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Optical Nonlinear Properties and Optical Switching of Henna (Lawson) Films

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Abstract : The nonlinear absorption coefficient and nonlinear refractive index have been investigated for Henna (Lawson) (2-hydroxyl-1,4 naphthoquinone) film using Z scan experiment at wavelengths 488 nm 514 nm and 633nm. The Z-scan measurements indicated that the Henna sample exhibited large nonlinear optical properties at low input power. The sample exhibited saturation absorption process at wavelengths 488 nm and 514 nm and exhibited a reverse saturation absorption process at wavelength 633nm. Intensity dependence of the nonlinear absorption β reveals that the Henna can be considered as a potential candidate for optical limiting at 633 nm. The optical nonlinearities were utilized to demonstrate all optical switching.
Key words: nonlinear absorption, nonlinear refractive index, optical switching, Henna, Lawson'2- hydroxyl-1,4 naphthoquinone

1. Introduction

There has been a considerable interest in searching for materials exhibiting nonlinear optical effects. These effects are of great technological importance for use in future applications within electronic and photonic devices [1, 2]. An example of interest that has attracted a lot of attention is the light intensity induced refractive index changes and absorption changes. The nonlinear refractive index changes are commonly described by the relationship $n= n_o+n_2I$, where n_o is the linear refractive index, I is the intensity of the light and n_2 is a nonlinear refractive index coefficient. The nonlinear absorption coefficient changes are described by $\alpha= \alpha_o+\beta I$, where α is a linear absorption coefficient and β is a nonlinear absorption coefficient. These coefficients are an effective parameter that contains many interesting nonlinear optical effects, such as laser induced grating, soliton pulse propagation in waveguides [3,4], optical switching [5-7] and self- focusing, self- defocusing, self-phase modulation [8-11]. Light induced nonlinear optical effects; in general have applications such as optical bistability optical limiting [12-13]. Many materials such as photo refractive materials [14], Buckminster-fullerenes [15,16], fluoride glasses [17] and polymers [18] have been investigated for these applications.

This work presents measurements of the nonlinear absorption coefficient and nonlinear refractive index of Hanna (Lawson) (2- hydroxyl-1,4 naphthoquinone) film with a low power continuous wave (cw) laser at wavelengths 488nm, 514nm and 633nm using the Z- scan technique[19]. The pump and probe technique was used to demonstrate optical switching nonlinearity. Henna has been studied for solar cell applications [20]. To our knowledge the Henna has not been previously considered for optical nonlinearity measurements.

2. Experimental details

Henna is extracted from a plant called *Lawsonia Interims* whose colour is due to the compound called Lawson (2- hydroxyl-1,4 naphthoquinone) [21]. The molecular structure is shown in figure 1a. A soxhlet extractor was used for the extraction of dye from 80 g of Bahraini Henna leaves, where 100ml of methanol is used for the extraction process. Henna film was fabricated by depositing the collected extract on the glass substrate slide and by casting the film under the atmospheric pressure. The film was left in the incubator at 37^{o}C for twenty four hours. The linear absorption of the film is shown in figure 1b

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The Z-scan technique was used to measure the nonlinear properties of Henna thin film. This technique relies on the fact that the intensity varies along the axis of the convex lens and is maximum at the focus. Hence, by shifting the sample through the focus, the nonlinear refraction can be measured by observing

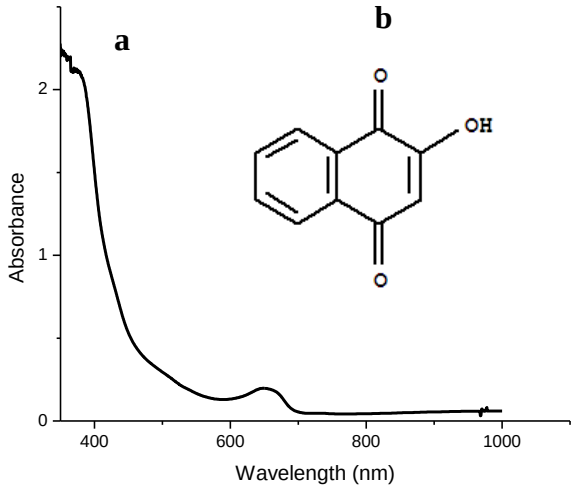

Fig.1. (a) Linear absorption of the Henna Dye (Lawson) film.
(b) Chemical structure of the Henna Dye (Lawson).

The spot size variation at the plane of finite aperture /detector combination. The experiment was performed with an air-cooled Ar ion laser beam operating at 488 nm and 514nm and He-Ne laser at 633 nm with an average power of 20 mW. The beam was focused to a beam waist of 20 μm with a lens of 5 cm focal length, giving a typical power density of 1.3x10 ^{7} W/m^{2}. The transmission for the sample was measured with and without an aperture in the far-field of the lens, as the sample moved through the focal point. This enables the nonlinear refractive index (closed aperture) to be separated from that of the nonlinear absorption (open aperture).

3. Results and discussion

The nonlinear refraction of the sample causes a spatial beam broadening or narrowing in the far field and thus modifies the fraction of light that passes through the aperture as the sample position is changed. A typical peak–valley (valley–peak) transmittance curve is obtained when the nonlinear refractive index of the medium is negative (positive). It is possible from the peak to valley variation of the measured transmittance curve to measure the nonlinear refractive index.
The difference between normalized peak–valley transmittance $\Delta T_{p\text{-}v}$ is given by

$$\Delta T_{p-v} = 0.406(1-S)^{0.25}\left|\Delta\phi\right| \quad (1)$$

where $\Delta|\phi|$ is the on axis nonlinear phase shift at focus and S is the linear transmittance of the aperture and is given by

$$S = 1-\exp(-2r_a^2 / w_a^2) \quad (2)$$

where r_a is the radius of the aperture and W_a is the radius of the laser at the entrance of the aperture. The nonlinear phase shift is given by

$$\Delta|\phi| = \frac{2PL_{eff}}{\lambda w_o^2} n_2 \quad (3)$$

where n_2 is the nonlinear refractive index, P is the laser power, λ is the laser wavelength, w_o is the beam waist at focus and $L_{eff} = (1-\exp(-\alpha_o d))/\alpha_o$ with α is linear absorption coefficient at 514 nm, d is the sample thickness and L_{eff} is the effective thickness of the sample.
The normalized transmittance through closed aperture for 100 μm thick Henna film is shown in Fig. 2. This normalized transmission curve is characterized by a prefocal peak followed by a postfocal valley. This peak valley configuration implies that the nonlinear refractive index of the film is negative (n_2<0).

The a bove equations were used to determine the value of refractive index n_2 at three different wavelengths (488nm, 514nm,633nm) at intensity $1.34x10^7$ W/m^2 . The results are summarized in table 1. These values are of two orders larger than the nonlinear refractive index measured in CdO film [22] and

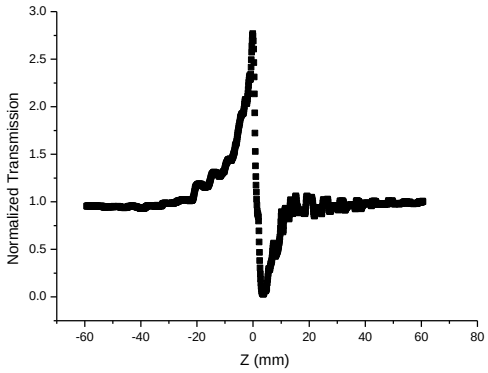

Fig. 2. Normalized transmittance (closed aperture) of Henna film at 488 nm at peak intensity of 1.34 x10^7 w/m^2.

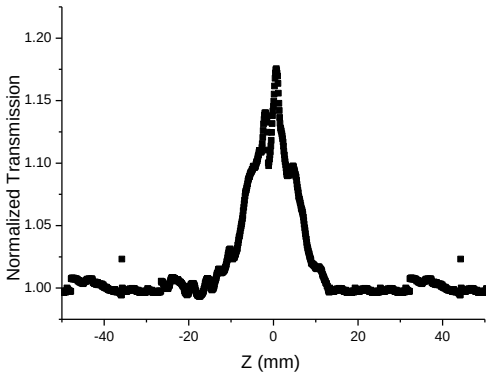

Fig.3. Normalized transmittance (open aperture) of Henna film at 514 nm at peak intensity of 1.34 x10^7 W/m^2

one order smaller than the nonlinear refractive index measured in Phthalocyanine thin film of PMMA [23]. The open Z- scan was performed at wavelengths 488nm, 514nm and 633nm. Fig.3 shows the normalized transmittance without an aperture as a function of the distance along the lens axis z, for a 100 μm thick Henna film at 488nm. The transmission is symmetric with respect to the focus (z =0), where it has a maximum transmission. This demonstrates that the sample exhibits Saturation Absorption,(SA) (negative nonlinear absorption effect), similar effect was observed at 514 nm.

Fig. 4 shows the normalized transmittance without an aperture at 633nm. The transmission is symmetric with respect to the focus (z =0), where it has a minimum transmission. This demonstrates that the sample exhibits Reverse Saturation Absorption,(RSA) (positive nonlinear absorption effect),

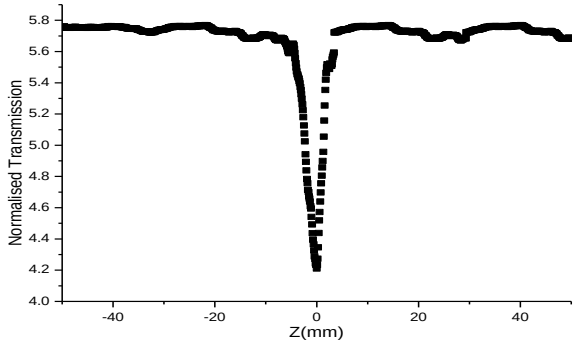

Fig. 4 Normalized transmittance (open aperture) of Henna film at 633 nm at peak intensity of 1.34 x10^7 W/m^2.

The normalized transmission for the open z scan is given by

$$T = 1 - \left(\frac{1}{2\sqrt{2}} \frac{\beta I_o L_{eff}}{(1+x^2)}\right) \tag{4}$$

where x = z / z_0 (with $z = w_o^2 / \lambda$) is the diffraction length of the Gaussian beam and w_o is the beam's waist, and $L_{eff} = (1 - \exp(-\alpha_o d)) / \alpha_o$ with α_o is a linear absorption coefficient and d is the sample

thickness, L is the effective thickness of the sample. I_o is the intensity of the laser beam at the focus. The normalized transmission T_o at the focal point (z =0) is

$$T_o = 1 - \frac{\beta I_o L_{eff}}{2\sqrt{2}} \tag{5}$$

for nonlinear absorption coefficient β the equ. 5 can be written as

$$\beta = (1 - T_o)\frac{2\sqrt{2}}{I_o L_{eff}} \tag{6}$$

Equ. (6) is used to calculate the values of nonlinear absorption coefficient β at a three different wavelengths (488nm, 514nm,633nm) at intensity 1.34×10^7 W/m^2 .The calculated values of β are summarized in Table 1

The nonlinear absorption coefficient β is related to the Δσ and given by [24]:

$$\beta = \frac{\lambda N_o \Delta\sigma}{4 \pi I_s} \tag{7}$$

where $\Delta\sigma = \sigma_{ex} - \sigma_g$ is the difference between excited state absorption cross section σ_{ex} and ground-state absorption cross section $\sigma_g = \alpha_o / N_o$, N_o is the total concentration (N_o= 2.27 x10^{18} m^{-3}) and I_s is a saturation intensity. In the case when only SA is present, the normalized transmission related to saturation intensity I_s given by

$$T = 1 - \left[\frac{1}{2\sqrt{2}}\left[\frac{\alpha_o I_o L_{eff}}{(1 + I/I_s)(1 + x^2)}\right]\right] \tag{8}$$

The fit of Eq. (8) to the z scan data the experimentally determined value of I_s and are shown in table 1. The ground state absorption cross section was calculated from $\sigma_g = \alpha_o / N_o$. Using Eq.7, the excited state absorption cross sections σ_{ex} were calculated for the wavelengths used in the experiment are shown in Table 1. The values of nonlinear absorption coefficient reported here is in agreement with the values reported for C60 and fast green FCF Dyes at 633 nm [23, 25, 26].

Table: 1 Shows the values of absorbance A, ground state absorption cross section σ_g, excited state absorption cross section σ_{ex}, saturation intensity I_s, nonlinear refractive index n_2 and nonlinear absorption β for Henna film.

λ nm	A	σ_g (cm^2)	σ_{ex} (cm^2)	I_s (W/m^2)	n_2 (cm^2/W)	β (cm/W)
488	0.331	2.44 x10^{-17}	1.99 x10^{-17}	9 x10^5	5.4 x10^{-6}	5.27 x10^{-5}
514	0.256	2.01 x10^{-17}	7.4 x10^{-17}	1.2 x10^6	2.6 x10^{-6}	4.44 x 10^{-6}
633	0.185	1.5x10^{-17}	1.25 x10^{-16}	2.2 x10^6	4.6 x10^{-6}	6.76 x 10^{-6}

It is a clearly evident from figures 3 and 4 that the open aperture scans indicating the intensity dependent absorption. This intensity dependant is due to Saturation Absorption (SA), (fig 3) and Reverse saturation Absorption (RSA), (fig 4). The saturation absorption may be explained by considering three level systems consists of HOMO band state, LUMO band state and other states/state generated by impurity or defects. When the sample is exited by a wavelength close to the band gap, the molecules are excited from the HOMO to LUMO band states. As the intensity increases the depopulation of HOMO band may occur and this lead to increase in the transmission. This phenomenon occurs when the excited state absorption cross section σ_{ex} is smaller than the ground state absorption cross section σ_g. The calculated values of σ_{ex} and σ_g at wavelengths 488 nm and 514 nm shows that $\sigma_{ex} < \sigma_g$ and the values are shown in Table 1 , in this case the increased transmission observed in the Fig. 3 is due to saturation absorption (SA). The decrease of the transmission with increasing the intensity observed for Henna film at 633 nm may be explained as follow: after population of LUMO band, the excited molecules decays to the one of impurity/ defect states and since the decay time from this state to the HOMO state is long the population on these states will build up. At higher intensity the molecules from these states may be re-excited back to LUMO band or to higher defect states results in the decrease in the transmission. This phenomenon occurs when the excited state absorption cross section σ_{ex} is larger than the ground state absorption cross section σ_g. The calculated

values of σ_{ex} and σ_g at wavelengths 633 nm shows that $\sigma_{ex} > \sigma_g$, and displayed in table 1, in this case the decrease in the transmission observed in the Fig 4. is due to Reverse Saturation Absorption (RSA). These results suggest that the nonlinear properties of Henna are wavelength dependant where at 488nm and 514 nm the material behaves as SA while at 633 it behaves as RSA.

Fig 5 shows that the calculated value of β at wavelength 633nm, decreases with increasing the input intensity which is a signature of optical limiting process, therefore Henna can be considered as a potential material for optical limiting.

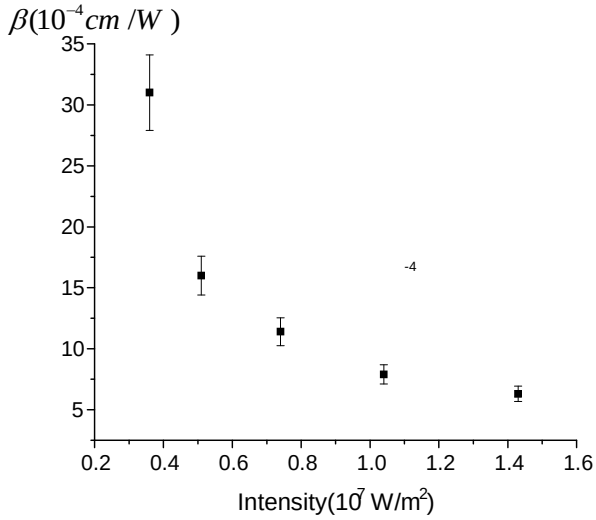

Fig . 5. Variation of nonlinear absorption coefficient β at λ= 6 33 nm as a function of input intensity

The pump and probe experiment was used to demonstrate inverted optical switching. The laser beam from He-Ne laser (λ=633 nm, 20 mW) was used as a pump beam and a weak laser beam from Nd:YG laser (λ=532 nm,1 mW) was used as the probe beam. Both beams were collinearly focused on the sample by the same lens. The probe beam was isolated from the pump beam using a filter and monochromator. The experiment was performed by moving the sample to a postfocusing position (minimum transmission in Z scan experiment). The pump beam was modulated with a mechanical chopper beam. The input of the pump beam and probe beam transmission was detected by photodiode and displayed on a PC oscilloscope (BitScope BS12000). Fig. 5 shows the traces of the waveforms for the pump beam (upper trace) and the probe beam (lower trace). It is clear from the figure that when the pump beam is "on state", the probe beam is intensively absorbed and is in "off state. Similar effect is demonstrated in Phthalocyanine [5]. Henna material is under investigation for use other logic gates such as AND, NOR, etc.

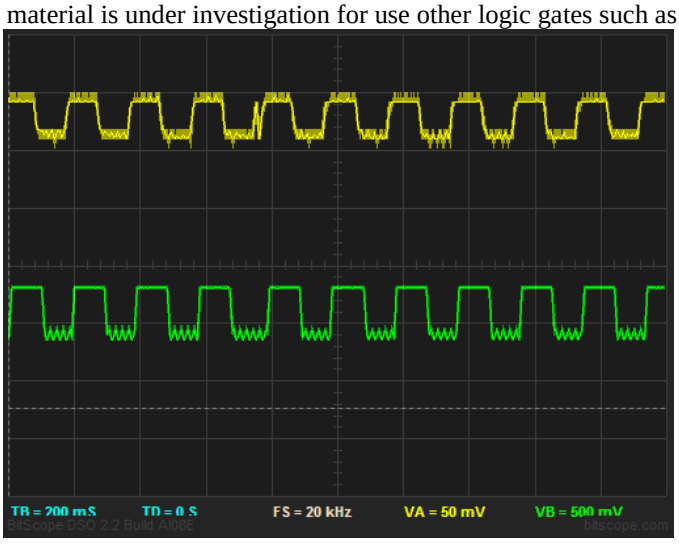

Fig.6. Waveform traces for the probe beam (upper trace) and the pump beam (lower trace).

In conclusion, nonlinear absorption coefficient and nonlinear refractive index have been investigated for Hanna thin films using Z scan experiment at wavelengths 488 nm 514 nm and 633nm. The Z-scan measurements indicated that the Henna exhibited large nonlinear optical properties at low input power. The sample exhibited saturation process

at wavelengths 488 nm and 514 nm and exhibited a reverse saturation absorption process at wavelength 633nm. The intensity dependence of the nonlinear absorption β reveals that the material can be considered as a potential candidate for optical limiting at 633 nm. Optical switching based on absorption of the probe was demonstrated. Investigations are in progress to study optical limiting behavior of the Henna at wavelengths higher than 633nm. Also the studies of the physical mechanism involved in optical switching is in progress. Low power pumping is important for device manufacturing with respect to cost and compactness and threshold damage. Henna has advantages over other materials for its stability and its viability in nature.

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