced with a convergent lens to collect the entire transmitted beam from sample cell.

The optical limiting measurement was conducted using the same set-up used earlier except fixing the sample cell beyond the lens focal point i.e., at the valley position of the closed-aperture Z-scan technique and increasing the input power on the sample and measuring the total transmitted beam.

**III. Results**

**1. Diffraction ring pattern technique**

Fig.(3) shows sample results of the variation of the diffraction ring patterns against input power. Where it can be seen from Fig.(3) that the area of each pattern and number of rings for each pattern increases with the increase of input power, and the outer most ring in each pattern is the most intense in power than the inner ones, an indication of self-defocusing occurring in the pepper oil. Fig.(4) shows the temporal evolution of one selected diffraction ring pattern. This is an indication that each ring pattern does not born instantaneously. Fig.(5) shows the effect of the wave front of the laser beam approaching the sample on the diffraction pattern i.e., convergent wave front when the sample at 1 cm before the focal point of the lens and divergent wave front when the sample at l cm beyond the focal point of the lens (Deng et al., 2005).

**3. Z-scan technique**

Closed aperture Z-scan trend is shown in Fig.(6) where it can be seen that a peak transmittance followed by a valley one an indication once more that the nonlinearity is based on self-defocusing i.e. negative nonlinear refractive index. The reason for self-defocusing of the laser beam is of thermal origin. As a result of the use of a continuous wave laser in the current study this lead to warming of the sample due of the absorption of laser energy by pepper oil and this causes a spatial Gaussian distribution of temperature in the pepper oil and this lead to a spatial variation of the refractive index, that acts as a thermal lens resulting in phase distortion of the propagating beam. The open aperture Z-scan scan resulted in a straight horizontal curve indicating that the sample does not have nonlinear absorption coefficient at incident intensity at 473 nm.According to thermal lens model the value of nonlinear refractive index, $n_2$, can be obtained from the difference between the transmittances at peak and valley, $\Delta T_{p-v}$, for pepper oil using Eq. (2) (Sendhil et al., 2005):

$$n_2 = \frac{\Delta T_{p-v}\lambda}{4\pi d\ I} \qquad (2)$$

*A.* where I(=1.721 $kW/cm^2$) is the laser beam intensity, so that $n_2 = 0.47 \times 10^{-8}\ cm^2/W$.

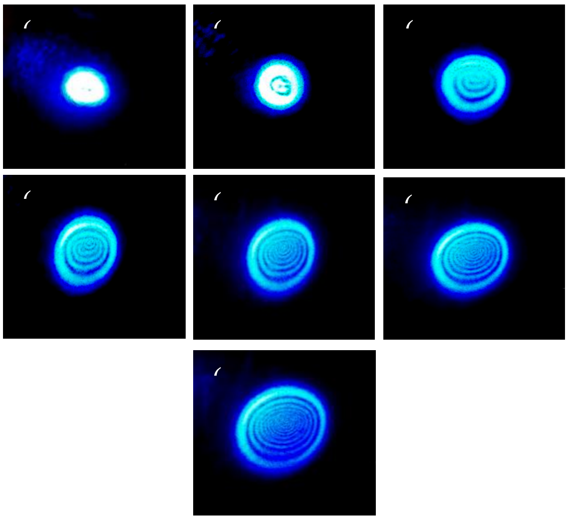


Fig.3. Far field diffraction ring patterns of pepper oil at (a) 7.5 (b) 16.5 (c) 24 (d) 30.5 (e) 35.5 (f) 42 (g) 50 mW input laser power.

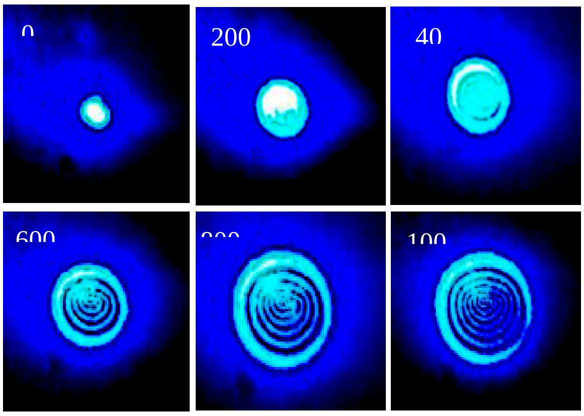



Fig.4. Temporal evolution of one diffraction ring pattern for pepper oil at input power of 50 mW.

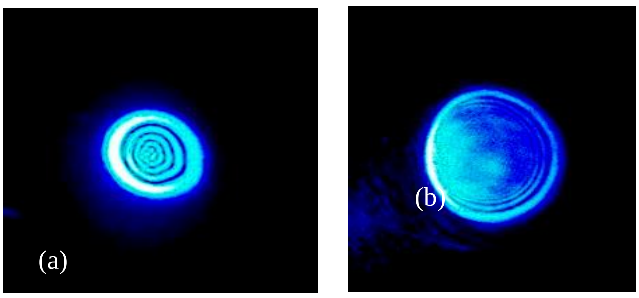



Fig.5. Far field diffraction rings pattern of pepper oil using input power of 50 mW for sample (a) 1 cm before the lens focus (b) 1 cm beyond the lens focus.

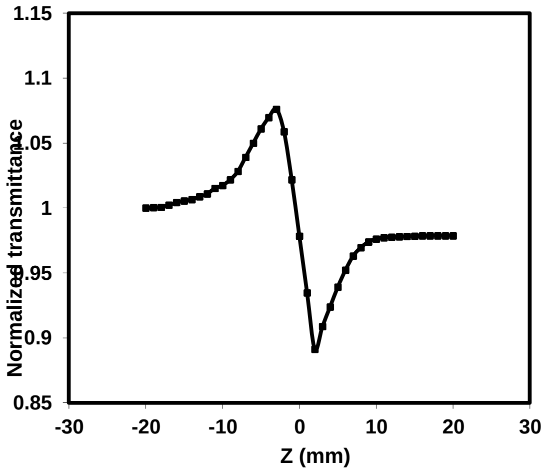



Fig.6. Closed aperture Z-scan data for the pepper oil.

## 4. Optical limiting measurements

Fig.(7) shows the relation between output power and input power where it can be seen that the curve reveal two outputs, a linear increase between input and output one followed by a constant output versus input. Fig.(8) is the normalized output or transmittance versus the input power. From this curve the limiting threshold value for pepper oil, which is defined as the value of the input power when the transmittance is reduced by half can be obtained. From Fig. (8) the limiting threshold value for pepper oil is 28 mW.

### 5. Calculation of the Total Change in refractive index, Δn, and nonlinear refractive index, $n_2$

Every ring born in each diffraction ring pattern is an indication of a change in phase, $\phi$, of the laser beam traversing the pepper of thickness, d, by $2\pi$

$$\Delta\phi = 2\pi N \qquad (3)$$

radians so that for $N$ rings to appear the total phase shift the laser beam, ($\Delta\phi$), suffers can be written as (Ogusu *et al.*, 1996):

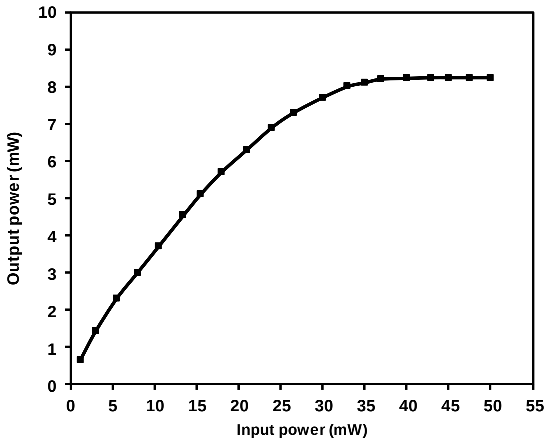



**Fig.7. Optical limiting of pepper oil.**

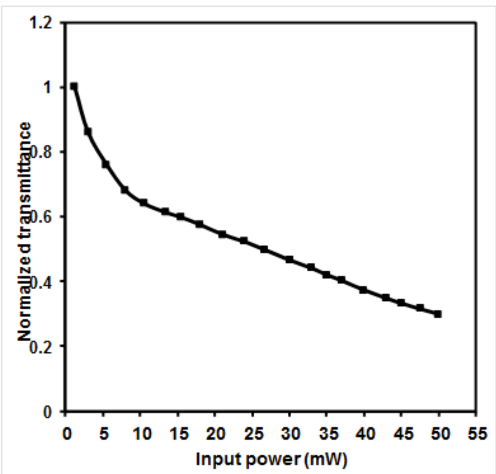



**Fig.8. Normalized transmission of optical limiting for pepper oil.**

The phase total change, $\Delta\phi$, can be related once more to the thickness of the sample thickness, *d*, as follows:

$$\Delta\phi = k\Delta \qquad (4)$$

$k$ ($=2\pi/\lambda$) is the laser beam wave vector and $\Delta$ is the change in the optical length that can be written as

$$\Delta = d\Delta n \qquad (5)$$

$\Delta n$ is the total change in refractive index so that

$$\Delta\phi = \frac{2\pi}{\lambda} d\Delta n \qquad (6)$$

From equations (3) and (6)

$$\Delta n = \frac{N\lambda}{d} \qquad (7)$$

I is the laser beam intensity that can be written as

*P* is the laser beam input power and $\omega'$ is the laser

$$I = \frac{2P}{\pi\omega'^2} \qquad (9)$$

$$\omega' = 1.22\frac{f\lambda}{\omega} \qquad (10)$$

beam radius falling on the sample where it can be related to the beam radius that fall on the lens ω by the following equation (Self , 1983)

f is the lens focal length, $\lambda$ = 473 nm, and ω can be related to the laser beam radius as it leaves the laser output coupler, $\omega_o$, by (Yariv, 1976)

*z* the distance between the laser output coupler and the

$$\omega = \omega_o[1 + (\frac{z}{z_o})^2]^{1/2} \qquad (11)$$

lens ($z$ = 40 cm), $\omega_o$ = 1.5 mm (at $1/e^2$) and $z_o$ can be written as follows (Yariv, 1976):

$$z_o = \frac{\pi\omega^2 n'}{\lambda} \qquad (12)$$

so that for $d$ = 1 mm, $n'$ in the refractive index of air, $P$ = 50 mW, $N$ = 9 , $\Delta n = 4.257 \times 10^{-3}$ and $n_2$ = $4.979 \times 10^{-7}$ $cm^2/W$. The difference between $n_2$ values due to diffraction ring patterns and Z-scan of two order of magnitude is due to the difference between input power levels used in both techniques.

**6. Modeling of the diffraction ring patterns**

It was mentioned that the laser beam used in this work have Gaussian, $TEM_{oo}$, wave front. Based on

$$E(r, z_o) = E(0, z_o)\exp(-\frac{r^2}{\omega_p^2})\exp(-\frac{ikn_o r^2}{2R}) \qquad (13)$$

this the Fresnel-Kirchhoff integral can be used to numerically reproduce the experimental diffraction patterns given in Fig.(3) Defining the electric field of

$$n_2 = \frac{\Delta n}{I} \qquad (8)$$

the laser beam at the entrance of the pepper oil cell as follows (Chavez-Cerda *et al.*, 2006):

$\omega_p$ is the laser radius (=$\omega$), k=2π/λ) is the laser beam wavelength, $R$ is the beam wave front radius, $n_o$ is the air refractive index and E(0,$z_o$ ) is the electric field falling on the sample cell. As the beam traverses the sample it suffers a total phase shift, $\phi(r)$, added to the Gaussian beam at a point (r) written as:

where

$$\phi(r) = k(\frac{n_o r^2}{2R}) + \Delta\varphi(r) \qquad (14)$$

$$\varphi(r) = \Delta\varphi_o \exp(-\frac{2r^2}{2R}) \qquad (15)$$

$$\Delta\varphi_o = k\Delta n(z_o, 0)d \qquad (16)$$

$$\Delta n(z, r) = n_2 I(z_o, r) \qquad (17)$$

$\Delta\varphi_o$ is the peak nonlinear phase-shift in the beam and d is the sample thickness. The total change of refractive index, Δ*n*, is related to the beam intensity, I, as follows:

The complex electric field, E(r,$z_o$+d), after the beam propagate along the nonlinear material is written as

$$E(r, z_o + d) = E(0, z_o)\exp(-\frac{r^2}{\omega_p^2}) \exp(-\frac{\alpha d}{d})\exp(-i\varphi(r) \qquad (18)$$

follows

The laser beam intensity, $I(\rho)$, falling on the screen situated in the far field after traversing the nonlinear pepper oil can written (see Fig.(9) as follows(Chavez-Cerda et al., 2006):

$$I(\rho) = I_o \left| \int_0^\infty J_o(k\theta(r)) \exp[-\frac{r^2}{\omega_p^2} - i\varphi(r)]rdr \right|^2 \qquad (19)$$

$$I_o = 4\pi^2 \left| \frac{E(o, z_o)\exp(\alpha d/2)}{i\lambda D} \right|^2 \qquad (20)$$

$I_o$ is the light intensity at the entrance of the sample cell, $J_o(x)$ is the zero order Bessel function, $\theta$ is the far-field diffraction angle. $I_o$ can be written as follows λ=473 nm, $\omega_p$=19.235 µm, $R$=30 cm, $d$= 0.1 cm, $D$= 75 cm, α = 55.3 $cm^{-1}$, and $\rho = D\theta$ , are given in Fig.(3).

To solve equation (19) Mat Lab system were used. Fig.(10) shows the numerically calculated intensity distributions of the laser beam in two dimension (left column) and in one dimension (right column) where it can be seen the accepted agreement compare to the experimental results shown in Fig.(3).

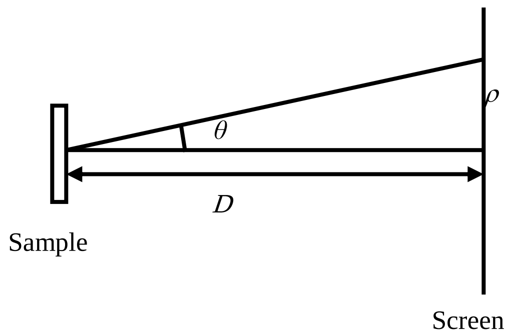



**Fig. 9. Definition of experimental *θ*, *D*, and *ρ* used in the theoretical analysis of the diffraction patterns.**

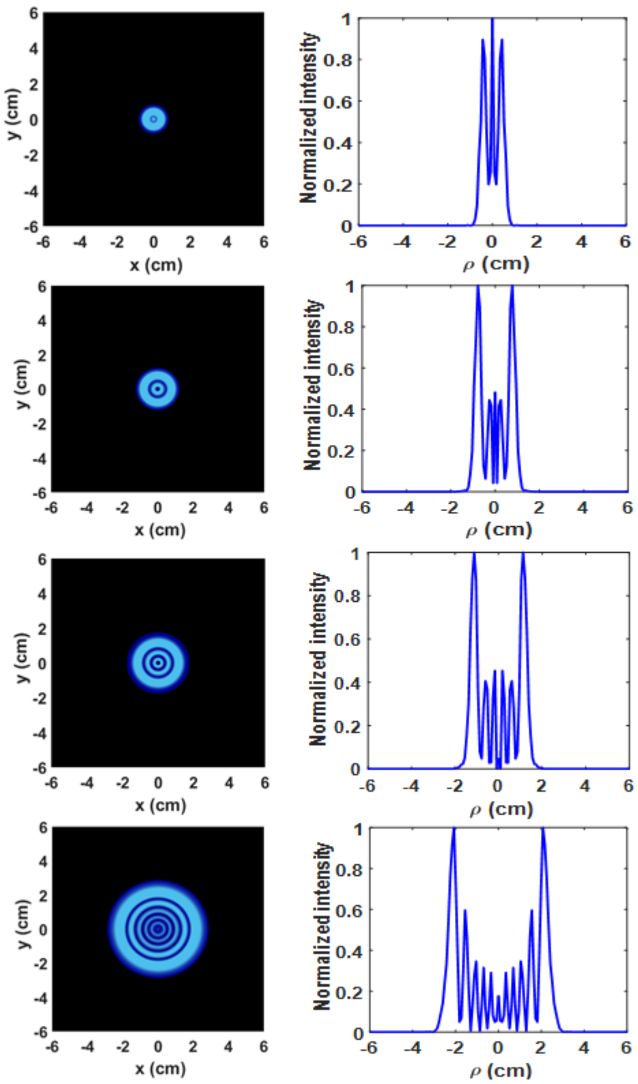
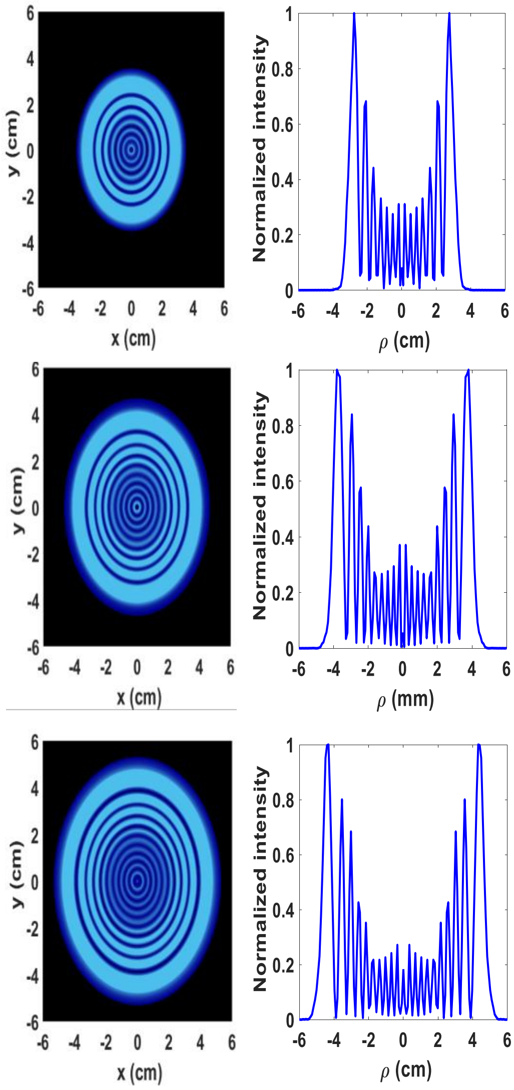



**Fig.10. Numerically calculated far-field intensity distribution of rings, left column is two dimensional and right column is one dimensional distributions at input power (mW): (a) 7.5 (b) 16.5 (c) 24 (d) 30.5 (e) 35.5 (f) 42 (g) 50 mW ,from top to bottom, Continue**




## IV. Conclusion

The nonlinear optical properties of pepper oil were investigated experimentally by the diffraction ring patterns and Z-scan techniques. It is observed that bright and dark rings resulted due to the passage of laser beam through pepper oil. It is found that the number of rings in each pattern and diameter of the outer most of rings in each pattern are intensity dependent. The far-field intensity distribution of rings are numerically calculated using Fresnel-Kirchhoff diffraction integral. Good agreement between theoretical and experimental results was obtained. The nonlinear refractive index of pepper oil was calculated via diffraction ring patterns and Z-scan techniques. When comparing the value of nonlinear refractive index which was calculated by Z-scan with the nonlinear refractive index value of some conventional materials such as acridine orange doped polymer film (Sukumaran and Ramalingam, 2005), 2,8-Phenazinediamine, N8, N8, 3-trimethyl hydrochloride (1:1) dye solution (Al-Ahmad *et al.*, 2012), azo dye[1-am ino-2-hydroxynaphthalin sulfonic acid-[3-(4-azo)]-4- amino diphenyl sulfone] (Al-Ahmad *et al.*, 2012), crystal violet dye doped poly(methylmethacrylate) (Sukumaran and Ramalingam, 2006) and phloxine B dye doped PMMA (Hassan *et al.*, 2013), it is worth noting that the nonlinear refractive index value of pepper oil is larger than those materials. Therefore, the objective of this research has been achieved by finding a material that possesses high nonlinear optical properties. The optical limiting property of pepper oil was investigated. Through the results shown by pepper oil, it is believed that this sample can be a promising candidate in the field of optical applications.

## V. References


Abdulkader, A.; Amjad F.; Hassan, Q.M.; Al-Asadi, M.; Ahmed S.; Bakr,H.; Sultan, H.A. and Emshary, C.A. (2018), Linear, nonlinear and optical limiting properties of carbon black in epoxy resin, Optik ,160 100–108.

Abed-Ali, I.T. and Emshary, C.A. (2017). Determination of nonlinear refractive index of

Paprika oil and pepper oil under CW visible laser illumination. J. Thi- Qar Univ. 12, 90-101.

Al-Ahmad, A.Y.; Hassan, Q.M.; Badran H.A. and Hussain, K.A. (2012). Investigating some linear and nonlinear optical properties of the azo dye (1-amino-2-hydroxy naphthalin sulfonic acid-[3-(4-azo)]-4-amino diphenyl sulfone), Opt. Las. Tech. 44,1450-1455.

Al-Ahmad, A.Y.; Badran H.A.; Hassan, Q.M.A. and Emshary, C.A. (2012).The influence of solvents on formation of self- focusing in organic dye using CW laser light, Adv. Appl. Sci. Res., 3 706-712.

Ali, Q. M. and Palanisamy, P.K. (2007), Optical phase conjugation by degenerate four-wave mixing in basic green 1 dye-doped gelatin film using He–Ne laser, Opt. Las. Tech. 39, 1262–1268.

Ali, Q. M., Palanisamy, A.; Manickasundaram, S. and Kannan, P. (2006), Sudan IV dye based poly(alkyloxymethacrylate) films for optical data storage, Opt.Commun.267236–243.

Al-Mudhaffer, M.F.; Al-Ahmad, A.Y.; Hassan, Q.M.A. and Emshary, C.A. (2016). Optical characterization and all-optical switching of benzenesulfonamide azo dye**,** Optik 127,1160–1166.

Chavez-Cerda, S.; Nascimento, C.M.; Alencar, M.A.; de Silva, M.G.; Meneghetti, M.R. and Hickmann, J.M. (2006). Experimental observation of the far field diffraction patterns of divergent and convergent Gaussian beans in a self-defocusing medium. Annals of optics, XXIX ENFMC

Dneprovskii, V.S.; Furtichev, A.I.; Klimov, V.I.; Shen, L.; Nazvanova , E.V.; Okorokov, D.K. and Vandishev, U.V. (1988). Nonlinear optical properties of CdS and optical bi-stability, Phys. Stat. Sol. (b) 150 ,839 - 844.

Deng, L.; He, K.; Zhou, T. and Li, C. (2005). Formation and evolution of far-field diffraction patterns of divergent and convergent Gaussian beams passing through self-focusing and self-defocusing media, J. Opt. A:Pure Appl. Opt.7,409-415.

El-Fadl, A.; Mohamad, A.; Galal, A.; El-Moiz, A.B. and Rashad, M. (2005). Optical constants of Zn1_xLixO films prepared by chemical bath deposition technique. Physica B, 366, 44-54.

Gordon, J.P.; Leite, R.C.; Moore, R.S.; Porto, S.P. and Whinnery, J.R. (1965). Longe transient effects in lasers with inserted liquid samples, J. Appl. Phys. 36,3-8.

Hassan, Q.M.A. (2017). Investigation on the nonlinear optical properties and optical power limiting of parsley oil, Int. J. Photo. Opt. tech. 3,17-20.

Hassan, Q.M. A.; Al-Ahmad, A.Y.; Al-Mudhaffer, M.F. and Badran H.A. (2013). Third-order optical nonlinearities and optical-limiting properties of phloxine b dye doped pmma films investigated by z-scan technique, Rom. J. Phys. 58 ,962-969.

Hassan, Q.M.A.; Sultan, H.A.; Baker, H..; Hashim D.H. and Emshary, C.A. (2017). Diffraction patterns and nonlinear optical properties of Henna oil, J. Coll. Edu. Pure Sci .7 90-103.

Manickasundaram, S.; Kannan, P.; Kumaran, R.; Velu, R.; Ramamurthy, P.; Hassan, Q.M.A.; Palanisamy, P.K.; Senthil, S. and Narayanan, S. (2011). Holographic grating studies i[n pendant xanthene dyes containing poly(alkyloxymethacrylate)s. J. Mater. Sci: Mater. Electron. 22 ,25–34.

Manickasundaram, S.; Kannan, P.; Hassan, Q.M. and Palanisamy, P.K. (2008). Azo dye based poly(alkyloxymethacrylate)s and their spacer effect on optical data storage, J. Mater Sci: Mater Electron.19,1045–1053.

Manickasundaram, S.; Kannan, P.; Hassan, Q.M. and Palanisamy, P.K. (2008). Holographic grating formation in poly(methacrylate) containing pendant xanthene dyes. Optoelec. Adv. Mater. Rap. Commun. 2 , 324-331.

Ogusu, K.; Kohtani,Y. and Sao, H. (1996). laser induced diffraction rings from an absorbing solution. Opt. Rev., 3, 232-234.

Self, S.A. (1983). Focusing of spherical Gaussian beams, Appl. Opt. 22,658–661.

Sendhil, K.; Vijayan, C. and Kothiyal, M.P. (2006). Low-threshold optical power limiting of cw laser illumination based on nonlinear refraction in zinc tetraphenyl porphyrin, Opt. Laser Technol. 38, 512–515.

Sheik-Bakae, M.; Said, A.A.; We,T.H.; Hegan, D.J. and Van Styland, E.W. (1990). Sensitive measurement of optical nonlinearities using a single beam. IEEE J. Quan. Elctron., 26, 760-769.

Sukumaran, V. and Ramalingam, A. (2005). Spectral characteristics and nonlinear studies of acridine orange dye, Phys. Lett., A 341, 454–458.

Sukumaran, V. and Ramalingam, A. (2006). Spectral characteristics and nonlinear studies of crystal violet dye, Spectrochim. Acta Part A 63 ,673–676.

Sultan, H. A.; Hassan, Q.M. A.; Bakr, H.; Al-Asadi, A.; Ahmed, S.; Hashim, D. H. and Emshary, C. A. (2018) .Thermal-induced nonlinearities in rose, linseed and chamomile oils using CW visible laser beam , Can. J. Phys. 96, 157-164.

Yariv, A. (1976). Introduction to optical electronics, Holt, Rinehart and Winston, Newyork, USA.

Zamiri, R.; Parvizi, R.; Zakaria, A.; Sadrolhosseini, A.R.; Zamiri, G.; Darrouds, M. and Hus, M.S. (2012). Investigation on non linear-optical properties of palm oil/Silver nano- Particles. J.Europ. Opt Soc. Rap. Public. 7,12020-4.

Zhang, X.L.; Liu, Z.B.; Li, X.C.; Ma, Q.; Chen, X.D.; Tian, J.G.; Xu, Y.F. and Chen, Y.S. (2013). Transient thermal effect, nonlinear refraction and nonlinear absorption properties of graphene oxide sheets in dispersion, Opt. Ex p. 21,7511-7520.