Determination of nonlinear refractive index of paprika oil and pepper oil under cw visible laser illumination
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Abstract:
We report the observation of multiple diffraction rings patterns in paprika and pepper oils using continuous wave laser beams. The patterns occur at relatively low intensities. The number of rings together with outermost ring diameter of each pattern increases monotonically with increasing input power. The total changes in refractive index, $\Delta n$, and nonlinear refractive index, $n_2$, are determined using the number of rings observed in both samples and 473 nm and 532 nm laser beams. We obtained good values of $\Delta n = 0.01$ and $n_2 = 10^{-6}$ cm$^2$/w. These large values are attributed to thermal effects.

Keywords: Diffraction rings, visible laser beams, nonlinear refractive index, vegetable oils.

Introduction
Changes in refractive index induced by optical fields give rise to variety of nonlinear phenomena in photoresponsive materials [1,2]. When a light beam passes through an absorbing medium whose refractive index decreases as a function of temperature, the medium behaves as a negative lens [3]. In the spatial domain, the interplay between divergence of the propagating beam and the nonlinear response of the medium can lead to a diverse range of self-action behavior such as optical self-trapping, soliton formation and spontaneous pattern formation due to modulation instability [4,5]. A related phenomena is the formation of spatial rings which is understood to be induced by spatial self-phase modulation arising from the laser induced refractive index. The multiple diffraction rings have been demonstrated for the first time back in 1967 by Callen et al., [6]. A pump laser beam with Gaussian intensity extent can induced such diffraction rings due to the induced of phase shift with a bell-shaped transverse profile (Gaussian) [7].
The interference between light from two distinct points on the Gaussian curve that have the same transverse wave vector can lead to either a maximum constructive or minimum destructive interference point or circle.

The rotation around the Gaussian curve should lead to bright or dark rings when the phase difference is equal to \(( m \pi )\), \( m \) being even or odd respectively. Diffraction rings can be used to estimate the total change in refractive index, \( \Delta n \), \( n = n_0 + \Delta n \), where \( n \) is the refractive index in the presence of a laser beam and \( n_0 \) is the linear or background refractive index. \( \Delta n \) equals to \( n_2 I \), \( n_2 \) is the nonlinear refractive index and \( I \) is the laser beam intensity. During the last thirty years, tens of materials have been tested against diffraction rings for the sake of materials having the highest nonlinear refractive index, \( n_2 \), and fast response time [8-11].

Very little, or almost no vegetable oil has been tested for the same purpose [12]. In the present article, we are presenting comparative results of the nonlinear response of paprika (red) oil and pepper (black) oil using visible (437 nm and 532 nm) c.w laser beams.

**Experimental:**

**Samples and UV – visible spectroscopic results**

The paprika and pepper oils used in the experiments are available in the local market, their chemical structures are shown in figure (1).

![Chemical structures and molecular formulas for paprika and pepper oils](image)

Fig. (1): Chemical structures and molecular formulas for paprika and pepper oils.
A UV – visible spectroscopy have been used to characterizes paprika and pepper oils in the ranges (300 – 900 nm). The absorption (A) of both oils measured using double UV – visible spectrophotometer (type 6800 UV / vis Jenway. England).These measurements were performed at room temperature. Fig. (2): shows spectral distributions of absorbance in the spectral range (300 - 900) nm.

**Fig. (2): Spectral absorbance of (a) paprika and (b) pepper oils**

**(a) Experimental set – up**

The experimental arrangement, shown in figure (3), comprised an paprika oil and pepper oil in a glass cell, 1 mm thick. Two conventimal solid state laser (SDL – 473 – 050 T) and (SDL – 532 – 100 T) emitting blue (473 nm) and green (532 nm) light beam operating on the lowest order transverse (TEM00) modes given Gaussian spatial distribution, respectively. A glass lens of 50 mm focal length was used in the experiments and a digital camera (sony DSC -T99 - 8700 -82-25 mm) and a semitransparent 30 * 30 cm screen. The laser input power was measured using a power multi-range meter (type SDL -PM - 002).

**Diffraction rings**

**Patterns measurements**: The green laser output power can be varied.
between zero and 102 mW while the blue laser output power varied between zero and 66 mW. Both laser beam spot sizes radius is 1.5 mm (at \( \frac{1}{e^2} \)). The laser light intensity was calculated using the formula \( I = \frac{2P}{\pi \omega^2} \) where \( P \) is the laser output power and \( \omega \) is beam radius at each sample. Figs. (4) and (5) shows input power dependence of diffraction ring number for (A) paprika oil and (B) pepper oil respectively where it can be seen that number of rings for both samples increases monotonically with increases of input light power at \( \lambda = 532 \, \text{nm} \) and \( \lambda = 473 \, \text{nm} \) respectively.

Fig. (4). Variation of rings number in (A) paprika oil [I: 3.8 mW, II: 15.2 mW, III: 30.4 mW, IV: 68.4 mW] and (B) pepper oil [I: 7.4 mW, II: 29.6 mW, III: 66.6 mW, IV: 81.4 mW] using light beam of \( \lambda = 532 \, \text{nm} \) against input power.

Fig. (5): Variation of rings number in (A) paprika oil [I: 3.45 mW, II: 17.25 mW, III: 31.05 mW, IV: 37.95 mW] and (B) pepper oil [I: 11.25 mW, II: 26.25 mW, III: 41.25 mW, IV: 56.25 mW] using light beam of \( \lambda = 473 \, \text{nm} \) against input power.
Fig .( 6 ) and Fig .(7) shows two graphs of the increase of ring number with input power for both samples at $\lambda = 532$ nm and $\lambda = 473$ nm respectively.

Fig (6): Variation of rings number oil against input power at $\lambda = 532$ nm for paprika oil and pepper oil.

Fig (7): Variation of rings number against input power at $\lambda = 473$ nm for paprika oil and pepper oil.

Figs.( 8 and 9 ) shows the variation of diameter of external ring with input power for both samples at $\lambda = 532$ nm and $\lambda = 473$ nm respectively. It can be seen the increase of diameter for both samples monotonically.

Fig (8): Variation of external ring diameter against input power at $\lambda = 532$ nm for paprika oil and pepper oil.
With the positive 50 mm lens in its place, the beam radius, $\omega$, at the 1 mm cell when the laser is at the focal point of the lens can be calculated using the formula, $\omega = 1.22 f \lambda / \omega_0$ [13] where $\lambda$ is the laser beam wavelength, $f$ is the focal length of the lens and $\omega_0$ is the laser beam radius strikes the lens so that $\omega$ depend on the laser light wavelength i.e., $\omega = 19.24 \mu m$ for $\lambda = 473$ nm and $\omega = 21.63 \mu m$ at 532 nm.

To meet the nonlinear thin medium requirement case, the cell thickness must be less than the Rayleigh range (length), $ZR$ for positive lens, $ZR = \pi \omega_2 / \lambda$ [14] where $\omega$ and $\lambda$ have the same definitions mentioned earlier so that ($ZR = 2.76$ mm) $>1$ mm cell length at $\lambda = 532$ nm and ($ZR = 2.65$ mm) $>1$ mm at $\lambda = 473$ nm.

Estimation of nonlinear refractive index for both samples: We estimated the induced refractive index change, $\Delta n$, and the magnitude of the effective nonlinear refractive index, $n_2$, from the diffraction rings pattern data obtained earlier. The number of bright rings, $N$, can be approximated as [8]:

$$N = \frac{(\Delta \Phi)_{max}}{2\pi} \quad \ldots \quad (1)$$

Where, $(\Delta \Phi)_{max} = k \Delta$, is the phase change of the laser beam as a result of light traversing a distance $(d)$ in the nonlinear medium, $k$ is the wave vector $= \frac{2\pi}{\lambda}$ and $\Delta = d \Delta n$ is the optical path difference in the nonlinear and $\Delta n$ is the total change in the medium and refractive index. $\Delta n = In_2$, $n_2$
is the nonlinear refractive index, and I is the laser beam intensity. 

\[ (\Delta \Phi)_{\text{max}} = 2\pi N = \frac{2\pi}{\lambda} d\Delta n \]

so that

\[ \Delta n = \frac{\lambda N}{d} \] \hspace{1cm} (2)

\[ n_2 = \frac{\Delta n}{I} \] \hspace{1cm} (3)

According to equations (2) and (3) tables (1-4) summarizes the values of \(\Delta n\) and \(n_2\) as functions of input powers. The obtained results are explained as follows: For varying input power from zero and above there exist thresholds for obtaining the first ring from both samples which are different in magnitude depending on the amount of absorption from both samples and based on the wave-length used. As the input power increased so does the amount of power absorbed by the samples, hence local increase in temperature occurs in the shape of bell like shape as a result of the Gaussian distribution of the laser beam. The amount of temperature near the peak of the laser beam is higher than those around the wings of Gaussian distribution. Hence more reduction in refractive index occurs on the axis of the beam direction and more rings born around the central part of the beam i.e., the laser beam diverge or self-defocused around the central part of the beam. This is why the external ring is more intense than the inner ones. The change in diffraction rings radius, shown in fig(7), increased with increasing input power.

**Table (1)**: Variation of the nonlinear refractive index, \(n_2\), and change in refractive index, \(\Delta n\), for paprika oil with increasing input power at \(\lambda = 473\) nm

<table>
<thead>
<tr>
<th>(P_{\text{in\ mw}})</th>
<th>(n_2 \times 10^{-7}\ \text{cm}^2/\text{W})</th>
<th>(\Delta n \times 10^{-6})</th>
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Table (2): Variation of the nonlinear refractive index, $n_2$, and change in refractive index, $\Delta n$, for paprika oil with increasing input power at $\lambda=532$nm.

<table>
<thead>
<tr>
<th>$P_{in}$ mw</th>
<th>$n_2 \times 10^{-7}$</th>
<th>$\Delta n \times 10^{-6}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>15.2</td>
<td>2.571</td>
<td>532</td>
</tr>
<tr>
<td>22.8</td>
<td>3.429</td>
<td>1064</td>
</tr>
<tr>
<td>30.4</td>
<td>3.858</td>
<td>1596</td>
</tr>
<tr>
<td>38</td>
<td>4.114</td>
<td>2128</td>
</tr>
<tr>
<td>45.6</td>
<td>6.001</td>
<td>3724</td>
</tr>
<tr>
<td>53.3</td>
<td>7.348</td>
<td>5320</td>
</tr>
<tr>
<td>60.8</td>
<td>8.358</td>
<td>6916</td>
</tr>
<tr>
<td>68.4</td>
<td>9.143</td>
<td>8512</td>
</tr>
<tr>
<td>76</td>
<td>10.286</td>
<td>10640</td>
</tr>
<tr>
<td>83.6</td>
<td>11.222</td>
<td>12768</td>
</tr>
<tr>
<td>91.2</td>
<td>11.575</td>
<td>14364</td>
</tr>
<tr>
<td>98.8</td>
<td>11.869</td>
<td>15960</td>
</tr>
<tr>
<td>102</td>
<td>11.913</td>
<td>17024</td>
</tr>
</tbody>
</table>

Table (3): Variation of the nonlinear refractive index, $n_2$, and change in refractive index, $\Delta n$, for pepper oil with increasing input power at $\lambda = 473$ nm.
Table (4) : Variation of the nonlinear refractive index , $n_2$, and change in refractive index , $\Delta n$, for pepper oil with increasing input power at $\lambda=532\text{nm}$.

<table>
<thead>
<tr>
<th>$P_{in} \text{ mw}$</th>
<th>$n_2 \times 10^{-7} \text{ cm}^2/\text{W}$</th>
<th>$\Delta n \times 10^{-6}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>22.2</td>
<td>1.761</td>
<td>532</td>
</tr>
<tr>
<td>29.6</td>
<td>2.640</td>
<td>1064</td>
</tr>
<tr>
<td>37</td>
<td>3.169</td>
<td>1596</td>
</tr>
<tr>
<td>44.4</td>
<td>3.521</td>
<td>2128</td>
</tr>
<tr>
<td>51.8</td>
<td>3.773</td>
<td>2660</td>
</tr>
<tr>
<td>59.2</td>
<td>4.622</td>
<td>3724</td>
</tr>
<tr>
<td>66.6</td>
<td>5.282</td>
<td>4788</td>
</tr>
<tr>
<td>74</td>
<td>6.339</td>
<td>6384</td>
</tr>
<tr>
<td>81.4</td>
<td>6.242</td>
<td>6916</td>
</tr>
</tbody>
</table>

In general the obtained nonlinear refractive index in this work can be well compared with those obtained in many materials obtained by z scan techniques , thermal lens techniques and diffraction rings techniques . (1.94 $\times 10^{-7}$ ) cm$^2$/W was obtained by Mao etal [10] , $10^{-8}$ cm$^2$/W by Milan chian[15] , $7.7 \times 10^{-8}$ cm$^2$/W by Henari [16] , Krishnamurthy etal [17],$1.17 \times 10^{-10}$ cm$^2$/W by Henari and Cassidy [18], ($-4.4 \times 10^{-8}$ ) cm$^2$/W by Vinitha and Ramalingam [19] , ($1.63 \times 10^{-7}$ ) cm$^2$/W by Ali and palanisamy [20 ] , $10^{-7}$ cm$^2$/W by Majles etal.,[21], $10^{-7} – 10^{-6}$ cm$^2$/W by Majles etal ., [22]and $10^{-5}$ cm$^2$/W by Qi etal .,[23].The only available result in vegetable oil

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is the one published by Zamiri et al., [12], using palm oil doped with silver nanoparticles to enhance the nonlinearity in this material. Their value of $n_2$ was $3.07 \times 10^{-5}$ cm$^2$/W. et al.,

**Conclusion:**
We have demonstrated diffraction ring patterns in two vegetable oils viz., paprika and pepper using two low power continuous wave visible laser beams. It appears to us that the increase in number of rings reaches saturation at high input power. The observed ring patterns are attributed to thermal effects as a result of the high absorption coefficients in the two samples and interference occurring on the observation screen in the far field.

**Reference**


青色・赤色の食用油を用いたラジカルの存在状態の測定 \( \Delta n = 0.01 \) \( n_2 = 10^{-6} \text{cm}^2/\text{W} \) と評価した。これは、比較的高い熱影響を示す。