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Comparative Study of the Linear and Nonlinear Optical Properties for Different Iraqi Heavy and Light Crude Oils

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Abstract

Iraqi crude oil has some of the physical and chemical characteristics that distinguish it from other types of oil crudes in the world. Some of these features such us molecular composition, rheological, viscosity and emulsions are studied carefully by researchers. In this work, a comparative study of the linear and the non-linear optical properties for typical heavy and light Iraqi crude oil samples was studied utilizing the Z-scan technique. The He -Ne laser of wavelength 632.8 nm had been used for this purpose. These samples were collected from Basra and Kut oil fields. The values of the non-linear refractive index (n2), non-linear absorption coefficient (β), and third-order electrical susceptibility (χ3) were estimated respectively. All the samples have a negative refractive index with high nonlinearity. The obtained results will be helpful for researchers who work in the field of oil industries.

Key words

Iraqi crude oil, nonlinear optical properties, z-scan technique, petroleum industry.

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دراسة مقارنة للخصائص البصرية الخطية واللاخطية لعينات مختلفة من النفط العراقي الثقيل والخفيف

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الخلاصة

يتمتع النفط الخام العراقي ببعض الخصائص الفيزيائية والكيميائية التي تميزه عن بقية أنواع النفط الخام الأخرى في العالم. بعض هذه الميزات مثل التركيب الجزيئي، الريولوجيا، اللزوجة والمستحلبات، والتي تمت دراستها بعناية من قبل الباحثين. تم في هذا العمل سيتم تقديم دراسة مقارنة للخصائص البصرية الخطية وغير الخطية لعينات النفط الخام العراقي (الثقيلة والخفيفة) باستخدام تقنية المسح Z. وقد استخدم ليزر He -Ne بطول موجة 632.8 نانومتر لهذا الغرض. تم جمع هذه العينات من حقلي نفط البصرة والكوت. تم تقدير قيم معامل الانكسار غير الخطي (n_2) ومعامل الامتصاص غير الخطي (β) والحساسية الكهربائية من الدرجة الثالثة (χ3) على التوالي. جميع العينات لها معامل انكسار سالب مع لاخطية عالية. النتائج التي تم الحصول عليها ستكون مفيدة للباحثين الذين يعملون في مجال الصناعات النفطية.

Introduction

Crude oil is a liquid fuel source located underground, it is extracted through drilling, and it is a base part of vehicle fuel, plastics, chemicals, and other petroleum products. It represents a complex mixture of hydrocarbon and non-hydrocarbon compounds, and different chemical elements such as oxygen, nitrogen, sulfur, and a small amount of nickel or vanadium [1]. The crude oil can be categorized as either light or heavy according to the American Petroleum Institute (API^0) gravity [2]. Based on the API^0 index, the Brazilian National Petroleum Agency (ANP) classifies four different types of oil, as shown in Table 1 [3].

Table 1: Crude oil ordering by the National Petroleum Agency of Brazil (Adapted from ANP, 2000) [3].

Oil Class	API˙
Light	API˙ ≥ 31
Medium	22≤ API˙<31
Heavy	10≤ API˙<22
Extra-heavy	API˙≤10

The Light Crude oils are fluid oil that has a low density and that flow freely at room temperature. It has low viscosity, low specific gravity and high API^0 gravity due to the existence of a high amount of light hydrocarbon elements. While the heavy crude oil has a high viscosity and density, chemical complexity, high acidity, high sulfur, high level of metals, increase asphaltenes content, low API^0 gravity [3]. Latest training is examined various of oil properties, such as the rheological properties of heavy crude oil [4, 5], the performance of the emulsion [6], a viscosity [7, 8], the steady condition [9] and nonlinear properties of multi-grade oil [10]. In this work, we set out to compare the linear and the nonlinear optical properties of different heavy and light Iraqi crude oils extracted from Basra and Kut oil fields. This could be clarified by utilizing the Z-scan technique. It is the easiest and profits way by which signal and value of a nonlinear refractive index (n_2), nonlinear absorption (β) and third-order electrical susceptibility (χ^3) can be achievement independently [11, 12].

Experimental work

In this work, three types of Iraqi crude oil samples had been used. They were titled as: (B) for Basra heavy crude oil, (K1) and (K2) for Kut heavy and light crude oil respectively. About 2 ml of each sample was filled into a glass cell of 2mm thickness. The Z-Scan technique had been used to study the linear and the nonlinear optical properties of the oil samples. The He-Ne laser with wavelength 632.8 nm and power 5m watt had been used for this purpose. Fig.1 shows the Z-Scan technique experimental setup. The laser beam is focused with a convergent lens, f = 15 cm focal length. The beam waist diameter at the focus area w_0= 2.4183×10^{-2} mm and Rayleigh length of z_0 = 29 mm, the hole transmittance (S) is 2.188638×10^{-9}. The laser intensity (I_0) measured by a (1 cm^2) large area photodetector to be 67634.71 mW cm^{-2} and the radius of the beam (w_a) is 15114.6496 nm. The effective length (L_{eff}) is 1.438, 1.5039612, and 1.43353 for the B, K1, and K2 crude oil samples respectively. The closed and open apertures have prepared with an aperture of diameter about 1mm to investigate the (n_2), (β) and (χ^3) individually. A stepper motor-powered of 10 µm steep was integrated into a linear movement set in order to ensure the sample displacement with a 0.05 mm for each step.

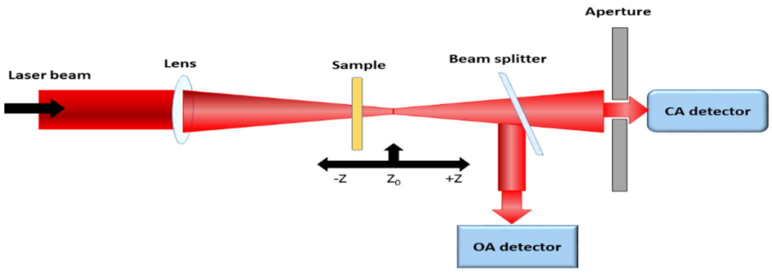

Fig.1: Experimental setup for the Z-Scan technique.

The absorption coefficient (α_0) of each sample is given by [13]:

$$\boldsymbol{\alpha_0 = \frac{1}{d}\ln\frac{1}{T}} \qquad \textbf{(1)}$$

Where d is the sample thickness, and T is the sample transmittance. The extinction coefficient (k) of the sample is:

$$\boldsymbol{k = \frac{\lambda\alpha_0}{4\pi}} \qquad (2)$$

The linear reflection coefficient (R):

$$\boldsymbol{R = \frac{1+\sqrt{1-exp(-\alpha d)+exp(\alpha d)}}{1+exp(-\alpha d)}} \qquad \textbf{(3)}$$

The linear refraction coefficient (n):

$$\boldsymbol{n = \frac{-(R+1)+\sqrt{-3R^2+10R-3}}{2(R-1)}} \qquad \textbf{(4)}$$

The magnitude and sign of the (n_2) in the closed aperture (CA) Z-Scan manner. The phase shift ($\Delta\emptyset_\circ$) of nonlinear media is given by [14]:

$$\boldsymbol{\Delta T_{P-V} = 0.406(1-S)^{0.25}|\Delta\emptyset_\circ|} \qquad \textbf{(5)}$$

where ΔTp-v is the difference between peak and valley of output power, and S is the hole transmittance provided by [15,16]:

$$\boldsymbol{S = 1-\exp\left(-\frac{2r_a^2}{w_a^2}\right)} \qquad \textbf{(6)}$$

where r_a is the radius of the aperture, w_a is the radius of the beam at the aperture. The sample is placed at the focus point of the lens, and then moved along the z-axis a distance of $\mp z_\circ$ which is given by the Rayleigh length $z_\circ$:

$$\boldsymbol{z_\circ = \frac{\pi w_\circ^2}{\lambda}} \qquad \textbf{(7)}$$

where w_o is the beam waist diameter, the nonlinear refractive index n_2 is given by **[**17]:

$$\boldsymbol{n_2 = \frac{\Delta\varphi_\circ}{kI_\circ L_{eff}}} \qquad \textbf{(8)}$$

where k is the wave number, λ is the wavelength of the laser beam, I_o is the laser intensity at focus. The L_{eff} is the effective length of the sample, and given by:

$$\boldsymbol{L_{eff} = \big(1-exp(-\alpha_\circ d)\big)/\alpha_\circ} \qquad \textbf{(9)}$$

In OA (Open Aperture) state, the transmitted light is measured when the aperture is removed, in this condition, the measurement gives information about the β which is given by: [18].

$$\beta = \frac{2\sqrt{2}\,\Delta T}{I_\circ L_{eff}} \qquad (10)$$

where ΔT is the peak value at open aperture (OA) of the Z-Scan curve. The third-order susceptibility, $\chi^{(3)}$, is determined by means of the following relation: [19, 20]

$$\left|\chi^{(3)}\right| = \sqrt{(Re\{\chi^{(3)}\})^2 + (lm\{\chi^{(3)}\})^2} \qquad (11)$$

where $\left(Re\{\chi^{(3)}\}\right)^2$ and $\left(lm\{\chi^{(3)}\}\right)$ are the real and imaginary part of third-order susceptibility, respectively, and are given by:

$$Re\{\chi^{(3)}\}(esu) = 10^{-4}\,\frac{\epsilon_0 c^2 n_0^2}{\pi}\,n_2\left(cm^2/W\right) \qquad (12)$$

and

$$lm\{\chi^{(3)}\}(esu) = 10^{-2}\,\frac{\epsilon_0 c^2 n_0^2}{4\pi^2}\,\beta\left(cm^2/W\right) \qquad (13)$$

where ϵ_0 is the vacuum electric permittivity (8.8542×10^{-16} C/Nm^2) and c (3×10^{10}cm/s) is the speed of light.

Results and discussion

The achieved consequences of the linear properties of the three crude oil samples are shown in Table 2.

Table 2: The linear optical parameters for the three crude oil samples.

Oil sample	Linear absorption coefficient $\alpha_\circ$ (mm)$^{-1}$	Reflectance (R)	Thickness of model d (mm)	Extinction coefficient k	Linear refraction coefficient n	Wavelength λ (nm)
B	0.35	2.351	2	17.624	1.0138	632.8
K1	0.3	2.114	2	15.107	1.12328	632.8
K2	0.211	5.4	2	13.085	0.60997	632.8

From the above results, it can be noticed that the extinction coefficient magnitude (k) of heavy oil samples (B and K1) were higher than the light one (K2). The reason behind this is that the proportions of the elements (the chemical composition) of heavy elements are greater than those in light elements such as asphalt and others. And the linear refractive index and linear absorption coefficient of the heavy oil samples are close and are greater than the linear refractive index of the light sample for the same reasons mentioned above. The reflectivity was smaller for the heavy oil samples as compared with the light oil sample. The density of the crude oil correlates toughly with the refractive index, and thus, it can be used as a precise way to approximate the API gravity. Based on that, the crude oil refractive index is a suitable factor in illustrating the crude oil properties.

1. The nonlinear refractive index coefficient (n_2) measurements

The nonlinear refractive index (n_2) was measured through the closed aperture experiment with aids of Eq.(8). The nonlinear behavior of each sample illustrated in Fig.2.

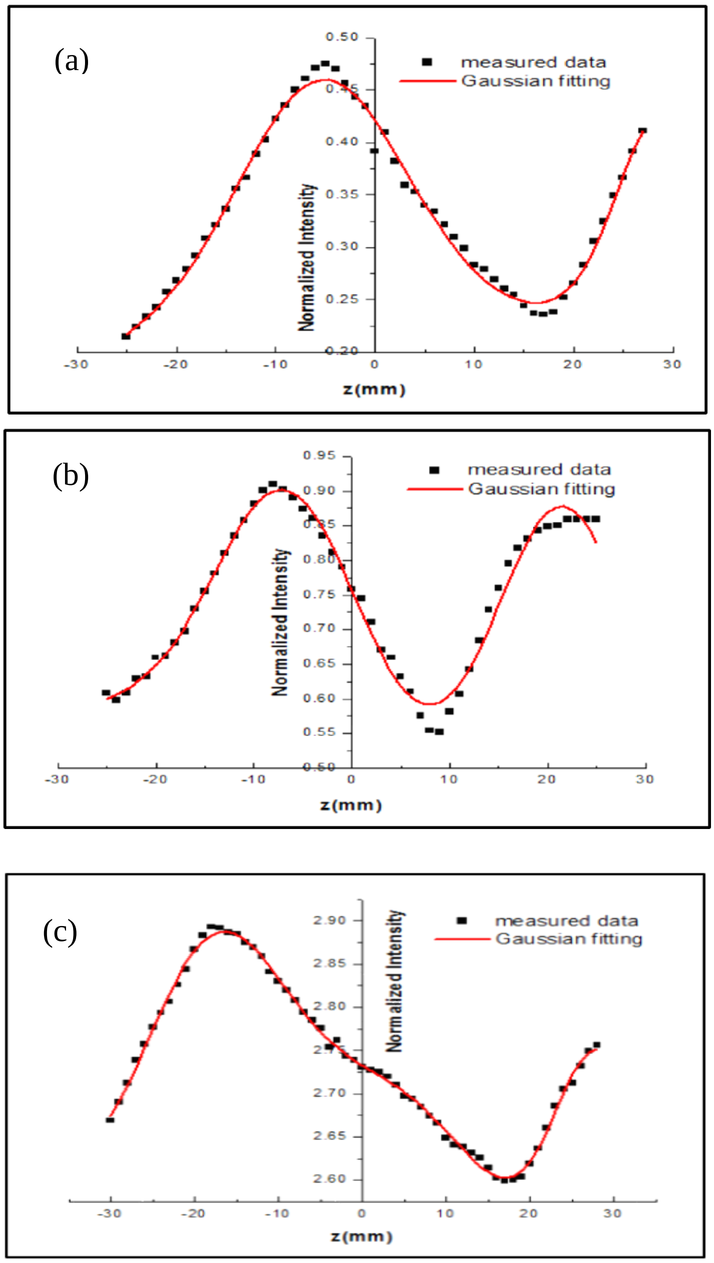

Fig.2: The Z-scan curves for CA of the three crude oil samples measured at 632.8 nm: (a) B sample, (b) K1 sample, and (c) K2 sample.

Fig.2 illustrates the behavior of the three crude oil samples when each sample is scanned along the Z-axes. A negative Z-scan profile for each sample is starting at ($Z < 0$), the intensity increases as the sample is motivated towards the focus producing a higher transmittance through the aperture due to self-lensing in the sample. The high transmittance value will drop to a minimum value as the sample is moved at ($Z > 0$)

because the sample shows a negative lensing to the beam. Table 3 listed the values of n_2 for these samples.

Table 3: The experimental results for the test samples in CA

material	ΔT_{p-v}	$\Delta\phi_\circ$	$n_2(\frac{cm2}{mW})$
B	0.238	0.615897	-6.34381×10^{-4}
K1	0.358	0.837620	-8.25108×10^{-4}
K2	0.29	0.714440	-7.35882×10^{-4}

From Table 3, the magnitude of phase shift $\Delta\phi_\circ$ is very small (less than π) for all samples denotes that the sample shows a light thermal lens effect.

2. The nonlinear absorption coefficient (β) Measurements

In the open aperture experiment, when all the samples are scan nearer to the focus, the obtained curves begin to drop until they reach to the minimum value (T_{min}) at the focal point, (Z=0). Later, the curves instigate to increase with the samples place (+Z) as shown in Fig.3.

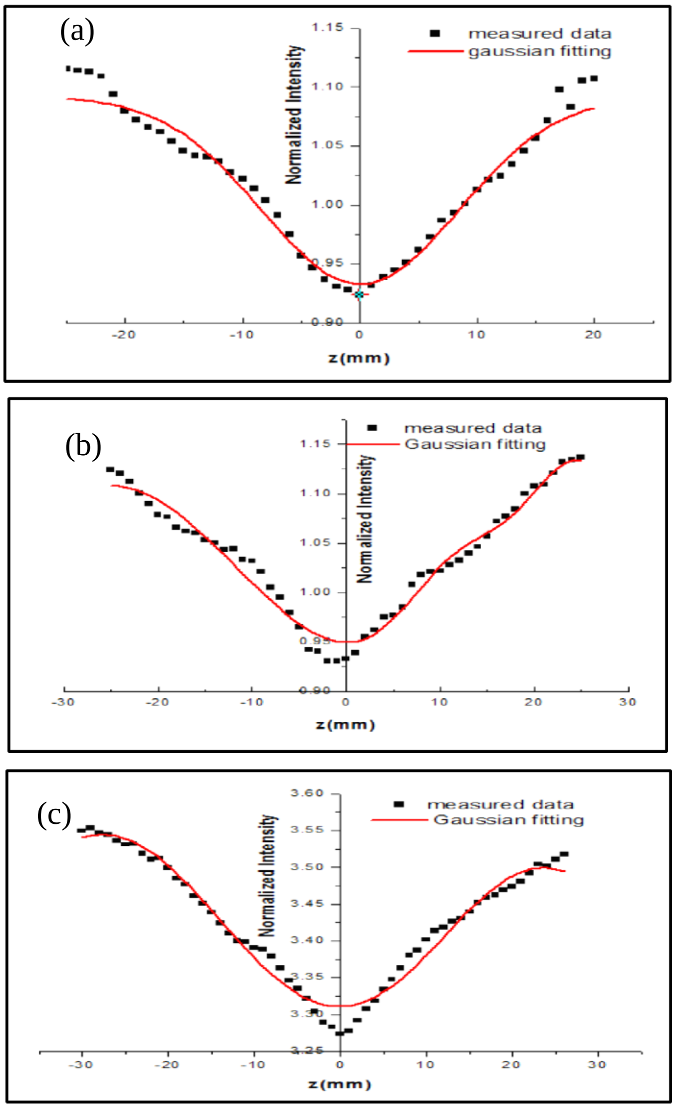

Fig.3: The far-field OA Z-scan curves of the three crude samples illuminated by He-Ne laser pulses at 632.8nm. (a) B sample, (b) K1 sample, and (c) K2 sample.

From Fig.3, the performance of transmittance showing linear behavior when the sample is positioned at (-Z). As the sample is shifted nearer to the right, the curve of transmittance is decreases to the minimum value (T_{min}) at the (Z=0) axes. Later, the transmittance curve tends to increase at the sample position (+Z). The obtained figure for each sample in open aperture Z- scan exhibits a symmetric around the focus, the nonlinear absorption coefficient (β) can be calculated using equation 10. The values of β of the three samples are listed in Table 4.

Table 4: The experimental results for the three samples in OA.

Oil sample	T_{min}	$\beta(\frac{cm}{mW})$
B	0.924	2.68942×10^{-5}
K1	0.938	2.58874×10^{-5}
K2	3.72	1.555673×10^{-5}

The values of β indications high absorption for the heavy oil (B and K1) samples as the comparison with light oil (K2) sample, this may be due to the high optical density of the heavy sample. To take out the effect of the absorption, we used the CA Z-scan curve divided by the OA Z-scan curve as shown in Fig.4.

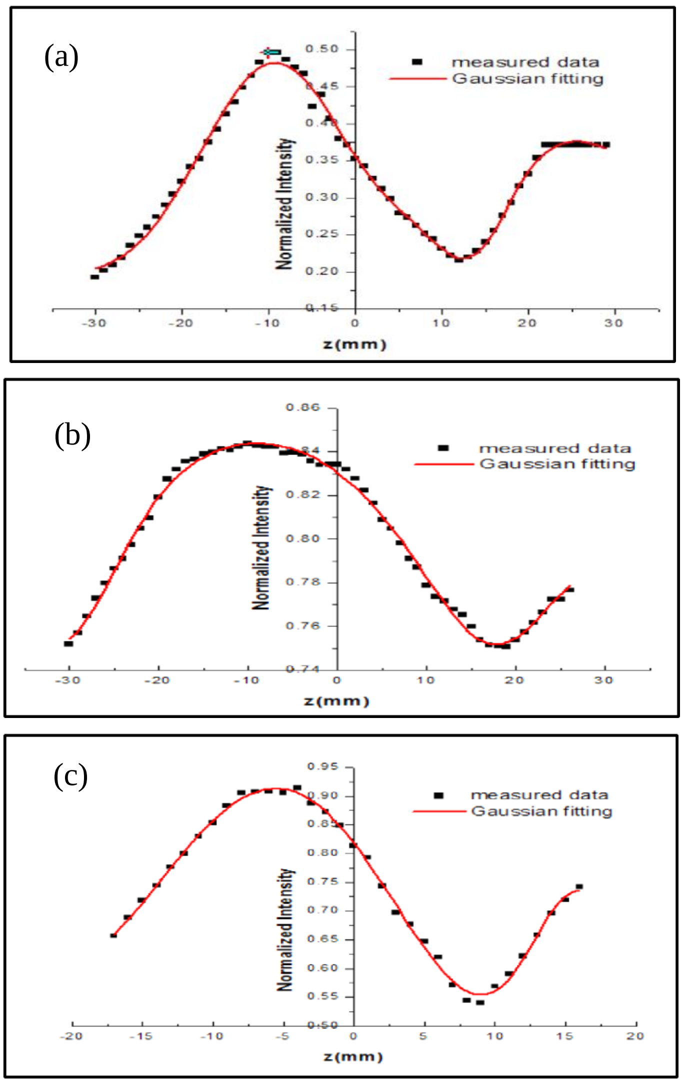

Fig.4: The division of the CA by OA Z-scan curves for the oil samples at 632.8 nm for, (a) B sample, (b) K1 Sample, (c) K2 sample respectively.

From the CA/OA Z-scan curves of the test samples at the high-intensity beam incident, we can observe the negative nonlinear refractive index (n_2). Which indicate that all the samples behaviors were self-defocusing (n_2) at 632.8 nm. The values of (n_2), and ($\chi^{(3)}$) are shown in Table 5.

Table 5: The experimental results for the test samples in CA/CO

Oil samples	ΔTp − v	$\Delta\emptyset_\circ$	$n_2(\frac{cm2}{mW})$	$\chi^{(3)}$(esu)
B	0.282	0.68980	-7.105078×10^{-4}	165.562×10^{-15}
K1	0.34	0.96079	-9.466447×10^{-4}	264.158×10^{-15}
K2	0.092	0.22665	-2.334522×10^{-4}	69.459×10^{-15}

from this table, the magnitude of the (χ^3) for the heavy crude oil samples was found to be higher than for the light oil samples, this could be due to the higher percentage of hydrocarbons. This also represents thermal effects for these samples since this is indirectly related to n_2. Heavy crude has high concentrations of sulfur and several metals, mainly nickel and vanadium. The present results are compared with another previous study for some crude oil samples presented by [21, 10] and listed in the Table 6.

Table 6: Comparison of the obtained linear and nonlinear optical properties of Iraqi crude oil samples with values of different oil samples at different wavelengths of light sources.

Oil samples	$\alpha_\circ$ (mm)$^{-1}$	Linear refraction coefficient (n)	$n_2(\frac{cm2}{mW})$	$\beta(\frac{cm}{mW})$	$\chi^{(3)}$(esu)	Source and Wavelength (nm)
Azadegan oil	0.459	-1.4237×10^{-4}	-2.2126×10^{-8}	—	—	Diode laser (660) nm.
Sorouh oil	0.505	-1.1014×10^{-4}	-1.7116×10^{-8}	—	—	Diode laser (660) nm.
20W-50 multi-grade oil	—	—	-2.3201×10^{-9}	1.3864×10^{-3}	3.1286×10^{-5}	Nd:YAG laser (532) nm
B	0.35	1.0138	-7.1050×10^{-4}	2.6894×10^{-5}	165.56×10^{-15}	He-Ne laser (632.8)nm
K1	0.3	1.12328	-9.4664×10^{-4}	2.5887×10^{-5}	264.15×10^{-15}	He-Ne laser (632.8)nm
K2	0.211	0.60997	-2.3345×10^{-4}	1.5556×10^{-5}	69.45×10^{-15}	He-Ne laser (632.8)nm

From this table, the three samples are showed positive linear refractive index (n), which displays the possibility of the super lens and other new phenomena to be actively developed by means of metamaterials. The two heavy crude oil samples (B and K1) show high nonlinear absorption (β) as comparison to the light crude oil sample (K2). This may due to Iraqi crude oil contains high salt content and it contain high concentrations of some metals with API^0 value of 24^0 according to the Exxon Mobil index table. It described by a high content of asphalt, weight of hydrocarbons.

Conclusions

The non-linear optical properties of Iraqi crude oil samples (heavy and light) were determined for laser wavelength of 632.8 nm. The non-linear absorption coefficient was obtained from the transmittance curves in open-cell configuration, for all samples we finding that the non-linear absorption effect is associated to saturable absorption. The non-linear refractive index was obtained from the transmittance curves for the closed-cell configuration, the sign of this parameter was negative for all tested samples beside they have high nonlinearity. In addition, the magnitude of the third-order electric susceptibility was calculated for each sample and it is observed that the order of magnitude of this value is considered very small. These results distinguish Iraqi oil from other types of oil in the world in terms of optical characteristics and allow us to observe that the heavy and light Iraqi crude oils offer high non-linear sensitivity.

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