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Solvents effect on the optical nonlinear properties of the sudan iv

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ABSTRACT

The optical nonlinear properties of three sudan iv solutions in DMSO, DMF and chloroform solvents are studied under irradiation with low power laser beam work at 473 nm. The diffraction ring patterns and Z-scan experiments were carried out separately to calculate the change in the index of refraction and the nonlinear index of refraction of each solution. The rings number per each pattern, the area of each pattern and the outer most ring diameter in each pattern increased with the increase of input laser beam intensity. Each pattern loses symmetry in the upper part as input intensity increased. The obtained diffraction ring patterns are simulated using Fraunhofer approximation of the Fresnel-Kirchhoff diffraction theory where good theoretical agreements with the experimental findings are obtained. According to both methods it is proved that sudan iv three solutions shows self-defocusing phenomena. Type of solvent appears to control the nonlinearities of sudan iv solutions. Optical limiting property is studied in the three solutions.

1. Introduction

The nonlinear optical properties of different materials were and still examined in the continuous wave (cw) regimes of visible low power laser beams by the three techniques viz., diffraction ring patterns [1,2], thermal lens [3,4] and by Z-scan [5,6]. These three techniques are simple and required little components viz., a laser device, a positive lens, a glass cell, a power meter, a semitransparent screen and a camera. So many materials have been tested for the sake of obtaining their nonlinear refractive indexes [7–21] for the possible use in different applications and devices [22–30]. Sudan dyes are available in various sorts viz., sudan i, sudan ii, sudan iii, sudan iv, sudan red B, sudan red G, sudan red 7B, sudan orange, and sudan black [31]. Generally all sudan dyes can causes potential risk to the health [32]. It received little attention from the point of view of nonlinear behavior. Sudan red G and sudan orange G optical linear and nonlinear properties and quantum parameters were studied by Esme and Sagdine [33], sudan i nonlinear optical response was studied by He and Wang [34]. Properties of optical limiting of sudan red B doped PMMA film, and films of sudan iv doped poly (alkyloxymethacrylate) for optical data storage were investigated by Qusay et al., [35,36]. Sudan iii have received much attention where ring patterns were generated under the effect of electrical field [37], measure of its nonlinear index of refraction [38], its photo-induced dichroism [39], its optical limiting behavior [40], its optical data storage [41] and its nonlinear optical properties were studied too [42]. Sudan iv optical nonlinear response was studied by Majles Ara et al. [43].

Through previous studies, which are mentioned above, it was concluded that sudan dyes have high nonlinear optical properties, so that one of sudan's dyes family, namely sudan iv is chosen, as a sample in this study. In this work the effect of three solvents viz., dimethyl sulfoxide (DMSO), dimethyl formamide (DMF) and chloroform on the optical nonlinear properties of sudan iv are presented

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via diffraction ring patterns and Z-scan techniques under low power cw laser beam of 473 nm wavelength. Also, the properties of optical limiting of the samples are reported.

2. Experimental

2.1. Material

Sudan iv was obtained from Thomas Baker. It is a dye belonging to the azo family, its color index number is 26105. Sudan iv formula is written as $C_{24}H_{20}N_4O$ and its molecular structure is shown in Fig. 1. Its average mass is 380.442 Da and its mono-isotopic mass is 380.163696 Da. Its melting point is at 199 °C, its linear refractive index is 1.645, its boiling point is 618.8 ± 55.0 °C at 760 mm Hg, and its density is 1.2 ± 0.1 g/cm³.

2.2. Spectroscopic study

The absorbance (A) spectra of the sudan iv solutions in the solvents DMSO, DMF and chloroform respectively at room temperature were obtained using a 6800 Jenway England spectrophotometer in the UV – visible range are shown in Fig. 2. It can be seen from Fig. 2 that the spectra of the sudan iv solutions in the solvents DMSO, DMF and chloroform are attributed to $\pi\text{-}\pi^*$ transition due to the conjugated double bonds of the benzene rings. With the use of Fig. 2 and the following relation, the absorption coefficients, α , of the sudan iv in the three solvents are calculated [44]

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

Where L is the thickness of sample. For L = 1 mm, the absorption coefficients of sudan iv solutions are: $\alpha_{\text{DMSO}} = 27.47$ cm⁻¹, $\alpha_{\text{DMF}} = 37.07$ cm⁻¹, $\alpha_{\text{Chloro}} = 48.1$ cm⁻¹ respectively. All absorption coefficients were calculated for the same solution concentration of 1 mM using a quartz cell of thickness 1 mm at the wavelength 473 nm.

2.3. Experimental set-up

The set-up of experiment shown in Fig. 3 was used in the obtaining of diffraction ring patterns. It comprised a solid state laser device emitting cw, single transverse, TEM₀₀, mode laser beam with low power at 473 nm. A power meter, a glass lens of 50 mm focal length was used to focus the laser beam onto the glass sample cell of 1 mm thickness and a 30 x 30 cm semitransparent screen was used to cast the diffraction ring patterns placed at 80 cm away from the samples cell exit. The Z-scan technique i.e. closed and open aperture Z-scans were carried out using the same set-up used to obtain the diffraction ring patterns with the fixing of the sample cell on a moving stage to scan the samples cells between (-z) and (+z) across the focus of the positive lens (z = 0). An aperture with a circular shape of 2 mm diameter was used to cover the detector which replaced the screen to measure the transmitted laser beam power from the samples cells. This is the closed aperture (CA) Z-scan, while the open aperture (OA) Z-scan measurements was carried out using the same set-up of CA Z-scan except replacing the circular aperture with another positive lens, to collect the transmitted laser beam from the samples cells. To carry out the experiment of optical limiting the same set-up of Z-scan measurements was used except that each sample cell was fixed behind the lens, in the valley position of the Z-scan CA experiment. Measurements were carried out via. varying the input power and measuring the corresponding output power.

3. Results

3.1. Diffraction ring patterns

The obtained ring patterns in the sudan iv solutions in DMSO, DMF, and chloroform solvents are shown in Fig. 4. Four distinct features are noticed in the obtained patterns (i) sever distortions occurs in the upper half of each ring pattern due to convection currents in the vertical direction which increases as input power increased, so that the upper part of each pattern grew in lower ratio compare to the lower ones. (ii) each outer most ring intensity of each pattern is large compare to the inner ones, a manifestation of self-defocusing as input power increased. (iii) as the input power increased the number of rings in each pattern increased too. (iv) as the

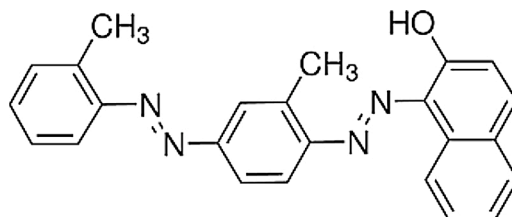


Fig. 1. The sudan iv molecular structure.

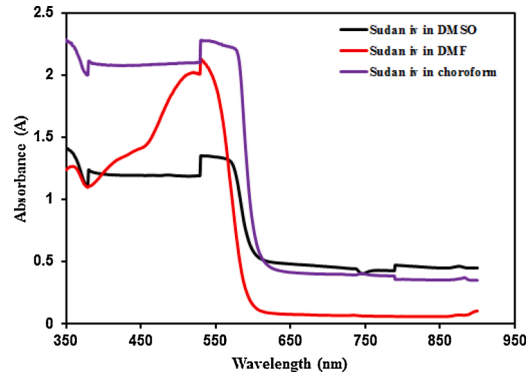


Fig. 2. Spectra of the absorbance (A) for sudan iv solutions in DMSO, DMF, and chloroform solvents.

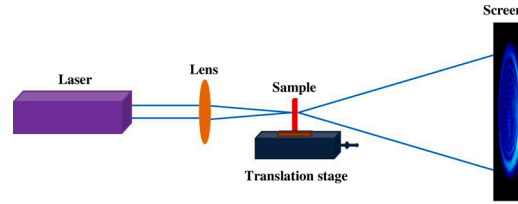


Fig. 3. Set-up of experiment for the formation of diffraction ring patterns.

input power increased the area of each pattern increased too.

As the laser beam leave the laser device output coupler the beam spot size, ω_0 , is 1.5 mm. With the use of a positive lens of focal length, $f = 5$ cm, beam wavelength, λ , of 473 nm, the distance between the laser device output coupler and the lens was 40 cm, the laser beam spot radius, ω' , fall on the entrance of sample cell is calculated via. the following equation [45]

$$\omega' = 1.22 \frac{f\lambda}{\omega} \tag{2}$$

ω is the radius of laser beam that fall on the lens, so that $\omega' = 19.235 \mu\text{m}$.

Taking the maximum number of rings, N , obtained at the same input power (53 mW) for sudan iv solutions as $N_{\text{DMSO}} = 17$, $N_{\text{DMF}} = 20$, and $N_{\text{chloro}} = 29$. It is believed that the birth of one ring lead's to a change of the laser beam phase by 2π radian as it passes through the nonlinear medium, so that the total phase shift, φ , acquired by the laser beam can be written as:

$$\varphi = 2\pi N \tag{3}$$

For sample thickness, L , a change in the medium index of refraction, Δn , the total change of phase, φ , is given by

$$\varphi = k\Delta L \tag{4}$$

where

$$k = \frac{2\pi}{\lambda}$$

and

$$\Delta L = L\Delta n \tag{5}$$

so that

$$\varphi = \frac{2\pi}{\lambda} L\Delta n \tag{6}$$

i.e.,

$$\Delta n = \frac{N\lambda}{L} \tag{7}$$

and the nonlinear index of refraction, n_2 , can be obtained using the relation [46]

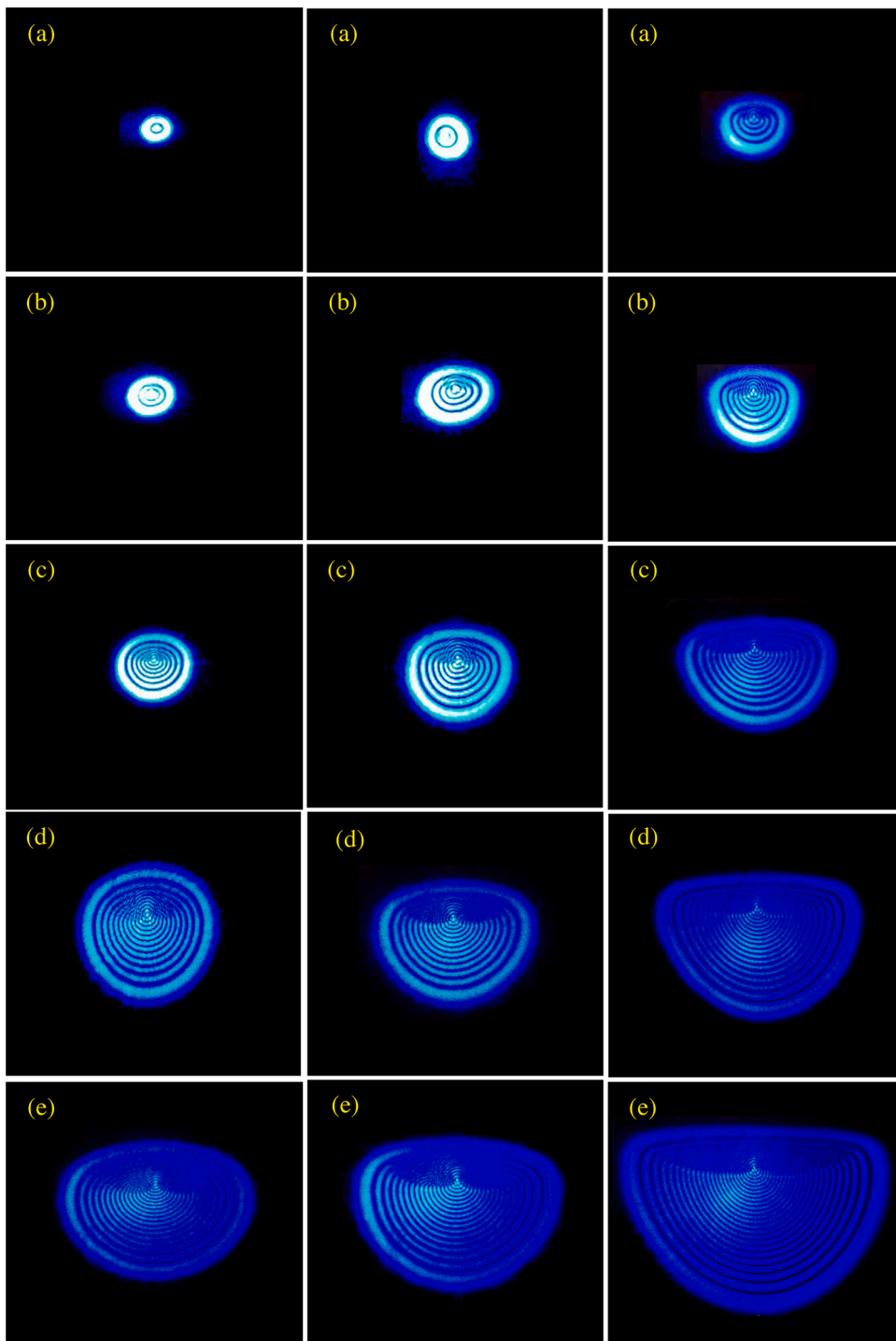


Fig. 4. Experimentally obtained diffraction ring patterns for the laser beam power of (mW) (a) 9, (b) 12, (c) 17, (d) 26, and (e) 47 passes through the sudan iv solutions in DMSO (left column), in DMF (middle column) and in chloroform (right column) respectively.

$$n_2 = \frac{\Delta n}{I} \tag{8}$$

I is the intensity of laser beam where

$$I = \frac{2P}{\pi\omega^2} \tag{9}$$

P is the input power of laser beam, so that the incident intensity on the sudan iv solutions is $I = 9124 \text{ W/cm}^2$, and Δn and n_2 of the sudan iv three solutions are as given in Table 2.

3.2. Simulating the diffraction ring patterns

To theoretically investigate the experimentally obtained diffraction ring patterns, when a cw laser beam having Gaussian intensity distribution traverses the sudan iv solutions, let the laser beam propagating along the z-direction and its intensity vary in the x-y plane. Let the laser beam power, P, its spot radius, ω , its wave front radius of curvature, R, and its propagation constant, k, so that the laser beam complex amplitude at the nonlinear medium entrance can be described by following formula [47]:

$$U(x, y, t, z = 0) = \left(\frac{2P}{\pi\omega^2}\right)^{1/2} \exp\left(-\frac{x^2 + y^2}{\omega^2}\right) \left(-ik \frac{x^2 + y^2}{2R}\right) \tag{10}$$

When the nonlinear medium absorbs part of the laser beam energy local heat of the nonlinear medium occur with a bell shape distribution as a result of the distribution of Gaussian laser beam. The heating of the nonlinear medium causes a reduction of the index of refraction of the nonlinear medium, so that a negative thermal lens is generated that causes a dispersion of the laser beam in the x-y plane perpendicular to the z-direction. Due to the change in the nonlinear medium index of refraction, the laser beam suffers changes in its phase. These effects leads to the formation of diffraction ring patterns. Since the highest heat change occur at the peak of the Gaussian beam and lowest at the beam wings, two types of heat transfers mechanisms occurs viz., conduction horizontally and convection vertically. When both are equal the diffraction ring patterns usually appears circular around the z-axis and asymmetric when one of them is larger than the other. In the experimental part (Fig. 4) asymmetries in the vertical direction as power increased are noted an indication that the vertical convection current overcome the horizontal conduction current. Such effect must be taken into account. By defining a convection velocity at the laser beam center, v_x , as follows [47]:

$$v_x = \frac{\beta g [\Delta T]_{max} \pi h^2}{16\mu} \tag{11}$$

β and μ are the sample thermal expansion and viscosity, g is the gravity acceleration, $[\Delta T]_{max}$ is the maximum change in the sample temperature and h is the distance from the center of the beam for which convection still occurs. Due to the heat resulted in the medium, the index of refraction, $n(x, y, t)$, changes which leads to changes in the beam phase, $\Delta\phi(x, y, t)$. Using Fraunhofer approximation of the Fresnel-Kirchhoff diffraction integral, the intensity distribution, $I(x', y', t)$, of the laser beam post traversing the sample of thickness L on the screen in the far field a distance d from the samples cells exits, defining new spatial variables, x' , and, y' , can be written as [47]:

$$I(x', y', t) = \left|\left(\frac{2P}{\pi\omega^2}\right)^{1/2} \frac{i\pi\omega^2}{\lambda d} \exp(ikd) \exp\left(-\frac{\alpha L}{2}\right) \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy \exp\left(-\frac{x^2 + y^2}{\omega^2}\right) \exp\left[i\left(-k \frac{x^2 + y^2}{2R} + \Delta\phi(x, y, t)\right)\right] \exp\left(-ik\left(\frac{xx' + yy'}{d}\right)\right)\right|^2 \tag{12}$$

Eation (12) is solved via. the Mat Lab system with the parameter values given in Table 1, the simulation results of the diffraction ring patterns in the sudan iv three solutions against input powers (mW), (9,12,17,26,47) are shown in Fig. 5 respectively. Good agreements can be noticed when compare the simulation results with experimental ones shown in Fig. 4. Figs. 6–8 are simulation results of one dimensional intensity laser beam distributions on the screen for sudan iv in DMSO, DMF and chloroform solvents respectively. The x-axis and y-axis distributions are shown in the left and right columns respectively. Fig. 9 are laser beam phase spatial

Table 1
Parameters of DMSO, DMF and Chloroform solvents.

Parameters	DMSO	DMF	Chloroform
Polarity	7.2	6.3	4.4
Dipole moment	3.96	3.86	1.040
Density (gmcm ⁻³)	1.092	0.944	1.492
Thermal conductivity (Wm ⁻¹ k ⁻¹)	0.199	0.175	0.13
Specific heat (J.gm ⁻¹ k ⁻¹)	1.956	2.050	0.9566
Thermal diffusivity (m ² /sec)	3.56×10^{-6}	9.57×10^{-8}	0.81×10^{-7}
Refractive index	1.479	1.4305	1.4459
Viscosity (m ² /sec)	1.996	0.92	0.57
Heat capacity (Jk ⁻¹ mol ⁻¹)	153	146.05	91.47

Table 2

Optical nonlinear parameters of sudan iv in DMSO, DMF and chloroform solvents calculated using diffraction ring patterns and CA Z-scan techniques at incident intensities of 9124 W/cm² and 860 W/cm², respectively.

Sample	Diffraction rings			Z-scan
	No. of rings N	$n_2 \times 10^{-6}$ (cm ² /W)	$\Delta n \times 10^{-3}$	$n_2 \times 10^{-7}$ (cm ² /W)
Sudan iv in DMSO	17	0.88	8.04	0.10
Sudan iv in DMF	20	1.03	9.46	0.14
Sudan iv in chloroform	29	1.5	13.71	0.17

distribution, $\Delta\phi(x,y,t)$, passing through sudan iv solutions (a) DMSO left column, (b) DMF middle column and (c) chloroform right column for input power (mW) from top to bottom (9,12,17,26,47). Fig. 10 is direct comparison of chosen input power of (47 mW) diffraction ring patterns in (a) sudan iv DMSO solution, in (b) sudan iv DMF solution and in (c) sudan iv chloroform solution. Blue color is the experimental and red color is the simulation results respectively where good agreement can be seen.

3.3. Z-scan

The nonlinear index of refraction, n_2 , and the nonlinear absorption coefficient, β , of a medium can be obtained via. conducting CA and OA Z-scans respectively. When performing the CA Z-scan experiment using the sudan iv solutions in DMSO, DMF and chloroform, the results are shown in Fig. 11. In this Figure a plot of transmittance against the position for each sample with respect to the lens focal point ($z = 0$) are obtained. The curves shows peaks followed by valleys, as it can be seen from the Fig. 11, an indication that each of the three dye solutions has negative nonlinear indexes of refraction i.e. self-defocusing. The original nonlinearity shown by the sudan iv solutions are thermal in nature which may be attributed to the use of cw laser beam. A horizontal straight lines were obtained when conducting the OA Z-scan experiments, which indicates that the sudan iv in the three solvents does not bare nonlinear absorption coefficients at 473 nm and incident intensity, respectively. Since the origin of observed nonlinearity is thermal in nature, so that the Cuppo et al. model (TLM) [48] can be used. The nonlinear phase shifts, θ , and the nonlinear refractive indexes, n_2 , of sudan iv solutions via. the TLM can be calculated using the following relations [48,49]:

$$|\theta| = \frac{\Delta T_{p-v}}{2} \quad (13)$$

$$n_2 = \frac{\Delta T_{p-v} \lambda}{4\pi L I_0} \quad (14)$$

where ΔT_{p-v} and I_0 are the peaks minus valleys normalized transmittances of the CA Z-scans data and the incident laser beam intensity respectively. Using input power of 5 mW, i.e., intensity $I = 860$ W/cm², so that the values of nonlinear index of refraction of the sudan iv solutions in DMSO, DMF and chloroform, are determined and the values are given in Table 2.

The differences in the values of nonlinear indexes of refraction of the three sudan iv solutions obtained by the diffraction ring patterns and Z-scan are due to the difference in the input intensities used in both techniques where those used in the diffraction ring patterns are larger the Z-scan's.

3.4. Optical limiting

Finding a material that can work as an optical limiter, OL, for the use with the cw, low power laser beam is one of the objectives of the present study. It is an important objective that many researchers have looking for in order to achieve, so that in this subsection the possibility of sudan iv three solutions as OLs are examined using the modified experimental set-up given in subsection 2.3 by measuring the output power corresponding to the input power using a photo detector connected to the power meter. The optical limiting experimental results are shown in Fig. 12. From Fig. 12 it can be seen that the sudan iv in the three solvents showed the properties of the OL. At low input power in each solution it was found that linear change of the output power as the input power increased. However, as the input power continues to increase, this will lead to the appearance of self-defocusing effect in the spot size of the laser beam. As a result of this effect, the laser beam spot size becomes larger than the area of the aperture of the photo detector, resulting in a small increase in the value of the output power since part of the laser beam spot size does not enter the aperture of the photo detector. As a result of self-defocusing the output power will become constant for high input power.

In order to determine that the three sudan iv solutions, can be used as OLs, first the threshold values, T_H , defined as the input power value when the transmittance is reduced to half must be calculated, second, such threshold value must be compared with the values for materials known as OLs. The T_H can be determined by plotting the transmittances of the laser beam through the sudan iv three solutions versus the input power (Fig. 13). The estimated values of T_H for sudan iv in the DMSO, DMF and chloroform are 18.7 mW, 17.3 mW and 15.8 mW respectively. These T_H values can be well compared with materials known as OLs, such as L-Lysine doped Oxalic acid single crystals ($T_H = 18$ mW) [50], Yb:YAG nanoparticles ($T_H = 19.3$ mW) [51], solution of sodium para-nitrophenolate dihydrate ($T_H = 25$ mW) [52], sodium penta borate ($T_H = 35$ mW) [53], graphene oxide ($T_H = 22.5$ mW) [54] and TTCS solution ($T_H = 18.6$ mW) [55]. It seem that sudan iv in the chloroform T_H is less than the values for these materials, therefore, the sudan iv dissolved in the chloroform can be a good candidate to be used as an OL. The mechanism responsible for showing the studied samples

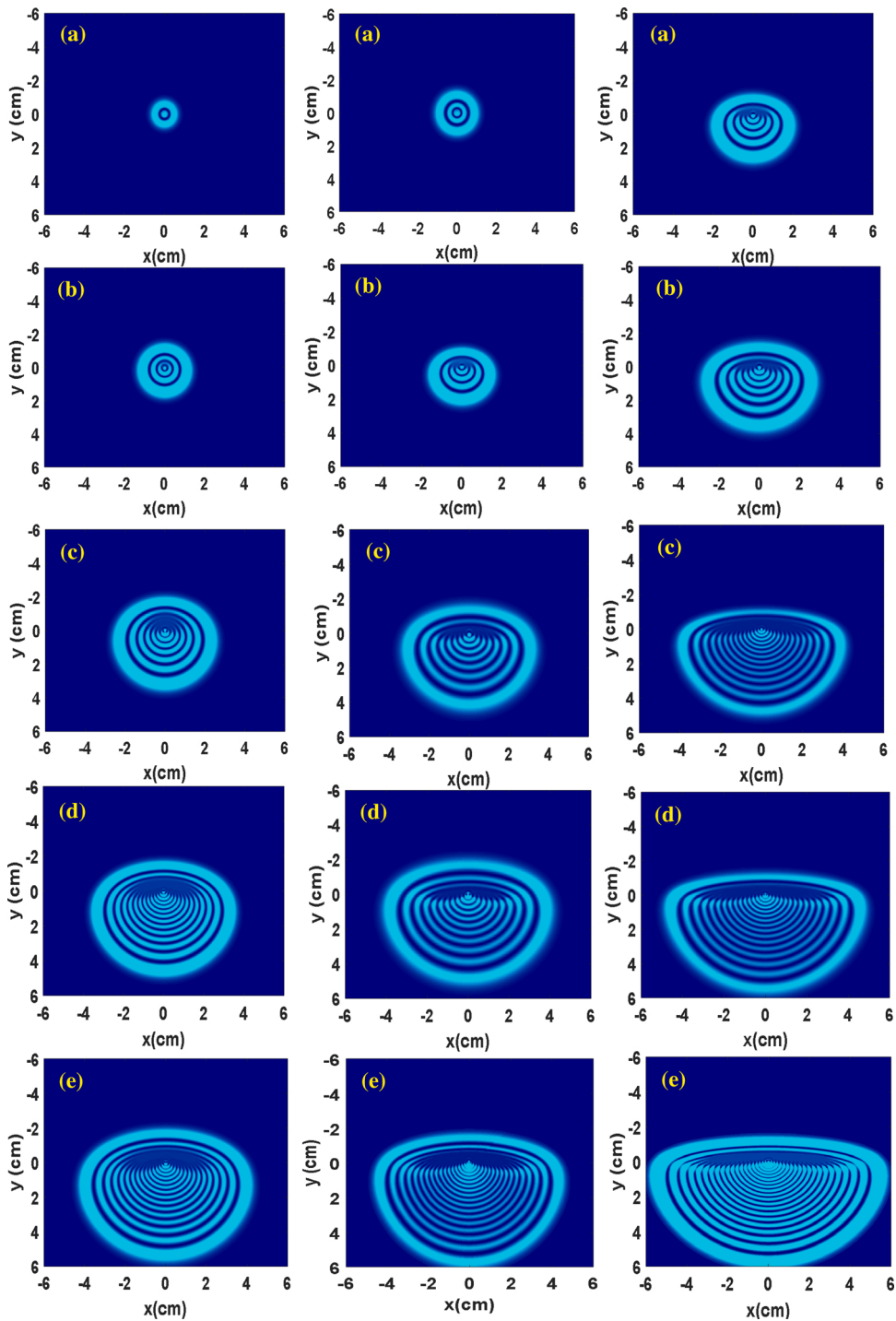


Fig. 5. Simulation results of the diffraction ring patterns for input power (mW) (a) 9,(b)12,(c)17,(d)26 and (e)47 for sudan iv solutions in DMSO (left column), in DMF (middle column) and in chloroform (right column) respectively.

the behavior of the optical limiter is the nonlinear refractive index (thermal lens). According to the obtained results it seem that the refractive index change is responsible for the reduction in the intensity of the transmitted beam.

3.5. Discussion

The resulting diffraction ring patterns are attributed to the thermal nonlinearity due to the localized absorption of part of the tightly

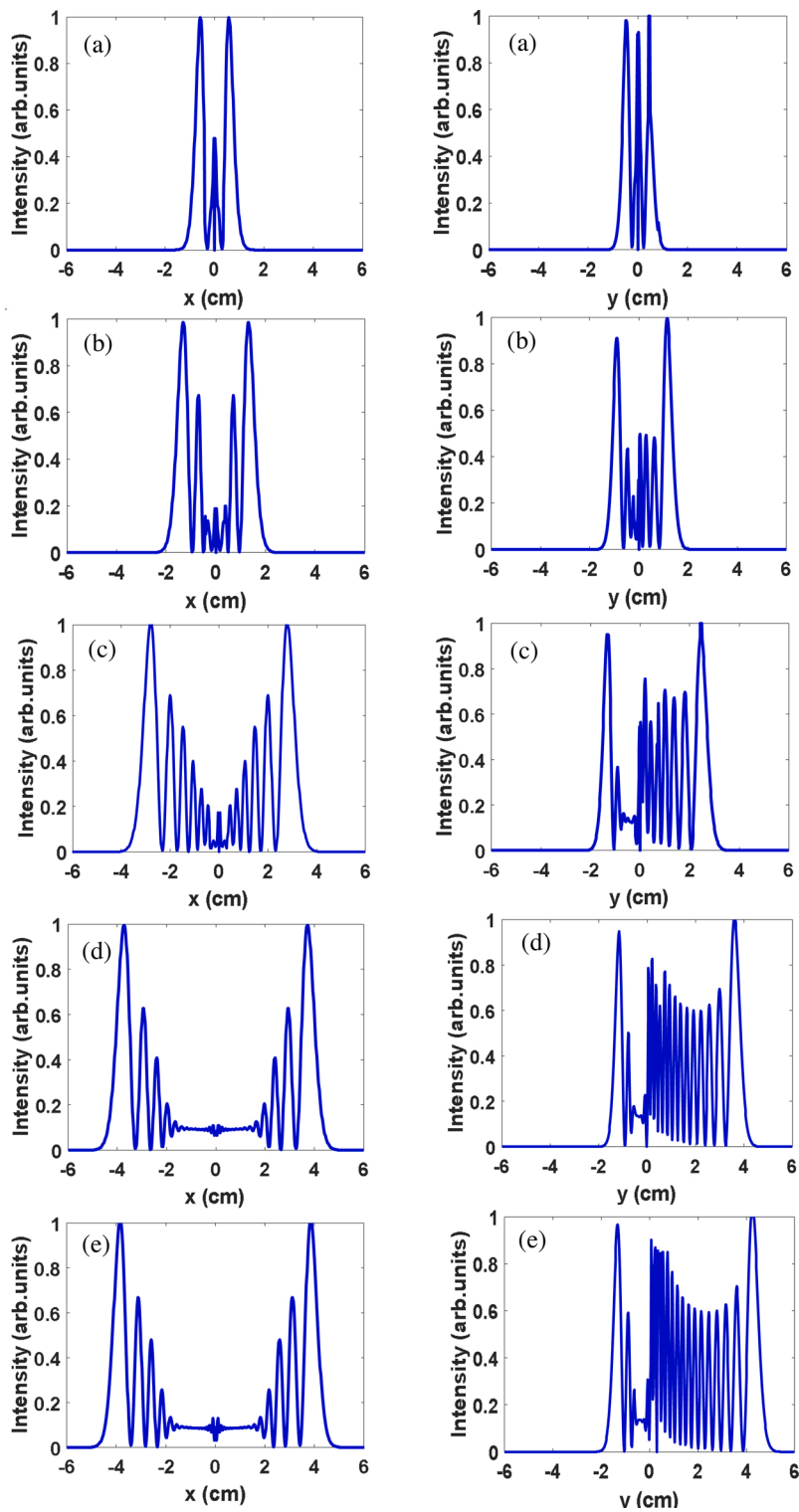


Fig. 6. Simulation results of the one dimensional intensity distribution of the laser beam, left column and right column are the x-axis and y-axis respectively in sudan iv DMSO solution at input power (mW) (a) 9,(b) 12,(c) 17,(d) 26 and (e) 47.

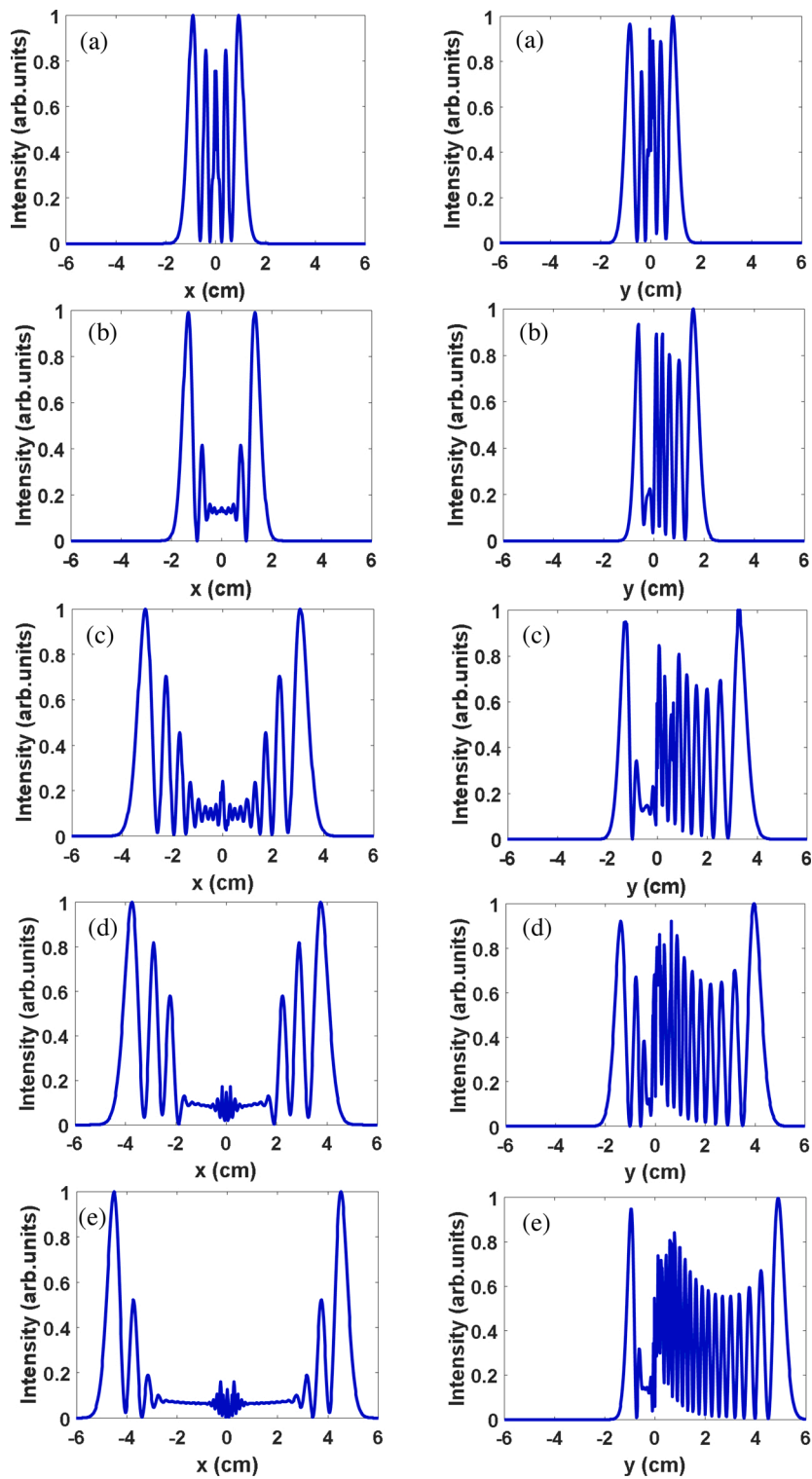


Fig. 7. Simulation results of the one dimensional intensity distribution of the laser beam, left column and right column are the x-axis and y-axis respectively in sudan iv DMF solution at input power (mW) (a) 9,(b) 12,(c) 17,(d) 26 and (e) 47.

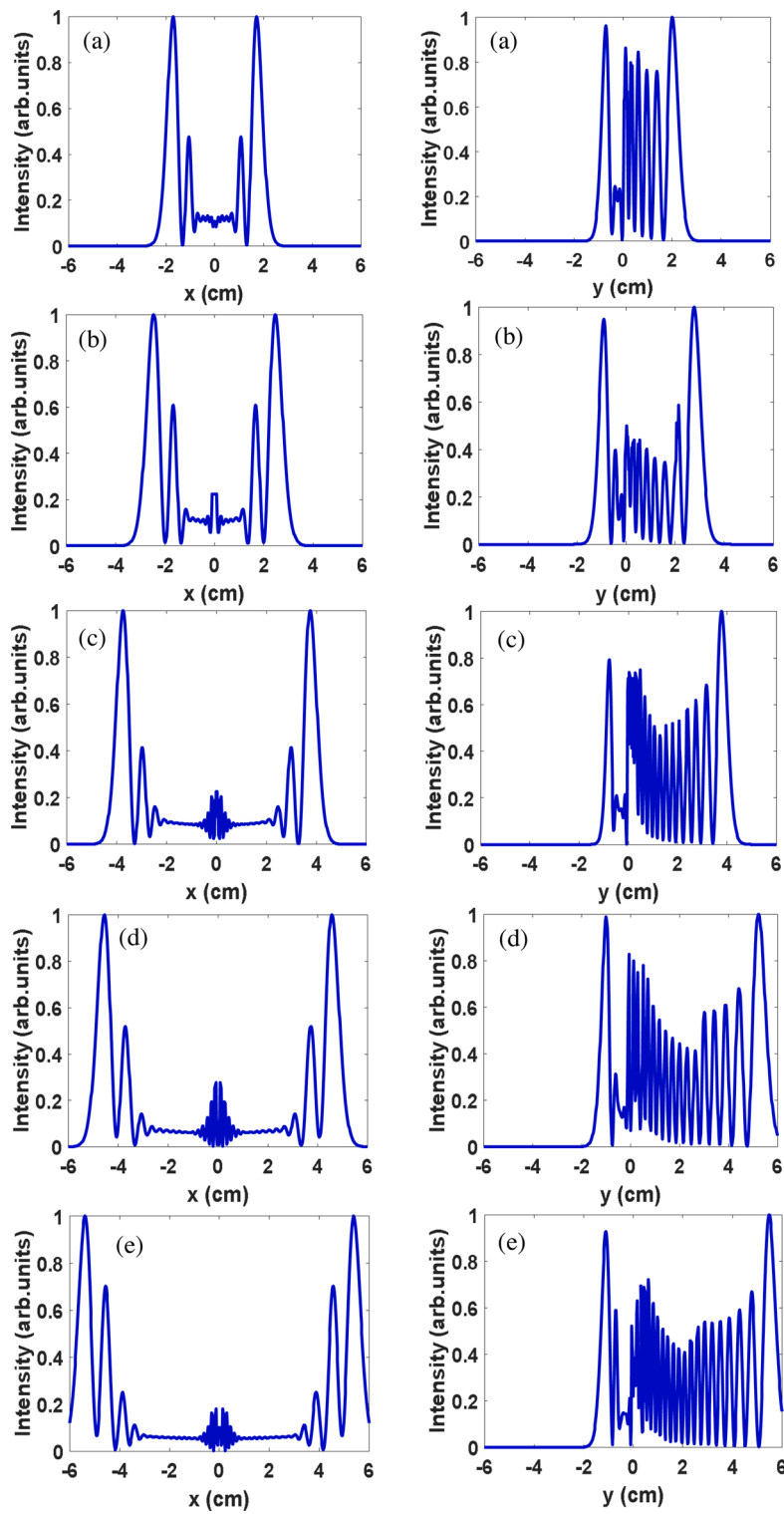


Fig. 8. Simulation results of the one dimensional intensity distribution of the laser beam, left column and right column are the x-axis and y-axis respectively in sudan iv chloroform solution at input power (mW) (a) 9,(b) 12,(c) 17,(d) 26 and (e) 47.

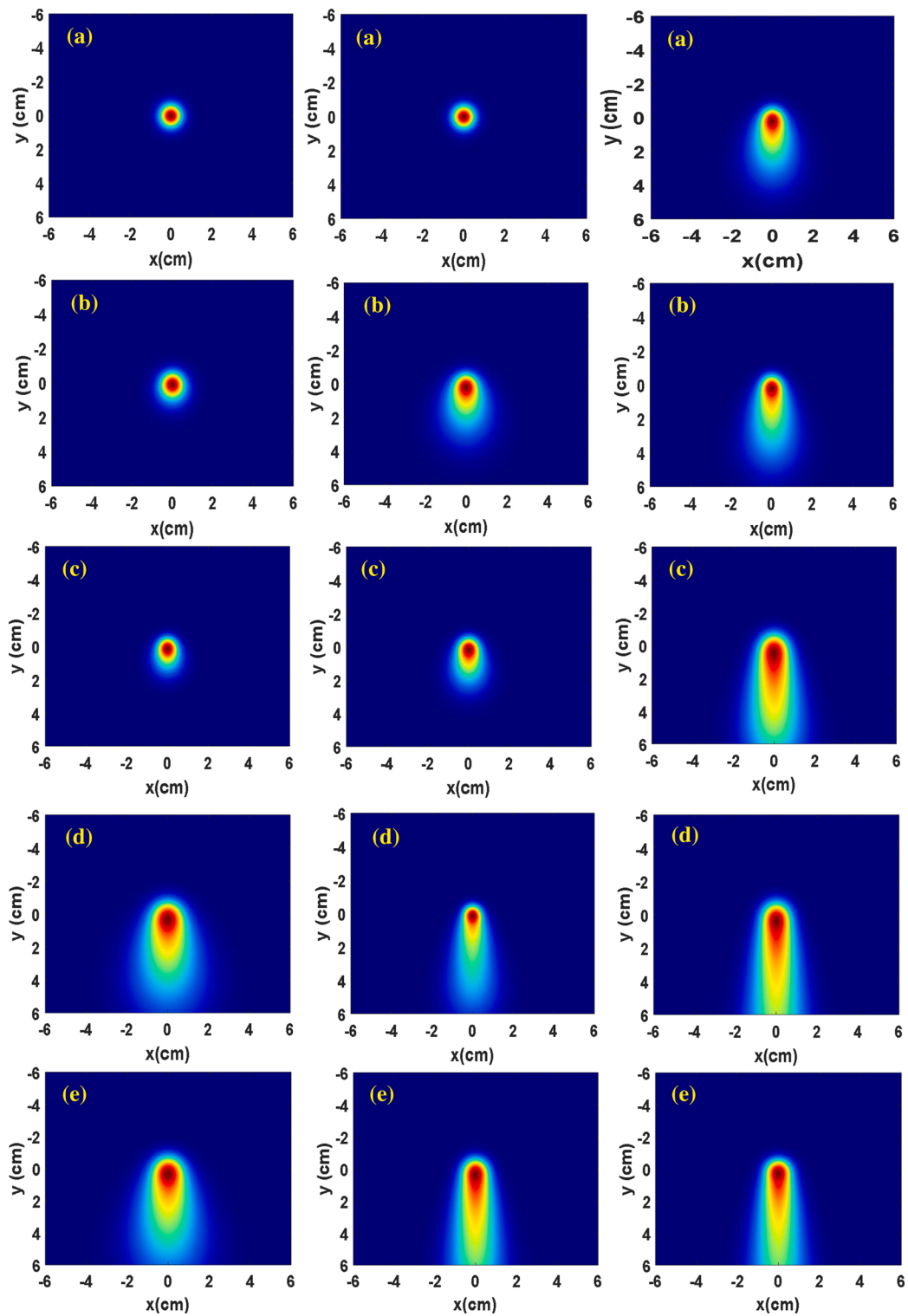


Fig. 9. Laser beam phase spatial distribution, $\Delta\phi(x,y,t)$, as the beam passes through the sudan iv in DMSO, left column, in DMF, middle column, and in chloroform, right column, for input power (mW) (a) 9,(b) 12,(c) 17,(d) 26 and (e) 47.

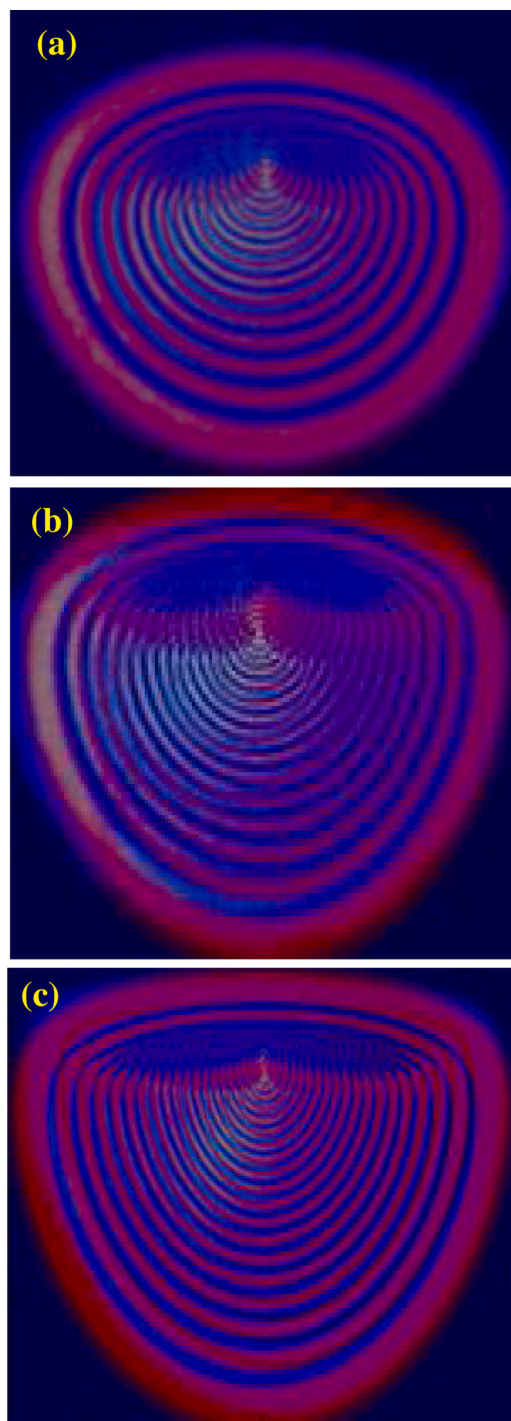


Fig. 10. Direct comparison between experimental (blue) and simulation (red) diffraction ring patterns results for input power of 47 mW in (a) sudan iv in DMSO solvent, in (b) sudan iv in DMF solvent and in (c) sudan iv in chloroform solvent.

focused laser beam energy at 473 nm that propagate in the sudan iv solutions. The spatial distribution of the temperature leads to the spatial distribution of the index of refraction of the dye solutions which act as negative thermal lenses. The negative thermal lens causes a severe phase distortion of the laser beam during its passages through the three samples. It is believed that the nonlinearities in solute dye such as the sudan iv can be enhanced in solvents of low specific heats and reduced vice versa [56]. High thermal conductivity solvents (see Table 1) is able to conduct heat faster resulted via absorption of part of the cw laser beam energy much higher than the low conductivity solvents. Solutions of dye such as those studied in this work have different absorption coefficient, α , as given

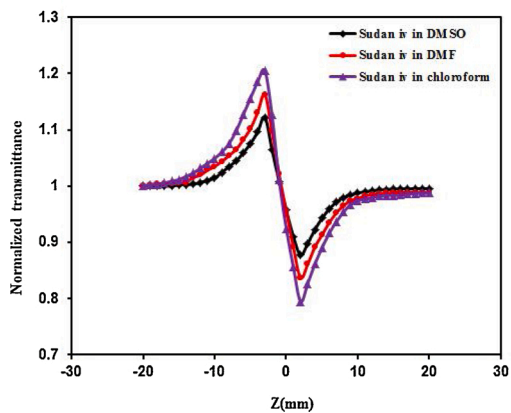


Fig. 11. Normalized CA Z-scan results for sudan iv in DMSO, DMF and chloroform solvents.

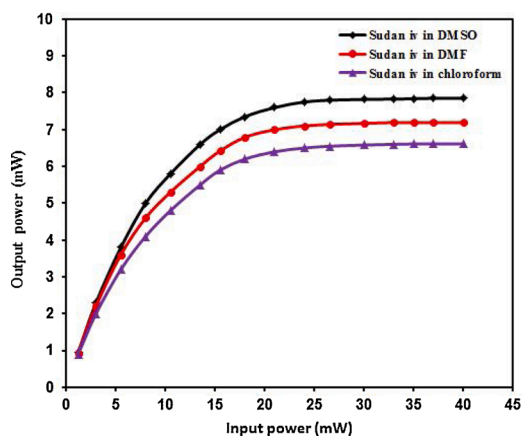


Fig. 12. Output power versus input power for sudan iv in DMSO, DMF and chloroform solvents.

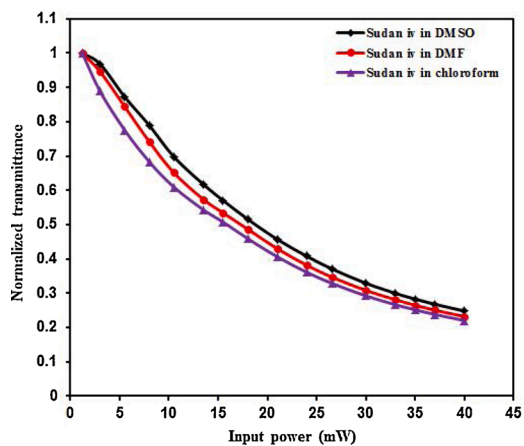


Fig. 13. Normalized transmittance versus input power for sudan iv in DMSO, DMF and chloroform solvents.

in Table 1. The one with high α , means that absorbs more energy from the incident laser beam. Also the sample with low specific heat enhances the nonlinearity more than the one with high specific heat. Taking these three parameters into account leads to the conclusion that sudan iv solute in the solvent chloroform should have the high nonlinearity resulted when a cw laser beam incident on the sudan iv i.e. nonlinear refractive index. The results given in Table 2 agree well with this conclusion. Such results agree with works of other researches [57–61].

4. Conclusions

Diffraction ring patterns resulted when low power, cw, visible, laser beam with TEM₀₀ intensity profile passes in the three sudan iv solutions in DMSO, DMF and chloroform. Each diffraction ring pattern lost symmetry in the upper part due to convective current. Based on the number of rings in each pattern the change in the total refractive indexes and nonlinear refractive indexes in sudan iv solutions are calculated. Diffraction ring patterns are simulated theoretically using the Fresnel-Kirchhoff diffraction theory with good agreement compared to the experimental results. Z-scan technique experiments led to the calculation of the nonlinear index of refraction of sudan iv solutions. Both techniques viz. diffraction ring patterns and Z-scan prove that self-defocusing phenomena occurs in the three sudan iv solutions at the same concentration of 1 mM and low input, cw, laser intensity at 473 nm wavelength. It is proved that the nonlinearities of the sudan iv solutions are controlled by the physical properties of each solvent such as the specific heat and the thermal conductivity. Sudan iv solutions are good candidate for the use as optical limiters.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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