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Investigation of nonlinear optical properties of Sudan red G dye solutions using diffraction rings patterns and z-scan technique

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Abstract—Nonlinear optical properties of Sudan red G (SRG) dye solutions in DMF solvent at different concentrations using diffraction ring patterns and z-scan techniques were investigated. The samples were exposed to two different wavelengths of light sources, continuous wave laser operating at 532 nm green laser and blue laser operating at 473 nm. The diffraction ring patterns of the SRG solutions at three different dye concentrations and over laser power ranging from 5 to 50 mW have been recorded, showing asignificant nonlinear optical behaviors. The findings show that the number of diffraction rings increases as laser power increases, leading to increase the nonlinear refractive index (n₂). It was also observed that when the dye concentration increases from 0.25 mM to 1 mM and at the incident intensity of 6800 W/cm², the refractive index increases from 0.78×10^{-6} to $1.33 \text{ cm}^2/\text{W}$ for the sample exposed by green laser and from 0.48×10⁻⁶ to 1.18×10⁻⁶ cm²/W for the sample exposed by blue laser. From open and closed aperture z-scan curves, the nonlinear absorption coefficient (b) and nonlinear refractive index of the samples were evaluated at laser intensity of 680 W/cm². As dye concentration increases from 0.05 mM to 0.1 mM, β increases from ~6.9×10⁻³ cm/W to ~11.16×10⁻³ cm/W at wavelength 532 nm and from ~5.78×10⁻³ cm/W to ~10.08×10⁻³ cm/W at wavelength 473 nm. Moreover, the value of n₂ are increases from ~1.23×10⁻⁷ cm²/W to ~4.25×10⁻⁷ cm²/W at wavelength 532 nm and from ~0.8×10⁻⁷ cm²/W to ~3.33×10⁻⁷ cm²/W at wavelength 473 nm, when the dye concentration increases from 0.05 mM to 0.1 mM. The optical limiting (OL) of the dye solutions was tested as a function of dye concentration and the thresholds values of the dye solution at concentration of 0.075 mM are ~12 mW and ~8.5 mW at the wavelength 532 nm and 473 nm, respectively. The high values of β and n_2 , and the significant optical limiting behavior obtained in the present work suggest that the Sudan red G dye could be used for optical nonlinear devices and device protection applications.

Keywords—Z-scan technique, diffraction ring patterns, Sudan red G dye, nonlinear parameters, optical limiting.

I. INTRODUCTION

In recent decades, the study of linear and nonlinear optical properties of materials has represented a fertile and attractive field for researchers to carry out important research related to materials that exhibit high nonlinear properties, such as semiconductor materials, organic, inorganic and nanomaterials [1-4]. To investigate the suitability of materials for a potential application, it is important to study the nonlinear optical properties of that material such as the type of nonlinearity, its size, spectrum, response time, e.tc. [5]. Organic materials have important advantages over other materials, which makes them more suitable for studying nonlinear optical properties. Dyes have occupied an important space among the studied organic materials due to their highly nonlinear properties, such as storing optical data [6], optical limiting (OL) [7-9], optical stabilization [10], phase coupling [1], optical switches [11] and optical modulation [12].

Among the dyes that have been studied are azo dyes. Azo dyes constitute more than 50% of the total production of dyes in the world. They are characterized by ease of preparation, strong and safe, cover a wide range and have good stability properties. Azo dyes are aromatic compounds that contain both azo and diazo groups. The azo group (-N=N-) is attached to an atomic group belonging to any organic group (short) [13].

Sudan dyes are available in different types, such as Sudan I, Sudan II, Sudan III, Sudan IV, Sudan Red B, Sudan Red G, Sudan Red 7B, Sudan Orange, Sudan Black and Sudan Blue [1,14]. Recently, the nonlinear optical properties and optical limiting of some Sudan dyes were investigated using diffraction ring lase patterns and z-scan techniques due to that Sudan dyes have high values of the nonlinear refractive index and the nonlinear absorption coefficient [1,15,16].

The nonlinear optical response in azo dyes may result from an electronic and/or non-electronic response process. Changes in density and temperature are non-electronic responses resulting from non-radiative interactions. Electronic nonlinearity occurs due to the non-linear response of electrons associated with the applied optical field. Its importance lies in its presence in all insulating materials. On the other hand, organic materials that have a nonlinear response exhibit significant optical properties and electronic nonlinearity due to their delocalized electrons. The characteristic response time for electronic nonlinearity is estimated at 10–16 seconds [17,18].

Three common techniques for studying nonlinearity using low-power visible wavelength lasers are direction scanning, diffraction ring patterns, and thermal lensing to calculate absorption and refraction coefficients for potential optical applications [19].

In this work we successfully investigated the nonlinear optical properties of Sudan red G dye solution in DMF

solvent to study the nonlinear optical properties and optical limiting. A high nonlinear refractive index and significant optical limiter characteristics were obtained. We use two techniques to evaluate the nonlinear parameters, the diffraction ring patterns and z-scan. Three different concentration of dye solutions were prepared to study the nonlinear properties using two light sources, green laser at 532 nm and blue laser at 473 nm. The open and closed aperture z-scan curve of the samples was measured as a function of dye concentration at the two laser sources. The nonlinear refractive index (n₂) and the nonlinear absorption coefficient (β) at two different wavelengths, 532 nm and 473 nm, were determined as a function of concentrations. The optical limiting property and the thresholds values of the dye solutions were also investigated.

II. EXPERIMENTAL

A. Materials

Sudan red G (SRG) dye ($M_W = 278.31$ g/mol), chloroform and DMF solvents were purchased from the Merck company. The molecular formula of SRG ($C_{17}H_{14}N_2O_2$) was illustrated in Fig. 1.



Fig. 1: Molecular formula of SRG dye.

B. Samples preparation

Figure 2 shows the preparation of the SRG dye solutions, where 0.011g of SRG dye dissolved in 20 ml DMF solvent, Resulting in a SRG solution at concentration of 2 mM. After that, the concentration of the solution is diluted to obtain lower concentrations, as shown in the table (1).



Fig. 2: Schematic diagram of Sudan red G (SRG) dye solutions preparation.

Table 1. The concentrations of the dye solutions that are used to study	the
nonlinear optical properties	

Initial dye	Volume	DMF	New	Used	
concentrati	SKG	(ml)	(mM)	λ	λ
on (mM)	(ml)			= 532 <i>nm</i>	= 473 <i>nm</i>
2	2	2	1	DRP	DRP
1	2	2	0.5	DRP	DRP
0.5	2	2	0.25	DRP	DRP
1	1	9	0.1	Z-Scan, OP	Z-Scan
0.1	3	1	0.075	Z-Scan, OP	Z-Scan
0.1	2	2	0.05	Z-Scan, OP	Z-Scan, lOP
0.05	2	2	0.025		OP

C. Linear optical properties of SRG dye solutions

Figure 3(a) shows the absorption spectra of the SRG dye solutions at three different concentrations, 0.05 mM, 0.075 mM, and 0.1 mM. As seen from this figure, all spectra have two peaks, one is located in the ultraviolet region (~305 nm) and the second is located within the visible region (~490 nm) with a shoulder at about 520 nm. Electronic absorption spectra of this azo dye in solution often exhibit two types of bands. The structure of the indicators under investigation contains the π - π ^{*} transition of the aromatic system, which has been primarily responsible for the smaller wavelength band in the UV region at ~330 nm. The second band is characterized as the $n-\pi^*$ transition, which is located in the 495 nm domain and exhibits a prominent charge-transfer characteristic (CT transition). It is believed that this band is charge-transfer because its broad nature and sensitivity to the kind of group coupled to the azo linker [20].



Fig. 3: (a) The absorbance spectra, and (b) the absorbance coefficient of SRG dye solutions as a function of wavelength.

It was also noticed that the absorption values of the SRG solutions increase with increasing dye concentration as a result of the increase in the number of dye molecules in the solution. Figure 3(b) presents the relation of the absorption coefficient of the SRG dye solutions with the wavelength, showing an increase in the absorption coefficient as dye concentration increased. The absorption coefficient (α) is calculated from the formula [19].

$$\alpha = 2.303 A/t$$

Where A and t are absorbance and sample thickness (t=1 cm) of the SRG dye solutions, respectively.

III. RESULTS AND DISCUSSIONS

A. Diffraction Ring Patterns (DRPs)

The diffraction ring patterns of SRG dye solutions were investigated using a CW laser beam from a solid-state laser at a wavelength of 532 nm with variable output power from 0 to 60 mW. As illustrated in Fig. 4, the beam is focused by a lens with a focal length of 50 mm. The sample (SRG dye solution) placed in a transparent cell and located at the focal length of the lens, where the Rayleigh length is $Z_R = 2.76$ mm. During the incident of the beam laser into the sample, DRPs are generated as a result of interaction between the solution and the beam laser. The DRPs are received on a transparent screen (the dimensions are 30x30 cm), where the distance between the sample and the screen is around 80 cm. To record the DRPs results, a digital camera was used to take the photos of the diffraction rings as a function of laser output power. The DRPs of dye solutions are also examined using a CW laser beam solid-state laser at a wavelength of 473 nm with a variable output power ranging between 0 and 70 mW. The Rayleigh length is determined at focus which is $Z_R = 2.456$ mm.



Fig.4: Schematic diagram of diffraction rings pattern setup.

Three different dye concentrations, 0.25 mM, 0.5 mM, and 1 mM were used to study DRPs as a function of incident laser power. Power-dependent DRPs of SRG dye solutions at three different concentrations illuminated by 532 nm and 473 nm laser sources were demonstrated in Fig. 5(a-c) and Fig. 6(a-c), respectively. The relation between the number of rings and the power of the incident laser beam for all samples was evaluated by using two different laser beam wavelengths, 532 nm and 473 nm, as plotted in Figs. 7(a) and 7(b), respectively. The results show the following information:

1- It is clearly evident that the number of diffraction rings increases as the incident laser power increases and it also increases when the SRG dye concentration increases (see Fig.7). This is attributed to generate a concave thermal lens in the medium which is turn shifts the phase of the laser waves by about 2π , showing bright and dark rings as a result of occurring the constructive and destructive interference processes [1]. This nonlinear phenomenon is highly affected by the temperature which is increased as laser power increases. It can be also noticed that the intensity of the inner rings is less than that of the outer rings. The reason is due to the phenomenon of self-focusing not occurring in the medium [21]. 2- The horizontal diameter (x) and vertical diameter (y) of the diffraction rings increases with increasing the incident power of the laser (see Figs. 8 and 9). It can also be seen that the horizontal diameter is larger than the vertical diameter, meaning that the diffraction rings are not circularly symmetrical.

3- The reason of why the horizontal diameter becomes greater than the vertical diameter when concentration of the SRG dye solution increases, is attributed to the increase in the sample's absorption of light dye as a result of increasing the number of molecules, and therefore part of the absorbed energy is transformed into thermal energy and the emergence of convection currents. In the horizontal and vertical directions, which leads to a process of rise and fall between the hot currents and the cooler ones, which leads to a decrease in the phenomenon of nonlinearity in the vertical direction compared with that in the horizontal direction [21]. 5 - The pattern area increases as a result of the increase in the number of diffraction rings.

The change in the phase of the incident laser beam ($\Delta \phi$) is calculated by using the following equation [22].

$\Delta \phi = 2\pi N$

where N is the number of diffraction rings. The change in the nonlinear refractive index of the sample can be determined from the formula [22]:

$$\Delta n = \frac{N\lambda}{d}$$

where λ is the wavelength of incident laser beam, d the thickness of sample (1mm). The laser intensity (I) is calculated from the relation [22]:

$$I = \frac{2 P}{\pi \omega^2}$$

where, P is the incident laser power, and ω is the radius of the laser beam at focus ($\omega \approx 21.635 \ \mu m$ for λ_{532} and $\omega \approx 19.235 \ \mu m$ for λ_{473}).

The nonlinear refractive index (n_2) is related to change in the refractive index (Δn) by the following formula [22]:

$$n_2 = \frac{\Delta n}{I}$$

The values of Δn and n_2 were calculated and listed in Table 2. As can be seen from this table, when the concentration of the dye increase from 1.25 mM to 5 mM, Δn values increase from $\sim 5.3 \times 10^{-3}$ to $\sim 9 \times 10^{-3}$ at the wavelength of 532 nm, and from $\sim 3.3 \times 10^{-3}$ to $\sim 8 \times 10^{-3}$ at the wavelength of 473 nm. It was also found that the nonlinear refractive index (n_2) of the SRG dye solutions increase with increasing the dye concentration, where under illumination by green laser, the n_2 values of the samples increase form $\sim 0.78 \times 10^{-6} \text{ cm}^2/\text{W}$ at 1.25 mM to $\sim 1.33 \times 10^{-6} \text{ cm}^2/\text{W}$ at 5 mM and when the dye solutions exposed by blue laser, the values of n_2 increase from $\sim 0.48 \times 10^{-6} \text{ cm}^2/\text{W}$ at 1.18 mM to $\sim 1.33 \times 10^{-6} \text{ cm}^2/\text{W}$ at 5 mM. In general, The values of Δn and n_2 of the samples illuminated by green laser is higher than those corresponding values of the samples exposed by blue laser.





Fig. 5: The diffraction rings patterns (DRPs) of the SRG dye solutions as a function of power laser. The excitation source is the green laser (532 nm).







Fig. 6: The diffraction rings patterns (DRPs) of the SRG dye solutions as a function of power laser. The excitation source is the blue laser (473 nm).





Fig.7: The number of the diffraction rings patterns as a function of incident laser power using (a) green laser, and (b) blue laser.



Fig. 8: The x and y dimensions of the diffraction rings patterns of the SRG dye solutions at three concentrations, (a) 1 mM, (b) 0.5 mM, and (c) 0.25 mM as a function of incident laser power using green laser.





Fig. 9: The x and y dimensions of the diffraction rings patterns of the SRG dye solutions at three concentrations, (a) 1 mM, (b) 0.5 mM, and (c) 0.25 mM as a function of incident laser power using blue laser.

Table 2. The values of the nonlinear refractive index for three dye concentrations and under illumination of two different laser wavelengths, 532 nm and 473 nm (I=6800W/cm²).

	λ=532 nm				λ=473 r	ım
Sample (SRG)	No.	$\Delta n imes 10^{-3}$	n_2 $\times 10^{-6}$ cm^2/W	No.	Δn ×10 ⁻³	n_2 $ imes 10^{-6}$ cm^2/W
0.25mM	10	5.32	0.78	7	3.31	0.48
0.5mM	12	6.38	0.94	14	5.67	0.83
1mM	17	9.04	1.33	17	8.04	1.18

B. Z-Scan technique

The Z-scan technique is considered the simplest and most common technique used to find the third-order nonlinear characteristics of samples that are exposed by a high light intensity source. Z-Scan technology is used to calculate the nonlinear absorption coefficient (β) and the nonlinear refractive index (n_2) of the medium. The setup of this technique was illustrated in Fig. 10. In this setup, a convergence lens of 5 cm focal length is used to focus the laser beam. Two different light sources were used, green laser (λ =532 nm) and blue laser (λ =473 nm). The laser intensity is fixed at 680 W/cm² which corresponds to the green laser power at 5 mW and to the blue laser power at ~4

mW. Then the sample (1mm-thick quartz-cell containing dye solution) is moved through the z-axis between the (-z) and (+z) direction along the laser beam propagation on both sides of the focal lens. The transmitting power is recorded as a function of the sample position after each movement along the Gaussian focus axis.



Fig.10: Schematic diagram of z-scan setup.

The z-scan experiment is performed in two arrangements: the first there is no cover in front of the photodetector aperture, which is called the open aperture and the second there is a cover which has a circular aperture with a diameter of (1mm) in front of the photodetector which is called the closed aperture.

After recording the change in transmittance power through a small aperture at the far-field position (closed aperture), the phase shift amplitude can be calculated. However, if the sample is moved through the focus and without placing a cover in front of the detector (open aperture), the density can be measured depending on the absorbance of the sample. When both methods, open and closed apertures, are used for measuring, the ratio of the signals determines the linear refractive index of the sample. The open aperture z-scan curves of SRG dye solutions at different dye concentrations using green laser and blue laser were shown in Fig 11 (a) and (b), respectively. It can be seen that the transmittance curve decreases as the dye concentration increases. The closed aperture z-scan curves are also measured as function of dye concentration using the green and blue laser as illustrated in Fig. 12 (a) and (b). The results show that the transmittance curve is decreasing with increasing the concentrations.





Fig.11: (a) Open aperture z-scan curves and (b) close aperture z-scan curves of SRG dye solutions at three different concentrations, 0.1 mM, 0.075 mM, and 0.05 mM using green laser with incident intensity (I=680W/cm²).



Fig. 12: (a) Open aperture z-scan curves and (b) close aperture z-scan curves of SRG dye solutions at three different concentrations, 0.1 mM, 0.075 mM, and 0.05 mM using blue laser with incident intensity ($I=680W/cm^2$).

In order to evaluated the nonlinear absorption coefficient and the nonlinear refractive index, the following mathematical formulas are used [23]:

$$|\Delta \varphi| = \frac{\Delta T_{p-v}}{0.406(1-S)^{0.25}}$$
$$L_{eff} = \frac{1 - \exp(-\alpha d)}{\alpha}$$
$$\beta = \frac{2\sqrt{2}\Delta T}{I. L_{eff}}$$
$$n_2 = \frac{\Delta \varphi \lambda}{2\pi L_{eff}}$$

Where $\Delta \phi$ is the on-axis phase shift, L_{eff} is the sample effective thickness, S is the linear transmittance of aperture (S_{532nm} = 0.393, S_{473nm} = 0.486), I is the intensity of

the incident laser (I=680 W/cm²), ΔT_{p-v} (T_{p} - T_{v}) is the difference between the normalized peak and valley transmittance, d is the sample thickness (1mm), ΔT is the normalized transmittance of the sample at z, λ is the wavelength of the laser source.

As seen from Fig. 13, the results show that the nonlinear optical parameters, n_2 and β increase as the dye concentration increases due to increase the dye molecules in the solution, leading to increase the linear absorption coefficient. It was also found that the values of these parameters of the samples exposed by green laser is higher than those values of the samples exposed by blue laser. This is attributed to that the linear absorption coefficient of the SRG dye at wavelength of 532 nm (green light) is higher than that at wavelength of 473 nm (blue light).

The results of nonlinear optical parameters obtained in the present work show high n_2 and β values of order 10^{-7} and 10^{-3} , respectively, which are in a good agreement with those values achieved in the previous works [1,16,19].



Fig. 13: (a) nonlinear absorption coefficient (β), and (b) nonlinear refractive index (n_2) as a function of dye concentration under two different laser sources, green and blue.

Table 3. The values of the nonlinear refractive index and nonlinear absorption coefficient of the SRG dye solutions at three different dye concentrations and under two different laser sources, green and blue

	λ=532 nm			λ=473 nm		
Sample (SRG)	α cm ⁻¹	$n_2 \ imes 10^{-7} \ cm^2/W$	$\beta \times 10^{-3} cm/W$	α cm ⁻¹	$\begin{array}{c} n_2 \\ \times 10^{-7} \\ cm^2/W \end{array}$	$egin{array}{c} \beta \ imes 10^{-3} \ cm/W \end{array}$
0.05	2.74	1.28	6.94	2.67	0.8	5.78
0.075	4.585	2.59	8.92	4.47	1.97	7.94
0.1	5.227	4.25	11.16	5.10	3.33	10.09

C. Optical Limiting

In this section, we studied the effect of reducing the optical energy of the SRG dye solution at two wavelengths, 532 nm and 473 nm for different dye concentrations of the dye. The setup of the optical limiting experiment was illustrated in Fig. 14. In this experiment, the closed aperture scanning technique is used but the sample is installed at a location after the focal length of the lens with a distance of 1cm and the aperture diameter was 2 mm. To evaluate the optical limit of samples, the output laser power (transmittance) was recorded as a function of the input laser power.



Fig. 14: Schematic diagram of optical limiting setup.

Figure 15 shows the optical limiting properties of the SRG dye solutions at two different wavelengths, 532 nm and 473 nm, for different dye concentrations. To evaluate the optical limit threshold of the dye solutions, the transmittance power as a function of input laser power of the samples under two different laser sources, green laser and blue laser for different dye concentrations was investigated and plotted in Fig. 16. From these two figures, it was found the following results:

1- For all dye concentrations and at low laser input power, the relation between the output power and the input power is a linear behavior. At high input power, the output power of the SRG dye solution is observed to be saturated. This behavior is highly effect by the dye concentration and the wavelength of the beam laser, as demonstrated in Fig. 15.

2- The ability of the samples to exhibit the optical limiting behavior depends on the dye concentration, where it is increased with increasing the concentration. This is attributed to that the linear absorption coefficient and the number of dye molecules increases as the dye concentration increases.

3- The optical limiting threshold values of the SRG dye solutions under incident laser power at wavelength of 473 nm are less than those values of the solutions under incident laser power at wavelength of 532 nm (see Table 4).

4- The SRG dye solutions exhibited low thresholds values, especially at high concentrations (see Table 4), meaning that these samples have high optical limiter efficiency which is very useful for devices protection applications.

5- The saturated laser output power and the optical limiting threshold are inversely proportional to the dye concentration (see Figs. 15 and 16), where the thresholds values decreases as dye concentration increases. The optical limiting behavior is due to thermally induced nonlinear refraction.

Table 4. The optical limiting threshold of the SRG dye solutions at two different wavelength, 532 nm and 473 nm for different dye concentrations.

λ=533	nm	λ=473nm	
Dye	Power	Dye	Power
concentration	Threshold	concentration	Threshold
	(mW)		(mW)
0.05 mM	15.4	0.025 mM	11.4
0.075 mM	12	0.05 mM	8.5
0.1 mM	8		



Fig. 15: Optical limiting characteristics of the SRG dye solutions at different dye concentrations under green and blue laser beam.





Fig. 16: Normalized transmittance of the SRG dye solutions as a function of input laser power at different dye concentrations under green and blue laser beam.

IV. CONCLUSION

Three different concentrations of Sudan red G (SRG) dye solution in DMF solvent were prepared to study the nonlinear optical properties of these solutions under a continuous laser beam with two wavelengths 532 nm and 473 nm. At dye concentrations of 0.25, 0.5, and 1 mM, the solutions exhibited clear diffraction ringpatterns (DRPs) at both laser beams, the green and blue lasers. The findings revealed that the number of diffraction rings increases with increasing the dye concentration and the power of the incident laser. The difference between the horizontal and vertical diameter increases with increasing the dye concentration due to generate a concave thermal lens in the medium. The nonlinear refractive index (n_2) was calculated using the diffraction ringpatterns (DRPs). At the incident intensity of 6800 W/cm² and as the dye concentration increases from 0.25 mM to 1 mM, the refractive index increases from 0.78×10^{-6} to $1.33 \text{ cm}^2/\text{W}$ for the sample exposed by green laser, and from 0.48×10^{-6} to 1.18×10^{-6} cm²/W for the sample exposed by blue laser. From open aperture and closed aperture Z-scan technique, it was noticed as an increase in the values of the nonlinear absorption coefficient and the nonlinear refractive index as the concentration of the dye increases. This is attributed to increase the number of dye molecules in the samples, leading to increase the light absorption by the solutions. The results also show that the n_2 values calculated by z-scan are lower than those values calculated by DRPs. This is due to that the dye concentrations and the incident laser intensity which are used in z-scan technique are less than those which are used in DRPs. Moreover, the optical limiting (OL) and the optical limiting threshold (OL_{Sh}) of the solutions was investigated as a function of dye concentration under two laser sources, the blue and green lasers. The results exhibit that the optical limiting and the threshold values are inversely proportional to the dye concentration, where the threshold values decreases as dye concentration increases. The samples exhibited low threshold values, especially at high concentrations, where at the dye concentration of 0.075 mM, the threshold values of the SRG dye solution are ~12 mW and ~8.5 mW at the wavelengths of 532 nm and 473 nm, respectively.

CONFLICT OF INTEREST

Authors declare that they have no conflict of interest.

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