Diffraction patterns and nonlinear optical properties of Nanocomposite (Gold nanoparticles / Epoxy resin polymer)

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ABSTRACT:
We observed and analyzed diffraction rings produced by CW Nd: YAG visible laser beam in a nanocomposite of gold nanoparticles doped by epoxy resin polymer. Change of refractive index, Δn, and nonlinear refractive index, n\textsubscript{2} were calculated based on the number of rings observed. A thermal effect is due to this major nonlinearity. With rising input power and sample thickness the number of rings increases. Spot diameter of rings was calculated and noted it increase with input power increasing and decreasing thickness of sample.

1. Introduction:
Nonlinear optics (NLO) are an optics branch that is concerned with the changes in material's optical properties when interacting with light. Nonlinear optical materials have been extensively explored for their diverse applications in all-optical switches, optoelectronic devices, 3-D optical memory devices, optical modulation, telecommunications, protection of human eyes and optical sensors, etc., and future applications in the sciences of biology and medicine [1, 2]. Continuous wave lasers are widely used in many applications, ranging from mW to kW [3]. Numerous techniques for measuring nonlinear refraction in various materials are described. The diffraction ring patterns are among those techniques and the easiest [4, 5]. This approach is a technological modification of the Z-scan. As in the D-scan method, an increase in sensitivity permitting measurements of nonlinearity caused by materials. For this technique, the power meter detector used in Z-scan was replaced by the transparent screen. In this research, The experimental procedure should include a continuous wave Nd: YAG laser second harmonic generating SHG (532 nm), a focal lens (10 cm) that used focus the laser beam on the sample, pinhole, screen and finally camera to record the diffraction rings [6].

2. Experimental Methods
2.1. Preparation of Nanocomposite:
We mix (10 ml) of type A epoxy resin with (5) ml of type B epoxy harder to prepare the nanocomposite of gold nanoparticles doped with epoxy and add (5) ml of gold nanoparticles
and place the mixture on a magnetic stirrer for (5) min. The mixture was modified after homogenization into a glass slide set with a spin coater [7] and the quantity increased each time (4, 6, 8, 10, 12 and 14) drops for (90) seconds and at speeds of (2000) rpm. Then, the samples were left to dry for (2) days at room temperature. Then after, the thickness for each sample was then determined by the thickness measuring tool, calculated the average thickness of the glass samples without the mixture, and taken with the mixture the average thickness of the glass samples and measured the difference between them, to obtain the thickness of each sample, it was around (2.5, 9, 10.5, 14.3, 20.5 and 34) μm.

2.2. Experimental Setup of D-scan technique
The experimental setup consisted of a CW Nd: YAG laser that operates at (532) nm and a positive (10) cm focal length glass lens to concentrate the laser beam at the sample [8]. The power input was calculated using a detector at an input power of (50, 60, 70, 80, 90) mW. The observed diffraction ring pattern is therefore referred to a laser-induced nonlinear phase change due to the intensity dependent of the refractive index and the lens [9]. Using a digital camera, the nano composite self-diffraction rings were observed at a distance of (2) m from the samples [10].

In this technique the induced refractive index change (Δn) and the nonlinear refractive index (n2) can be measured. The experimental structure is expressed in Fig. 1 [11].

![Experimental set-up for diffraction rings technique.](image.png)

**Fig. 1.** Experimental set-up for diffraction rings technique.

### 2.2 Diffraction ring patterns measurements
The experimental setup for the diffraction ring patterns is the same as stated, expect the transparent screen to replace the power meter detector. For the previous data as below, we can calculate the induced refractive index change (Δn) and the effective nonlinear refractive index (n2). Due to the Gaussian distribution of the laser beam used for the experiment, the relative phase shift, DF, experienced by the beam when crossing the thickness (L) sample can be written as [12]:

\[ Δφ = kLΔn \quad \ldots..(1) \]
Where:
K: is the wave number and equal \(2\pi/\lambda\).
\(\lambda\): is the wavelength of the laser beam.
L: is the thickness of the sample.

The nonlinear phase-shift \((\Delta \phi)\) of one axis can be determined by the number of rings \((N)\) noted as [13]:

\[ \Delta \phi = 2\pi N \]  

The induced refractive index \((\Delta n)\) varies from the above equations, and

This method enables the measurement of an efficient nonlinear refractive index \((n_2)\) [14]:

\[ \Delta n = \frac{\lambda N}{L} \]  
\[ n_2 = \Delta n/I \]

Where:
N: is the number of rings.
I: is the laser beam input intensity.

2. Results and discussion
The sample was located at the focal point of the lens or right behind it. When the laser power increased gradually, diffraction rings were observed on the screen when the laser power reached a certain value-threshold. As Laser power increases, the number of rings increases. Which means they were based on intensity. It was also observed that the diameter of the spot ring increases with increases in input power in each pattern. Using equations (2), (3) and (4), the relative phase shift \((\Delta \phi)\), the refractive index transition \((\Delta n)\) and the nonlinear refractive index \((n_2)\) for all samples, respectively, were calculated. It has also been observed that the ring spot diameter increases with increases in input power in each sample, while the spot diameter decreases with increased sample thickness. In addition, we found that the increase in the relative phase shift \((\Delta \phi)\), the nonlinear refractive index \((n_2)\) and the change in the refractive index \((\Delta n)\) with an increase in input power and a decrease with an increase in sample thickness.

The next image is acquired from the Nano-composite diffraction rings at thickness \((2.5)\) \(\mu m\) shown in Fig. 2. when the different input power of the laser source used:
Figure 2: Diffraction ring images for Nano-composite at thickness 2.5 µm with various input power (a) 50 mW (b) 60 mW (c) 70 mW (d) 80 mW (e) 90 mW.

Table (1) shows the values of ($n_2$) and ($\Delta n$) for Nano-composite with thickness (2.5) µm as follows:

Table. 1: Change of refractive index ($\Delta n$) and nonlinear refractive index ($n_2$) for Nano-composite at thickness 2.5 µm for different input power of the laser source.

| Power (mW) | Intensity (w/cm$^2$) | No. of rings | Spot diameter (cm) | $\Delta \Phi$ | $n_2$ (cm$^2$/W) x10$^{-3}$ | $\Delta n$ x10$^{-3}$ |
|------------|----------------------|--------------|-------------------|------------|----------------|------------------|
| 50         | 1990                 | 2            | 1.587             | 12.6       | 2.1           | 426              |
| 60         | 2388                 | 3            | 1.666             | 18.8       | 2.6           | 638              |
| 70         | 2786                 | 3            | 1.693             | 18.8       | 2.3           | 638              |
| 80         | 3183                 | 4            | 1.825             | 25.1       | 2.6           | 851              |
| 90         | 3582                 | 6            | 1.852             | 37.7       | 3.5           | 1277             |
The next image is acquired from the Nano-composite diffraction rings at thickness (9) µm shown in Fig. 2 when the different input power of the laser source used:

Figure 3: Diffraction ring images for Nano-composite at thickness 9 µm with various input power (a) 50 mW (b) 60 mW (c) 70 mW (d) 80 mW (e) 90 mW

Table (2) shows the values of (n₂) and (∆n) for Nano-composite with thickness (9) µm as follows:

Table 2: Change of refractive index (∆n) and nonlinear refractive index (n₂) for Nano-composite at thickness 9 µm for different input power of the laser source.

| Power (mW) | Intensity (w/cm²) | No. of rings | Spot diameter (cm) | ∆φ | n₂ (cm²/W) x10⁻⁴ | ∆n x10⁻³ |
|------------|-------------------|--------------|--------------------|-----|------------------|----------|
| 50         | 1990              | 2            | 1.190              | 12.6| 0.6              | 118      |
| 60         | 2388              | 3            | 1.349              | 18.8| 0.7              | 177      |
| 70         | 2786              | 4            | 1.481              | 25.1| 0.8              | 236      |
| 80         | 3183              | 5            | 1.508              | 31.4| 0.9              | 296      |
| 90         | 3582              | 7            | 1.666              | 44  | 1.4              | 413      |

The next image is acquired from the Nano-composite diffraction rings at thickness (10.5) µm shown in Fig. 4 when the different input power of the laser source used:
Figure 4: Diffraction ring images for Nano-composite at thickness 10.5 µm with various input power (a) 50 mW (b) 60 mW (c) 70 mW (d) 80 mW (e) 90 mW

Table (3) shows the values of \( n_2 \) and \( \Delta n \) for Nano-composite with thickness (10.5) µm as follows:

| Power (mW) | Intensity (w/cm\(^2\)) | No. of rings | Spot diameter (cm) | \( \Delta \phi \) | \( n_2 \) (cm\(^2\)/W) \( \times 10^4 \) | \( \Delta n \times 10^{-3} \) |
|------------|------------------------|--------------|-------------------|-----------------|-----------------|-----------------|
| 50         | 1990                   | 2            | 1.164             | 12.6            | 0.5             | 97              |
| 60         | 2388                   | 3            | 1.243             | 18.8            | 0.6             | 152             |
| 70         | 2786                   | 4            | 1.402             | 25.1            | 0.7             | 203             |
| 80         | 3183                   | 5            | 1.481             | 31.4            | 0.8             | 253             |
| 90         | 3582                   | 7            | 1.640             | 44              | 1               | 354             |

The next image is acquired from the Nano-composite diffraction rings at thickness (14.3) µm shown in Fig. 5. when the different input power of the laser source used:
Figure 5: Diffraction ring images for Nano-composite at thickness 14.3 µm with various input power (a) 50 mW (b) 60 mW (c) 70 mW (d) 80 mW (e) 90 mW

Table (4) shows the values of ($n_2$) and ($\Delta n$) for Nano-composite with thickness (104.3) µm as follows:

Table 4: Change of refractive index ($\Delta n$) and nonlinear refractive index ($n_2$) for Nano-composite at thickness 14.3 µm for different input power of the laser source:

| Power (mW) | Intensity (w/cm²) | No. of rings | Spot diameter (cm) | $\Delta \phi$ | $n_2$ (cm²/W) x10⁻⁵ | $\Delta n$ x10⁻³ |
|------------|-------------------|--------------|--------------------|--------------|----------------------|-----------------|
| 50         | 1990              | 2            | 0.952              | 12.6         | 3.7                  | 74              |
| 60         | 2388              | 3            | 1.190              | 18.8         | 4.6                  | 112             |
| 70         | 2786              | 4            | 1.269              | 25.1         | 5.3                  | 149             |
| 80         | 3183              | 5            | 1.428              | 31.4         | 5.8                  | 186             |
| 90         | 3582              | 6            | 1.561              | 37.7         | 6.2                  | 223             |

The next image is acquired from the Nano-composite diffraction rings at thickness (20.5) µm shown in Fig. 6. when the different input power of the laser source used:
Fraction ring images for Nano-composite at thickness 20.5 µm with various input power (a) 50 mW (b) 60 mW (c) 70 mW (d) 80 mW (e) 90 mW

Table (5) shows the values of (n$_2$) and ($\Delta n$) for Nano-composite with thickness (20.5) µm as follows:

Table 5: Change of refractive index ($\Delta n$) and nonlinear refractive index (n$_2$) for Nano-composite at thickness 20.5 µm for different input power of the laser source.

| Power (mW) | Intensity (w/cm$^2$) | No. of rings | Spot diameter (cm) | $\Delta \Phi$ | n$_2$ (cm$^2$/W) x10$^{-4}$ | $\Delta n \times 10^{-3}$ |
|-----------|---------------------|--------------|--------------------|-------------|-----------------|----------------------|
| 50        | 1990                | 2            | 0.952              | 12.6        | 0.26            | 52                   |
| 60        | 2388                | 3            | 1.111              | 18.8        | 0.23            | 78                   |
| 70        | 2786                | 4            | 1.269              | 25.1        | 0.37            | 104                  |
| 80        | 3183                | 5            | 1.349              | 31.4        | 0.40            | 130                  |
| 90        | 3582                | 6            | 1.455              | 37.7        | 0.43            | 156                  |

The next image is acquired from the Nano-composite diffraction rings at thickness (34) µm shown in Fig. 7. when the different input power of the laser source used:
Figure 7: Diffraction ring images for Nano-composite at thickness 34 µm with various input power (a) 50 mW (b) 60 mW (c) 70 mW (d) 80 mW (e) 90 mW
Table (6) shows the values of \( n_2 \) and \( \Delta n \) for Nano-composite with thickness (34) µm as follows:

Table. 6: Change of refractive index \( (\Delta n) \) and nonlinear refractive index \( (n_2) \) for Nano-composite at thickness 34 µm for different input power of the laser source.

| Power (mW) | Intensity (w/cm\(^2\)) | No. of rings | Spot diameter (cm) | \( \Delta \phi \) | \( n_2 \) (cm\(^2\)/W) \( \times 10^{-5} \) | \( \Delta n \times 10^{-3} \) |
|-----------|------------------------|--------------|-------------------|----------------|--------------------------------|--------------------|
| 50        | 1990                   | 3            | 0.846             | 18.8          | 2.3                             | 47                 |
| 60        | 2388                   | 4            | 1.058             | 25.1          | 2.6                             | 63                 |
| 70        | 2786                   | 5            | 1.111             | 31.4          | 2.8                             | 78                 |
| 80        | 3183                   | 6            | 1.269             | 37.7          | 2.9                             | 94                 |
| 90        | 3582                   | 7            | 1.349             | 44            | 3.1                             | 110                |

The relation of input power to spot diameter is shown in Figure (8). It has been noted that the spot diameter increases with increasing number of rings while decreasing with increasing thickness.
Figure 8: The relationship between the input power and the spot diameter for nanocomposite with different thickness in D-scan technique

In the following Figure (9), the relationship between the input power and the number of rings for nanocomposite with different thickness in D-scan technique are performed.
3. Conclusion

In summary, we noted several diffraction rings of a CW Nd: YAG laser light passing through a nanocomposite of gold nanoparticles with epoxy resin polymer at different thicknesses. We calculated the nonlinear refractive index coefficient ($n_2$) and the change in the refractive index ($\Delta n$) for nanocomposite samples for different thicknesses using the D-scanning technique with 532 nm wavelength of laser. With through input power increasing, the number of rings observed increases. The calculation of the change in the refractive index, including the relative phase shift, $\Delta \phi$, and the effective nonlinear refractive index, $n_2$ were noted to be increase with increase input power. It was also found that the diameter of the ring spot increases with increases in input power in each sample, although decreases with higher sample thickness.

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