

## Study the Third Harmonic Parameters for Different Liquid Laser Media

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### Abstract

Three different ethanol solutions of laser dyes as ( cryptocyanine ,Oxazine 4 as well as Cresyl violet ) had been prepared at ( $5 \times 10^{-4}$  M) , and study the linear optical parameters as (maximum absorption wavelength, absorbance  $A$ , transmittance  $T$ , absorption coefficient  $\alpha$  and refractive index  $n$  for three different solutions . Tm: YLF laser at 1907.8 nm , had been used to study the parameters of Third Harmonic generation in these solutions by using two cells from the same solution depending on the third harmonic power generated from them . Ordinary Refractive index  $n_1$  , 3<sup>rd</sup> harmonic refractive index  $n_3$  , the difference  $\Delta n$  between  $n_1$  and  $n_3$  , 3<sup>rd</sup> harmonic absorption coefficient  $\alpha_3$  , 3<sup>rd</sup> harmonic electrical conductivity  $\sigma_{elect}(3)$  , 3rd harmonic extinction coefficient  $\epsilon_{3\omega}$  and the phase shift  $\Delta\phi$ , gradient of 3<sup>rd</sup> harmonic electric field  $\frac{\partial E_3}{\partial z}$  , and conversion efficiency  $\eta$  , 3<sup>rd</sup> optical conductivity  $\sigma_{opt}(3)$  , The coherence length  $L_c$  , The optical density  $d$  , had been studied for these solutions at different Tm: YLF laser power in one time and at different laser incident angle on the solution cell in another time. It can be concluded that the Oxazine 4 laser dye is the best studied laser dye in 3rd harmonic generation efficiency; while the Cresyl Violet laser dye gives highest 3rd harmonic properties than other studied laser dyes. The main conclusion is that the incident laser power on the liquid laser dyes has the strongest effects in all the 3<sup>rd</sup> harmonic properties.

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**Keywords**— THG , Tm:YLF laser , cryptocyanine ,Oxazine and Cresyl violet laser dyes

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### Introduction

Cryptocyanine is the Synonym for 1,1'-Diethyl-4,4'-carbocyanine iodide and has the empirical Formula of  $C_{25}H_{25}IN_2$  It has the molecular Weight of 480.38. It has been classified as a laser dye of melting point of (250.5-253) °C, and the maximum laser wavelength at 648 nm and 703 nm.

In 1969, B. L. Booth, studied the absorption and fluorescence spectral properties of Cryptocyanine laser dye and the Breakdown of Cryptocyanine Dye using Ruby Laser Pulses[1].

In 1972, Roberta A. Hollier et al., Ultraviolet radiation within wavelength band adjacent to 300 nm created by the flashlamps used to pump the ruby rod, is accountable for each photochemical decomposition (irreversible bleaching) of methanolic solutions of cryptocyanine employed as passive shutters (Q-switches) in high-peak-power ruby lasers. The laser beam itself has slight or no irreversible influence on cryptocyanine[2].

In 1992, Thanga Thevar and J. Watson, presented the consequences and investigation of carried out experimentations on a dye Q-switch to assess its operating physical characteristics. The work has been achieved on a ruby laser, Q-switched by cryptocyanine in methanol solution. Performance examinations based on pump energy, output energy, dye absorbance and timing of pulse emergence are in detail depicted. These outcomes have been used to derive the efficiency of Q-switching for variable dye absorbance. Lastly, optimizing of Q-switch performance for diverse supplies of the laser was considered [3].

The Oxazine laser dye has the formula of  $C_{21}H_{22}ClN_3O_5$  and it has Molecular Weight of 431.87.

In 1976, K. H. Drexhage, et al., studied the fluorescence efficiency of oxazine dyes. Relationships with the molecular structure had investigated and dependency on solvent and temperature was clarified. Numerous fluorescence standards were suggested in this study [4].

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In 2007, A.Czímerová et al., studied the investigation of energy transfer between the laser dyes rhodamine 3B (R3B) and oxazine 4 (Ox4) adsorbed on the surface of synthetic Sumecton saponite (Sum). The process of energy transfer was studied for both saponite dispersions and oriented solid films[5].

In 2011, Amitabha Chakraborty et al., studied the influence of temperature on the structural geometry of the dimer (oxazine 4) in solution in conjunction with the dimerization equilibrium based on hydrophobic and electrostatic interactions[6].

The Cresyl Violet laser dye of  $C_{18}H_{15}N_3O_3$  molecular structure, has the Molecular Weight of (321.32 g/mol) , and it has the following properties as; 140 - 143 °C of melting point [7].

In 1972, P. Gacoin and Pierre Flamant studied the spectroscopic features of cresyl violet dye were investigated. A technique of purification was described to make possible increasing in the laser energy output at 665 nm by a factor of 4. The efficiency of  $1.7 \times 10^{-3}$  acquired with flashtube pumping was analogous with that of rhodamine 6G [8].

In 2003, Basheer Ahamed et al., presented theoretic and tentative study of the features of Nd: YAG laser pumped energy transfer distributed feedback Cresyl Violet dye laser [9].

In 2004, Michael J Holmes and Carl Mungan investigate the speed with which an optical dye in a solid plastic host conclusively lowers as it is luminously illuminated by visible light. Precisely, the organic dye Cresyl Violet perchlorate dispersed in plexiglas has optically motivated by a continuous-wave dye laser pumped through an argon-ion laser[10].

In this research we will study the third harmonic generation characteristics as; Ordinary Refractive index  $n_1$  , 3<sup>rd</sup> harmonic refractive index  $n_3$  , the difference  $\Delta n$  between  $n_1$  and  $n_3$  , 3<sup>rd</sup> harmonic absorption coefficient  $\alpha_3$  , 3<sup>rd</sup> harmonic electrical conductivity  $\sigma_{elect(3)}$  , 3rd harmonic extinction coefficient  $exc_3$  and the phase shift  $\Delta\phi$ , gradient of 3<sup>rd</sup> harmonic electric field  $\frac{\partial E_3}{\partial z}$  , and conversion efficiency  $\eta$  , 3<sup>rd</sup> optical conductivity  $\sigma_{opt(3)}$  , The coherence length  $l_c$  , The optical density  $d$  , had been studied for ( cryptocyanine , Oxazine 4 as well as Cresyl violet ) laser dyes solutions at different Tm: YLF laser power in one time and at different laser incident angle on the solution cell in another .

Theory :

Linear Optical properties :

The transmittance  $T$  of medium which is related with the absorbance (  $A$  ) that refers to the amount of absorbed photons by molecules , can be computed by [11-14]:

$$T = 10^{-A} \dots\dots\dots (1)$$

The absorption coefficient  $\alpha$  of an optical medium is related to the  $A$  absorbance based on eq.(2) [12- 13] :

$$\alpha_1 = \frac{1}{2.302A} \dots\dots\dots (2)$$

The refractive index  $n_0$  is associated with transmittance of medium by [12-14]:

$$n_0 = \frac{2 + \sqrt{4 - 4T^2}}{2T} \dots\dots\dots (3)$$

Third Harmonic properties:

The 3<sup>rd</sup> harmonic non-linear refractive index (Extra ordinary refractive index)  $n_3$  which is resulted by generated extraordinary 3<sup>rd</sup> harmonic wave due to applied laser , can be related with ordinary refractive index  $n_1$  which is resulted by Ordinary wave , as the following equation [ 15 ]:

$$n_3 = 3 n_1 \dots\dots\dots (4)$$

Where  $n_1$  can be computed using eq. (3) but for applied laser case. The difference between  $n_3$  and  $n_1$  can be given as [ 16-17 ]:

$$\Delta n = n_3 - n_1 \dots\dots\dots (5)$$

The 3<sup>rd</sup> harmonic non-linear absorption coefficient may be given as [ 18 ] :

$$\alpha_3 = \frac{-\ln\left(\frac{P_{third}}{P_0}\right)}{\ell} \quad \dots\dots\dots (6)$$

The 3<sup>rd</sup> harmonic non-linear optical conductivity can be formulated as [19 ] :

$$\sigma_{opt(3)} = \frac{\alpha_3 n_3 c}{4\pi} \quad \dots\dots\dots (7)$$

The 3<sup>rd</sup> harmonic non-linear electrical conductivity can be formulated as [ 20 ] :

$$\sigma_{elect(3)} = \frac{Z \lambda \sigma_{opt(3)}}{\alpha_3} \quad \dots\dots\dots (8)$$

Where  $\lambda$  is the Tm:YLF laser wavelength of  $(1.9 \times 10^{-6} m)$  . The 3<sup>rd</sup> harmonic non-linear extinction coefficient can be formulated as [21 ]:

$$exc_{(3)} = \frac{\alpha_3 \lambda}{4\pi} \quad \dots\dots\dots (9)$$

The coherence length  $\ell_c$  which verifies the phase –matching condition for 3rd harmonic generation to accumulate the incident, ordinary and extraordinary waves through laser media can be given as [22]:

$$\ell_c = \frac{\lambda}{6 \Delta n} \quad \dots\dots\dots (10)$$

The optical density  $d$  is the ratio between the coherence length and laser medium length as follow [23] :

$$d = \frac{\ell_c}{\ell} \quad \dots\dots\dots (11)$$

The phase shift  $\Delta\phi$  between the incident laser wave and 3<sup>rd</sup> harmonic wave can be written as follow [24-27] :

$$\Delta\phi = K \Delta n \ell \quad \dots\dots\dots (12)$$

Where  $K$  is the wave number for incident laser . The gradient of 3<sup>rd</sup> harmonic electric field  $\frac{\partial E_3}{\partial Z}$  , can be given as [28] :

$$\frac{\partial E_3}{\partial Z} = \frac{\omega_3 P_{\omega 1} \eta}{3} \quad \dots\dots\dots (13)$$

Where  $\omega_3$  is the angular frequency of generated 3<sup>rd</sup> harmonic wave ,  $P_{\omega 1}$  is the power of incident Tm:YLF laser and  $\eta$  is the 3<sup>rd</sup> harmonic efficiency which is defined as the percentage ratio between the generated 3<sup>rd</sup> harmonic power  $P_{\omega 3}$  to incident laser power  $P_{\omega 1}$  as below [29] :

$$\eta = \frac{P_{\omega 3}}{P_{\omega 1}} \quad \dots\dots\dots (14)$$

Experimental part :

*Samples Preparation :*

The  $(5 \times 10^{-4} M)$  molar concentration for each of ( Crypto cyanine , Oxazine 4 and Cresyl violet ) laser dyes in ethanol organic polar solvent , had been prepared to achieve liquid laser dye media.

*The Linear Optical Properties :*

The absorption spectrum for liquid samples of ( Crypto cyanine , Oxazine 4 and Cresyl violet ) laser dyes in ethanol, had been measured via UV-Vis spectrophotometer supplied from ( Scinco) company , type (Mega -2100) , made in ( Korea ). The linear optical properties like (transmittance , absorption coefficient  $\alpha$  and refractive index  $(n_o, n_i)$ ).

### The Third Harmonic Properties :

The THG output power was measured using power meter ( LP1 - mobiken) for different incident angles in the range from  $-20^\circ$  to  $20^\circ$  increments of  $5^\circ$  step. The studied samples are the cells of three different liquid laser dyes as (Crypto cyanine , Oxazine 4 and Cresyl violet ) in ethanol at  $(5 \times 10^{-4} \text{M})$  molar concentration , where the cell is of (1mm) thickness. The incident angle of essential beam  $\theta$  can be measured using Bevel square . This angle may be changed by move the sample forward to the left of the optical axis to make a positive angle and backward to the right of the optical axis to make a negative angle. Bevel square was used to measure the incident angle as shown in fig (1).

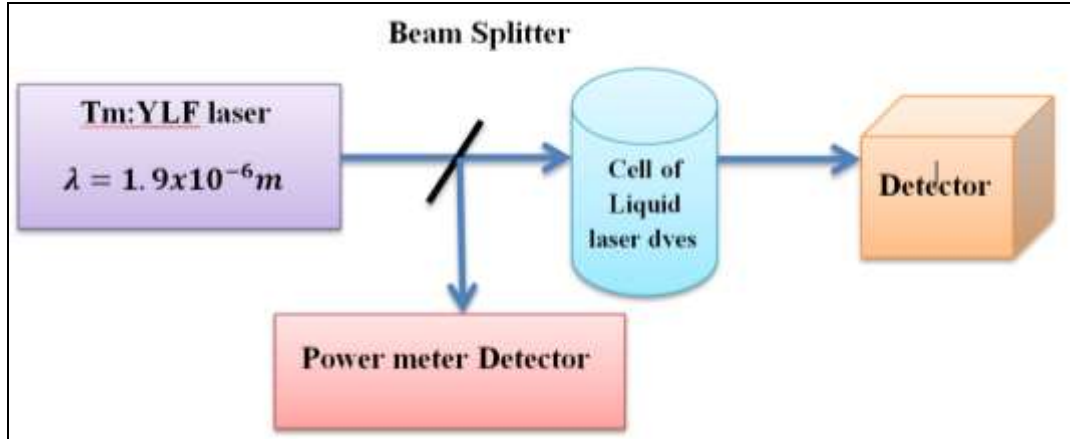


Figure:1 The Experimental Setup for 3<sup>rd</sup> harmonic generation using Tm:YLF laser and different liquid laser dyes

### Results:

#### Linear Optical Properties :

The linear optical properties such as; maximum absorption wavelength, absorbance  $A$ , transmittance  $T$ ,

absorption coefficient  $\alpha$  and refractive index  $n$  for three different liquid laser dyes as; Oxazine 4, Crypto cyanine and Cresyl violet , had been studied depending on the absorption spectra for them and using eqs.(1-3). These properties had been listed in table (1).

Table : 1

The Linear Optical Properties for Liquid laser dyes

Dyes	Maximum absorption Wavelength (nm)	Absorbance $A$	Transmittance $T$	Absorption Coefficient $\alpha \text{ (m}^{-1}\text{)}$	Refractive index $n$
Oxazine 4	615	0.91	0.402	0.477	4.765
Crypto cyanine	708	0.8	0.449	0.967	4.217
Cresyl violet	601	0.49	0.612	0.886	2.926

#### THG measuring:

The Ordinary refractive index  $n_1$ , the 3<sup>rd</sup> harmonic refractive index  $n_3$  , The refractive index difference  $\Delta n$  , the 3<sup>rd</sup> harmonic absorption coefficient  $\alpha_3$  , the 3<sup>rd</sup> harmonic electrical Conductivity  $\sigma_{elect(3)}$  , the 3<sup>rd</sup> harmonic extinction coefficient  $\epsilon_{ex3}$  and the phase shift  $\Delta\phi$  , for liquid Oxazine 4 laser dye at different Tm:YLF laser power and incident angle , had been studied using eqs.(3,4,5,6,8,9,12),respectively. These properties had been listed in table (2-a and b).While table (3-a and b) and table (4-a and b) show all the above mentioned properties at different Tm:YLF laser power and incident angle for both of Crypto cyanine and Cresyl violet laser dyes , respectively.

Table: 2

The Third Harmonic Properties for Liquid Oxazine 4 laser dyes at different: (a ) laser power at 20° incidence angle  
(b )incident angle using 80W of TmLYLF laser power

(a )

Laser Power (W )	Ordin. Refrac. index $n_1$	3 <sup>rd</sup> harmon. Refract. index $n_3$	$\Delta n$	3 <sup>rd</sup> harmonic absorption Coefficient $\alpha_3$ $\times 10^3 \text{ m}^{-1}$	3 <sup>rd</sup> harmonic electrical Conductivity $\sigma_{elect(3)}$ $\times 10^2 (\text{m}^2.\text{s}^{-1})$	3 <sup>rd</sup> harmonic Extinction Coefficient $\epsilon_{xc3}$ $\times 10^{-3}$	The Phase Shift $\Delta\phi$ $\times 10^{+6}$
7	9.898	29.694	19.796	1.609	26.949	0.243	3.271
15	8.211	24.633	16.422	1.427	22.357	0.215	2.713
30	6.513	19.539	13.026	1.203	17.733	0.181	2.152
45	4.313	12.939	8.626	0.820	11.742	0.124	1.425
50	4.012	12.036	8.024	0.693	10.918	0.108	1.326
65	3.732	11.196	7.464	0.544	10.160	0.082	1.233
80	2.763	8.289	5.526	0.446	7.523	0.0674	0.913

(b )

Incid. angle (deg.)	Ordin. Refrac. index $n_1$	3 <sup>rd</sup> harmon. Refract. index $n_3$	$\Delta n$	3 <sup>rd</sup> harmonic absorption Coefficient $\alpha_3$ $\times 10^3 \text{ m}^{-1}$	3 <sup>rd</sup> harmonic electrical Conductivity $\sigma_{elect(3)}$ $\times 10^2 (\text{m}^2.\text{s}^{-1})$	3 <sup>rd</sup> harmonic Extinction Coefficient $\epsilon_{xc3}$ $\times 10^{-3}$	The Phase Shift $\Delta\phi$ $\times 10^{+6}$
-20	8.774	26.322	17.548	8.399	23.890	1.270	2.899
-15	8.647	25.941	17.294	8.031	23.544	1.214	2.858
-10	4.638	13.914	9.276	7.794	12.628	1.179	1.532
-5	1.491	4.473	2.982	6.985	4.097	1.056	0.492
0	1.176	3.528	2.352	6.920	3.201	1.046	0.388
+5	1.595	4.785	3.19	7.013	4.342	1.060	0.527
+10	5.138	15.414	10.276	7.888	13.990	1.193	1.698
+15	6.513	19.539	13.026	8.111	17.734	1.226	2.152
+20	8.774	26.322	17.548	8.399	23.890	1.270	2.899

Table: 3

The Third Harmonic Properties for Liquid Crypto Cyanine laser dyes at different: (a ) laser power at 20° incidence angle (b )incident angle using 80W of TmLYLF laser power

(a )

Laser Power (W )	Ordin. Refrac. index $n_1$	3 <sup>rd</sup> harmon. Refract. index $n_3$	$\Delta n$	3 <sup>rd</sup> harmonic absorption Coefficient $\alpha_3$ $\times 10^3 \text{ m}^{-1}$	3 <sup>rd</sup> harmonic electrical Conductivity $\sigma_{elect(3)}$ $\times 10^2 (\text{m}^2.\text{s}^{-1})$	3 <sup>rd</sup> harmonic Extinction Coefficient $\epsilon_{xc3}$ $\times 10^{-3}$	The Phase Shift $\Delta\phi$ $\times 10^{+6}$
7	11.679	35.037	23.358	1.771	31.798	0.267	3.860
15	9.898	29.694	19.796	1.609	26.949	0.243	3.271
30	7.872	23.616	15.744	1.386	21.434	0.209	2.601
45	4.791	14.373	9.582	0.916	13.042	0.138	1.583
50	4.652	13.956	9.304	0.798	12.666	0.120	1.537
65	4.541	13.623	9.082	0.693	12.359	0.104	1.500
80	4.380	13.14	8.76	0.544	11.923	0.082	1.447

(b)

Incid. angle (deg.)	Ordin. Refrac. index $n_1$	3 <sup>rd</sup> harmon. Refract. index $n_3$	$\Delta n$	3 <sup>rd</sup> harmonic absorption Coefficient $\alpha_3$ $\times 10^3 \text{ m}^{-1}$	3 <sup>rd</sup> harmonic electrical Conductivity $\sigma_{\text{elect}(3)}$ $\times 10^2 (\text{m}^2.\text{s}^{-1})$	3 <sup>rd</sup> harmonic Extinction Coefficient $\epsilon_{\text{xc}3}$ $\times 10^{-3}$	The Phase Shift $\Delta\phi$ $\times 10^{+6}$
-20	11.020	33.06	22.04	8.622	30.006	1.304	3.642
-15	7.872	23.616	15.744	8.294	21.434	1.254	2.601
-10	6.250	18.75	12.5	8.072	17.018	1.221	2.065
-5	1.716	5.148	3.432	7.047	4.672	1.066	0.567
0	1.176	3.528	2.352	6.920	3.201	1.763	0.388
+5	1.470	4.41	2.94	6.980	4.002	1.055	0.485
+10	2.848	8.544	5.696	7.377	7.754	1.115	0.941
+15	3.732	11.196	7.464	7.377	10.161	1.115	1.233
+20	10.600	31.8	212	8.584	28.863	1.298	3.503

Table: 4

The Third Harmonic Properties for Liquid Cresyl Violet laser dyes at different: (a) laser power at 20° incidence angle (b) incident angle using 80W of TmLYLF laser power

(a)

Laser Power (W)	Ordin. Refrac. index $n_1$	3 <sup>rd</sup> harmon. Refract. index $n_3$	$\Delta n$	3 <sup>rd</sup> harmonic absorption Coefficient $\alpha_3$ $\times 10^3 \text{ m}^{-1}$	3 <sup>rd</sup> harmonic electrical Conductivity $\sigma_{\text{elect}(3)}$ $\times 10^2 (\text{m}^2.\text{s}^{-1})$	3 <sup>rd</sup> harmonic Extinction Coefficient $\epsilon_{\text{xc}3}$ $\times 10^{-3}$	The Phase Shift $\Delta\phi$ $\times 10^{+6}$
7	13.257	39.771	26.514	1.897	36.096	0.286	4.381
15	11.020	33.06	22.04	1.714	30.005	0.259	3.642
30	8.979	26.937	17.958	1.514	24.449	0.229	2.967
45	7.560	22.68	15.12	1.347	20.582	0.203	2.498
50	4.925	14.775	9.85	0.941	13.407	0.142	1.627
65	4.104	12.312	8.208	0.776	11.174	0.117	1.356
80	3.410	10.23	6.82	0.616	9.284	0.093	1.127

(b)

Incid. angle (deg.)	Ordin. Refrac. index $n_1$	3 <sup>rd</sup> harmon. Refract. index $n_3$	$\Delta n$	3 <sup>rd</sup> harmonic absorption Coefficient $\alpha_3$ $\times 10^3 \text{ m}^{-1}$	3 <sup>rd</sup> harmonic electrical Conductivity $\sigma_{\text{elect}(3)}$ $\times 10^2 (\text{m}^2.\text{s}^{-1})$	3 <sup>rd</sup> harmonic Extinction Coefficient $\epsilon_{\text{xc}3}$ $\times 10^{-3}$	The Phase Shift $\Delta\phi$ $\times 10^{+6}$
-20	15.937	47.811	31.874	8.987	43.394	1.359	5.267
-15	9.898	29.694	19.796	8.517	26.951	1.288	3.271
-10	7.872	23.616	15.744	8.294	21.434	1.254	2.601
-5	2.448	7.344	4.896	7.264	6.665	1.098	0.809
0	1.176	3.528	2.352	6.920	3.201	1.046	0.388
+5	3.000	9.00	6.00	7.418	8.168	1.122	0.991
+10	8.979	26.937	17.958	8.421	24.448	1.273	2.967
+15	11.340	34.02	22.68	8.650	30.877	1.308	3.748
+20	15.937	47.811	31.874	8.987	43.394	1.359	5.267

Fig.(2-a and b) show the third harmonic efficiency  $\eta$  for three different liquid laser dyes as; Oxazine 4, Crypto cyanine and Cresyl violet at different Tm:YLF laser power and incident angle, respectively . The third harmonic efficiency  $\eta$  had been calculated using eq.(14)

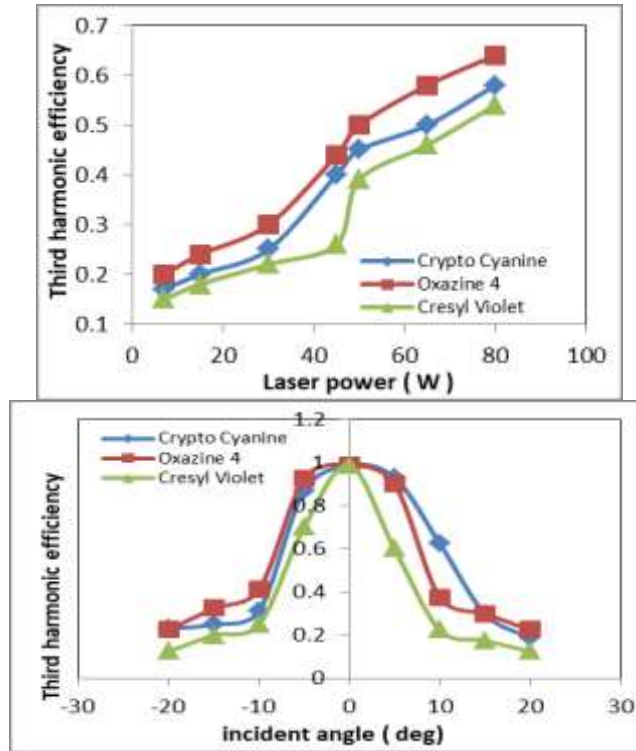


Figure: 2

The third harmonic efficiency  $\eta$  for three different liquid laser dyes as; Oxazine 4, Crypto cyanine and Cresyl violet at different: (a ) Tm:YLF laser power at 20° incidence angle (b) Incident angle using 80 W Tm:YLF laser

Fig.(3-a and b) show the optical density  $d$  for three different liquid laser dyes as; Oxazine 4, Crypto cyanine and Cresyl violet at different Tm:YLF laser power and incident angle, respectively . The optical density  $d$  had been calculated using eq.(11)

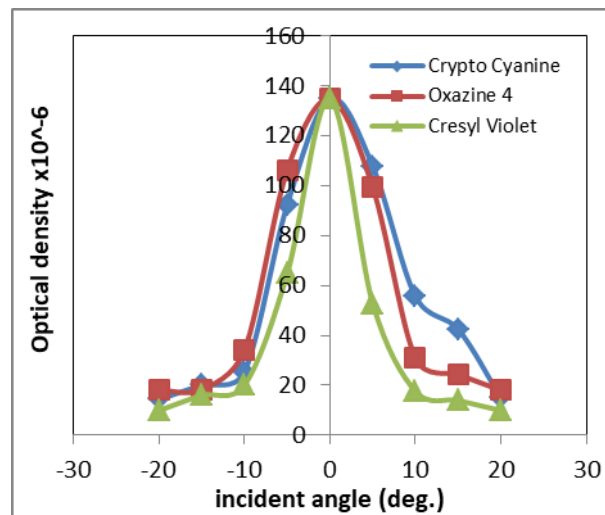


Figure: 3

The optical density  $d$  for three different liquid laser dyes as; Oxazine 4, Crypto cyanine and Cresyl violet at different:



- (a) Tm:YLF laser power at 20° incidence angle  
(b) Incident angle using 80 W Tm:YLF laser

Fig.(4-a and b) show the optical conductivity  $\sigma_{opt(3)}$  for three different liquid laser dyes as; Oxazine 4, Crypto cyanine and Cresyl violet at different Tm:YLF laser power and incident angle, respectively . The optical conductivity  $\sigma_{opt(3)}$  had been calculated using eq.(7)

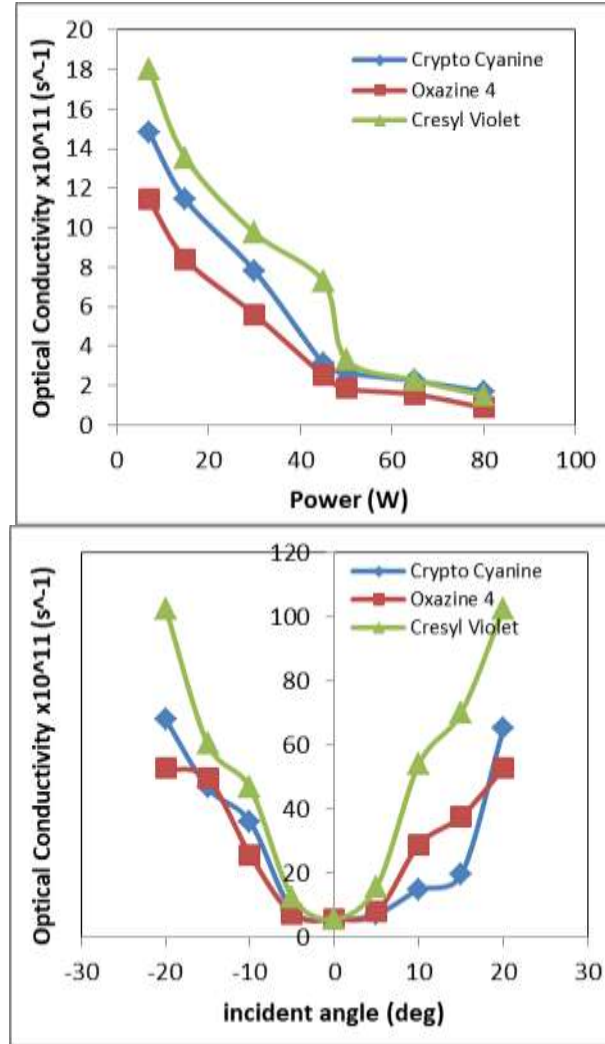


Figure: 4

The optical conductivity  $\sigma_{opt(3)}$  for three different liquid laser dyes as; Oxazine 4, Crypto cyanine and Cresyl violet at different :

- (a) Tm:YLF laser power at 20° incidence angle  
(b) Incident angle using 80 W Tm:YLF laser

Fig.(5-a and b) show the coherence length  $\ell_c$  for three different liquid laser dyes as; Oxazine 4, Crypto cyanine and Cresyl violet at different Tm:YLF laser power and incident angle, respectively . The coherence length  $\ell_c$  had been calculated using eq.(10)



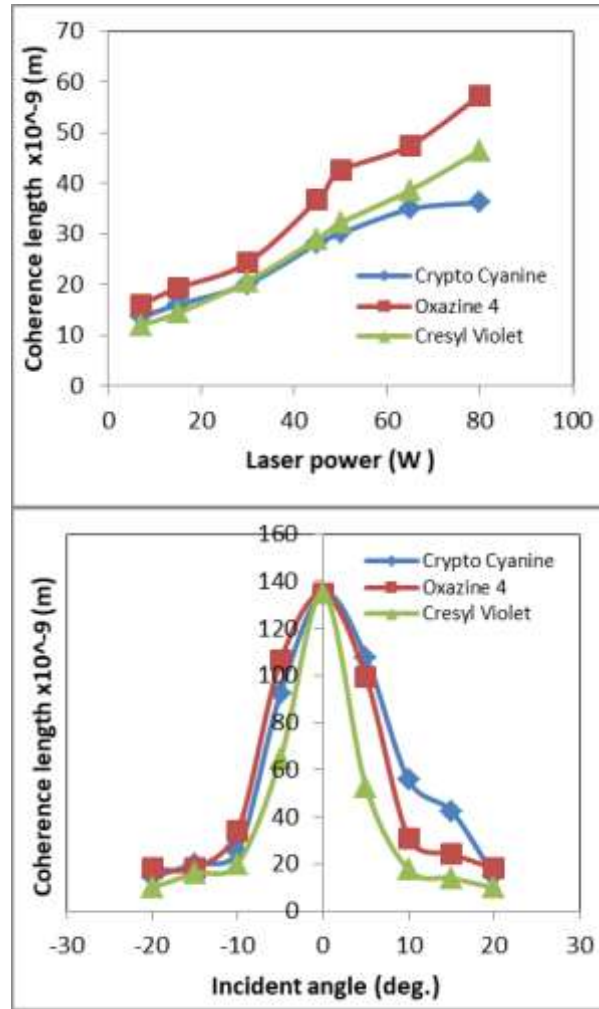


Figure: 5

The coherence length  $\ell_c$  for three different liquid laser dyes as; Oxazine 4, Crypto cyanine and Cresyl violet at different:

- (a ) Tm:YLF laser power at  $20^\circ$  incidence angle
- (b) Incident angle using 80 W Tm:YLF laser

Fig.(6-a and b) show the gradient of 3<sup>rd</sup> harmonic electric field  $\frac{\partial E_3}{\partial z}$  for three different liquid laser dyes as; Oxazine 4, Crypto cyanine and Cresyl violet at different Tm:YLF laser power and incident angle, respectively . The gradient of 3<sup>rd</sup> harmonic electric field  $\frac{\partial E_3}{\partial z}$  had been calculated using eq.(13)

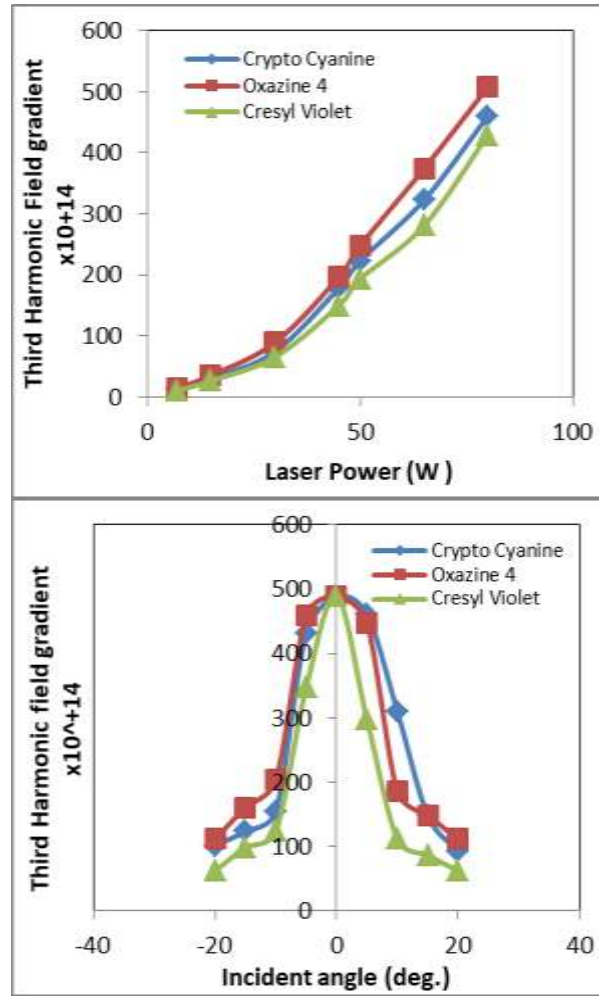


Figure: 6

The gradient of 3<sup>rd</sup> harmonic electric field  $\frac{\partial E_3}{\partial z}$  for three different liquid laser dyes as; Oxazine 4, Crypto cyanine and Cresyl violet at different:

(a) Tm:YLF laser power at 20° incidence angle

(b) Incident angle using 80 W Tm:YLF laser power

#### Discussion

Fig.(2-a and b) show the 3<sup>rd</sup> harmonic efficiency as a function of laser power and incident angle, respectively. The increasing of incident laser power on the laser dye solution causes an obvious linearly increasing in 3<sup>rd</sup> harmonic efficiency ( $\eta$ ). While the increasing of incident angle leads to form a curve behaviour for 3<sup>rd</sup> harmonic efficiency ( $\eta$ ). Since  $\eta$  has the maximum value at (0 degree) and starts to decrease with increasing of incident angle to positive part or to negative part. The Oxazine (4) laser dye has highest efficiency, while (Crypto cyanine) is the lower than one and the less efficiency had been appeared for Cresyl violet laser dye.

This behaviour can be attributed to the highest values for each of the optical density ( $d$ ), coherence length ( $\ell_c$ ) and the 3<sup>rd</sup> harmonic electric field gradient  $\frac{\partial E_3}{\partial z}$ , had been obtained at highest values of incident laser power, as shown in figs.(3,5-6)-a, respectively.

The closed curve behaviour for  $\eta$  as a function of incident angle in fig.(2-b) must be attributed to that the whole laser power allowed to convert to 3<sup>rd</sup> harmonic wave at (0 degree), while when the incident angle increases, the allowed laser power to 3<sup>rd</sup> conversion becomes less.

The same behavior mentioned above, are appeared to optical density ( $d$ ), coherence length ( $\ell_c$ ) and the 3<sup>rd</sup> harmonic electric field gradient  $\frac{\partial E_3}{\partial z}$ , at different values of incident angle as illustrated in figs.(3,5-6)-b, respectively. Fig.(4-a and b) show the 3<sup>rd</sup> harmonic optical conductivity ( $\sigma_{opt(3)}$ ) as a function of laser power and incident angle, respectively. The increasing of incident laser leads to ( $\sigma_{opt(3)}$ ) decreasing, while it has the open curve behavior when it had been studied at different values of incident angle. Where ( $\sigma_{opt(3)}$ ) has the minimum value at (0 degree) incident angle and it starts to increase with increasing of incident angle in positive part and in negative part, because of the whole laser power are allowed to convert to 3<sup>rd</sup> wave in (0 degree) incident angle and starts to decrease with the increasing of incident angle in both parts (positive and negative).

Table (1) shows that the (Oxazine 4) laser dye has the highest values for both of Absorbance (A) and refractive index (n) than for Crypto Cyanine and Cresyl Violet; while it has the lowest values for both of Transmittance (T) and absorption Coefficient ( $\alpha$ ) than others.

The data in tables (2- 4)-a illustrate that the increasing of incident laser power on the liquid laser dyes, causes an obvious decreasing for each of ( Ordinary index  $n_1$ , 3<sup>rd</sup> harmonic refractive index  $n_2$ , the difference  $\Delta n$  between  $n_1$  and  $n_2$ , 3<sup>rd</sup> harmonic absorption coefficient  $\alpha_3$ , 3<sup>rd</sup> harmonic electrical conductivity  $\sigma_{elect(3)}$ , 3rd harmonic extinction coefficient  $exc_2$  and the phase shift  $\Delta\phi$ ). This is caused by that the large applied laser power on the liquid laser dyes strongly changes the induced electrical field within laser dye which consolidates the refractive indices and all other 3<sup>rd</sup> properties. It is clear from above tables that the Cresyl Violet laser dyes has the strongest 3<sup>rd</sup> harmonic nonlinear properties at different values of incident laser power; while the Crypto Cyanine laser dye has less 3<sup>rd</sup> properties than one, and Oxazine 4 laser dye has the lowest 3<sup>rd</sup> harmonic nonlinear properties than other studied laser dyes. This behaviour can be attributed to that the Cresyl Violet has lowest absorbance than each of Oxazine 4 and Crypto Cyanine laser dye.

The data in tables (2- 4)-b show that the laser dye give highest 3<sup>rd</sup> harmonic properties when the incidence angle far from (0o) which has the lowest one in it. This due to that the incidence laser beam with any angle except (0o) induces more electrical conductivity  $\sigma_{elect(3)}$  and 3rd harmonic refractive indices and other properties. The Cresyl Violet laser dye give highest 3rd harmonic properties than Crypto Cyanine at different values of laser incident angle which has less 3rd nonlinear properties than one. The Oxazine 4 laser dye has the lowest values of 3rd harmonic properties.

### Conclusions

It can be concluded that the Oxazine 4 laser dye is the best studied laser dye in 3rd harmonic generation efficiency; while the Cresyl Violet laser dye gives highest 3rd harmonic properties than other studied laser dyes. The 0° of laser incidence angle causes highest 3<sup>rd</sup> harmonic generation efficiency and lowest 3<sup>rd</sup> nonlinear properties for each studied laser dyes. The main conclusion is that the incident laser power on the liquid laser dyes has the strongest effects in all the 3<sup>rd</sup> harmonic properties, but the incidence angle of laser beam, has the highest effect in the 3<sup>rd</sup> harmonic generation efficiency

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