Nonlinear responses and optical limiting behavior of 2-Chloro-5-nitroanisole dye under CW laser illumination

الأستجابات اللاخطية وسلوك الحد البصري لصبغة 2-Chloro-5-nitroanisole تحت إضاءة ليزر مستمر ذو الموجه الموجهه

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Abstract
Nonlinear optical properties of 2-Chloro-5-nitroanisole in chloroform solvent are studied employing different optical techniques. Experiments are performed using the diode laser beam at 532 nm wavelength and 50mW power. The effect of nonlinearity of 2-Chloro-5-nitroanisole in broadening the laser beam is observed. The optical limiting behavior is investigated by measuring the transmission of the samples. The nonlinear absorption coefficient is calculated using the open aperture Z-scan data, while its nonlinear refractive index is measured using the closed aperture Z-scan data. The nonlinear refractive index and absorption coefficient are found to be in the order of $1.24x10^{7}\ cm^{2}/W$ and $8.68x10^{-3}\ cm/W$ respectively. The results indicate that 2-Chloro-5-nitroanisole is a potential candidate for low-power optical limiting application. The optical limiting behavior of the solvent of 2-Chloro-5-nitroanisole is also demonstrated.
Keywords: Optical limiting , nonlinear refraction index , Z-scan , Organic dye

الملخص
دُرِسُت الخواص البصرية اللاخطية ل 2-Chloro-5-nitroanisole في مذيب الكلوروفورم باستخدام تقنيات بصرية مختلفة . أُجريت التجارب باستعمال حزمة ليزر ثنائي بطول موجي 532 نانوميتر وبقدرة 50 ملي واط . لقد لوحظ تاثير لاخطية 2-Chloro-5-nitroanisole في توسيع حزمة الليزر . تم التحقق من سلوك الحد البصري بواسطة قياس نفاذية العينات. معامل الامتصاص اللاخطي قد حسب باستخدام قيم المسح بأتجاه - Z للفتحة المفتوحة بينما معامل الانعكاس اللاخطي تم قياسه باستخدام قيم المسح بأتجاه - Z للفتحة المغلقة .لقد وجد ان معامل الانكسار ومعامل الامتصاص اللاخطي هما من رتبة $1.24x10^{-7}$ سنتمتر مربع / الواط و $8.68x10^{-3}$ سبتمتر/ الواط على التوالي . بينت النتائج ان 2-Chloro-5-nitroanisole هو مرشح محتمل لتطبيق الحد البصري واطئ الطاقة .كذلك تم عرض سلوك الحد البصري لمحلول 2-Chloro-5-nitroanisole.
الكلمات الدليلية: الحد البصري، معامل الأنكسار اللاخطي، المسح بأتجاه - Z ، صبغة عضوية

1. Introduction:
Nonlinear optics has received considerable attention due to their variety of applications in optoelectronic and photonic devices. Especially, nonlinear optical materials exhibiting strong two-photon absorption (TPA) are in great demand, due to their applications in three-dimensional fluorescence imaging and multi- photon microscopy, eye and sensor protection, frequency up conversion lasing, optical signal reshaping and stabilizing fast fluctuations of laser power [1–5]. A wide variety of materials have been investigated for third-order nonlinear optics, among which organic materials are attractive because of their optical and electronic properties which can be tuned and tailored by structural modification. The large and ultra fast nonlinear optical response has made organics particularly attractive candidates for high band width applications. There is quest to design and develop the novel nonlinear materials with large molecular two-photon absorption cross-sections to meet the present demand. Optical limiting is a nonlinear optical process in which the transmittance of a material decreases with increased incident light intensity. It has been demonstrated that optical limiting can be used for pulse shaping, smoothing and pulse compression [6]. The potential applications of optical limiting devices are sensor and eye protection.

In this study, optical nonlinearity induced in dye 2-Chloro-5-nitroanisole by CW diode laser with an output power of 50 mWatt at 532 nm was studied using Z-scan technique, based on the sample-induced changes in beam profile at the far field. The study was made for concentration of dye in chloroform solvent. The nonlinear refractive indices n_2 , and nonlinear absorption coefficients β were measured. The optical limiting behavior of the dye has been studied.

2. Experimental

The molecular structure of the 2-Chloro-5-nitroanisole is shown in Fig. 1. The linear absorption spectrum of the dye solution with the concentration of 0.05 mM in chloroform is shown in Fig. 2, which was acquired using a UV–VISNIR spectrophotometer (Type: CECEL 3500) .

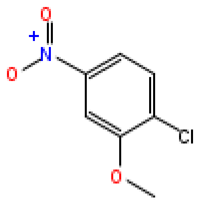

Fig. 1. The molecular structure of the 2-Chloro-5-nitroanisole

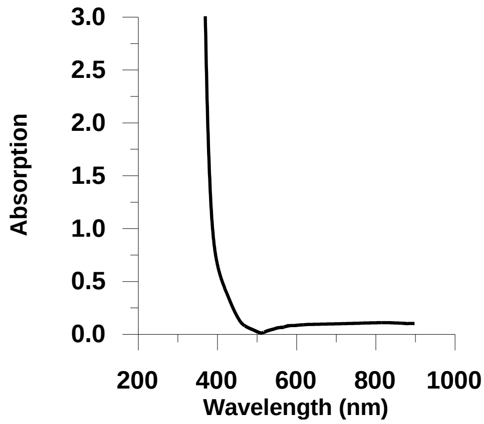

Fig. 2. The absorption of dye in chloroform with the concentration of 0.05 mM.

The schematic diagram of Z-scan technique is as shown in figure 3. By properly monitoring the transmittance change through a small aperture at the far field position (closed aperture), one is able to determine the amplitude of the phase shift. By moving the sample through the focus without placing an aperture at the detector (open aperture) one can measure the intensity dependent absorption of the sample. When both the methods (open and closed) are used in measurements of the ratio of signals determines the nonlinear refraction of the sample.

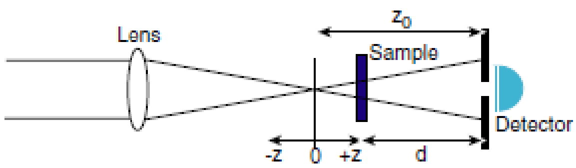

Fig. 3. Z - scan experimental setup.

3. The nonlinear optical measurement

The nonlinear coefficients of the 2-Chloro-5-nitroanisole was measured by using Z-scan technique, which is a well-known technique that allows the simultaneous measurement of both nonlinear absorption coefficient β and the nonlinear refractive coefficient n_2.
A spatial distribution of the temperature in the surface is produced due to the localized absorption of a tightly focused beam propagation through the absorbing sample. Hence a spatial variation of the refractive index is produced which acts as a thermal lens resulting in the phase distortion of the propagating beam. The difference between normalized peak and valley transmittance ΔT_{P-V} can be measured by Z-scan technique. The peak to valley ΔT_{P-V} is linearly related to the on- axis phase distortion $\Delta\Phi_0$ of the radiation passed through the sample **[18].** The relation is defined as,

$\Delta T_{p-v} = 0.406(1-S)^{0.25}|\Delta\Phi_0|$ ………………… (1)

and

$\Delta\Phi_0 = k\, n_2 I_0 L_{eff}$ ………………… (2)

Where $S = 1 - \exp(-2r_a/\omega_a)$ is the aperture linear transmittance with r_a is the aperture radius and ω_a is the beam radius at the aperture in the linear region, I_0 is the intensity of the laser beam at focus $Z = 0$, $L_{eff} = (1 - \exp(-a_0 L))/a_0$ is the effective thickness of the sample, L is the thickness of the sample, (a_0 =0.02788 cm^{-1}) is linear absorption coefficient of the 2-Chloro-5-nitroanisole solution and $k = 2\pi/\lambda$ is the wave number. The nonlinear refractive index n_2 can be obtained from equations 1 and 2, and the corresponding change in the refractive index $\Delta n = n_2 I_0$

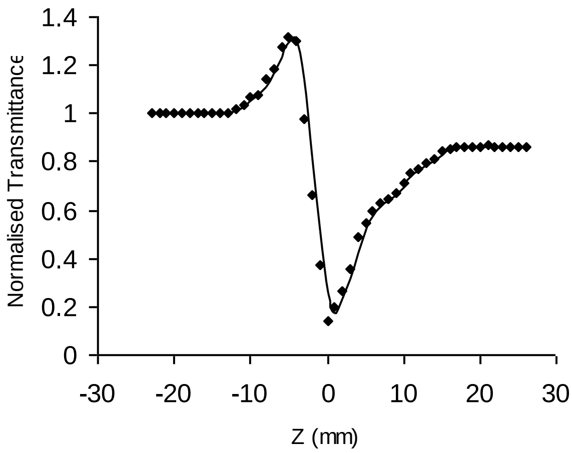

Fig. 4. Closed aperture Z-scan data for 2-Chloro-5-nitroanisole dye when the concentration of the solution is 0.05 mM at 1 mm thickness.

Fig.4 shows the closed aperture (CA) Z-scan curve of 2-Chloro-5-nitroanisole solution. From the asymmetric curve there is obvious nonlinear absorption existing in the solution and the close transmittance is affected by the nonlinear refraction and absorption.

In the Z-scan measurement, the effective thickness of the sample was L_{eff}= 0.998 mm. The linear transmittance of the aperture was $S = 0.39$ and the optical intensity at the focus point is 2.314 $kWatt/cm^2$. From the theoretical fit results, the peak to valley ΔT_{P-V} indicates the negative sign of nonlinear refractive index, n_2 (self-defocusing) for the 2-Chloro-5-nitroanisole solution. The Z-scan signature for sample gives the value of the transmission for peak to valley (ΔT) is 0.033.

The nonlinear absorption coefficient $\beta(cm/W)$ is obtained from fitting performed on the experimental data of the open aperture measurement with the equation.

$T(Z, S = 1) = \sum_{m=0}^{\infty} \frac{[-q_0(Z)]^m}{(m+1)^{3/2}}$ For $|q_0(0)| < 1$ ……………. (3)

where $q_0(z) = \beta I_0 L_{eff}/(1 + z^2/z_0^2)$.

Fig.5 shows the open aperture (S=1) Z-scan curve of sample. The enhanced transmission near the focus is indicative the for TPA process at high intensity ($z = 0$).

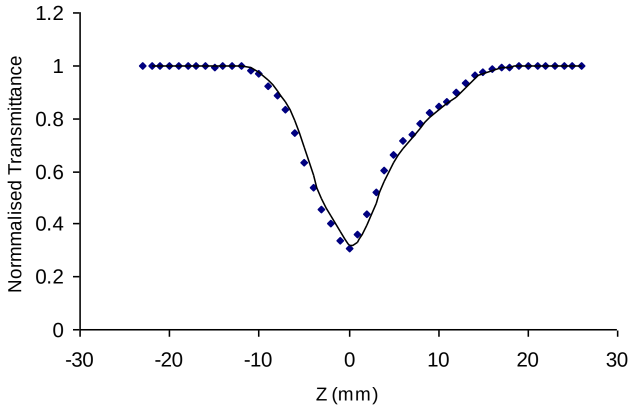

Fig. 5. Open aperture Z-scan data for 2-Chloro-5-nitroanisole when the concentration of the solution is 0.05 mM at 1 mm thickness.

In order to extract the pure nonlinear refraction part, we have computed the value of the closed aperture (CA) data by the open aperture (OA) data. Fig.6 shows the resulting curve corresponding to pure nonlinear refraction. The sign and magnitude of n_2 is determined from the relative position of the peak and the valley width z.

Generally the measurements of the normalized transmittance versus sample position, for the cases of closed and open aperture, allow determination of the nonlinear refractive index, n_2 , and the reversible saturation absorption (RSA) nonlinear coefficient, β [10]. Here, since the closed aperture transmittance is affected by the nonlinear refraction and absorption, the determination of n_2 is less straightforward from the closed aperture scans. Therefore, it is necessary to separate the effect of nonlinear refraction from that of the nonlinear absorption. A simple and approximate method to obtain purely effective n_2 is to divide the closed aperture transmittance by the corresponding open aperture scans.

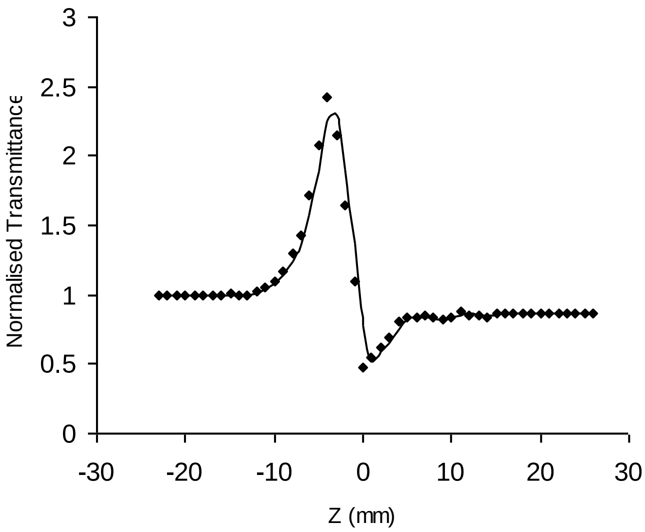

Fig. 6. Close aperture division Open aperture Z-scan data for 2-Chloro-5-nitroanisole when the concentration of the solution is 0.05 mM at 1 mm thickness.

4. Optical limiting

By optical limiting measurement the critical power of laser beam at which the nonlinearity starts to affect the transmission can be measured. For this experiment the sample is put near the focal plane of the lens and the input power is changed. The input power is measured by power meter 2 and power meter 1 is used to measure the output power of focused transmitted beam through the 2-Chloro-5-nitroanisole solution. The pinhole is not used in this experiment. In Fig. 7 the output power of laser beam is plotted as a function of incident power for (0.05 mM) concentration of sample .

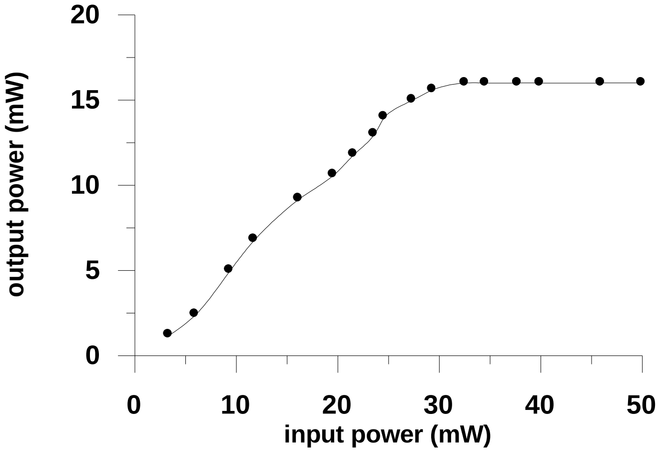

Fig.7. The nonlinear transmission behavior of 2-Chloro-5-nitroanisole of 0.05 mM concentration (1mm thickness).

The input power is in the range of (0–50) mWatt. As is shown clearly for incident beam power of above 25 mW, the transmission becomes nonlinear. At low incident power up to 25 mW, the output power varies linearly with a ratio of 0.73 for 0.05 mM concentration of 2-Chloro-5-nitroanisole solution. At incident power above 26 mWatt the output power tends to be constant, because its nonlinear absorption coefficient increases with increase in the incident irradiance. Generally in liquids, where the thermal expansion is large, high absorbance of the nonlinear material at the corresponding wavelength leads to increase in the temperature and density of the sample. Heating due to laser absorption is the responsible mechanism for changing the absorption coefficient and optical limiting effect [11-13]. Results confirm that 2-Chloro-5-nitroanisole solution is a good candidate for optical limiting at 532 nm CW lasers.

Conclusion

The nonlinear refraction index n_2 and nonlinear absorption coefficient β , of 2-Chloro-5-nitroanisole solution was studied using a single beam Z-scan technique under CW laser with excitation at 532 nm. The Z-scan measurement indicates that the sample exhibited large nonlinear optical properties. n_2, β and Δn values was found to be $1.24x10^7\ cm^2/W$, $8.68x10^{-3}\ cm/W$ and $2.86x10^{-4}$, respectively. From Fig 7 a, b there is an increasing trend for the values of n_2 and β as the concentration increases. This may be attributed to the fact that, as the number of dye molecules increases when concentration increases, more particles are thermally agitated resulting in an enhanced effect. The optical nonlinearity of the dye may be due to laser heating induced nonlinear effect. A laser beam, while passing through an absorbing media, induces temperature and density gradients that change the refractive index profile. This intensity-induced localized change in the refractive index results in a lensing effect on the optical beam. Thus the sample with many attracting linear and nonlinear optical properties is suitable candidate for optoelectronic applications. Based on nonlinear refraction the sample behaved as good optical limiters at low incident power up to 25mWatt, indicating this sample find potential applications in optical limiting and signal processing applications.

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