Integrated Linear and Nonlinear Optical Characterization of Olive Oil by 532 nm CW Laser Beam

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Abstract:

We present a comprehensive linear and nonlinear optical characterization of virgin and refined olive oil samples using a 532 nm continuous-wave (CW laser). The nonlinear optical properties were evaluated using the Z-scan technique to determine the nonlinear absorption and refraction coefficients. Additionally, the linear absorption characteristics were evaluated by UV-visible spectroscopy, giving the absorption edge values for both virgin and refined olive oil samples. The UV-visible absorption spectroscopy also indicated the type of nonlinear absorption, which is, in this case, two-photon absorption for both samples. This integrated linear and nonlinear approach can be effectively employed to assess the quality of the olive oil. Beam profiling was performed using the knife-edge technique, enabling accurate measurements of beam waist at the focus.

Introduction

The Z-scan technique, introduced by Sheik-Bahae et al. in 1989, has become a standard method for characterizing third-order nonlinear optical properties of material [1]. This single-beam technique enables the measurement of both nonlinear refraction (NLR) and nonlinear absorption (NLA) by translating a sample through the focus of a Gaussian laser beam and recording the transmitted intensity. In the closed-aperture Z-scan, an aperture is placed before the detector to detect beam distortions caused by NLR. In contrast, in the open-aperture Z-scan, the aperture is removed to exclusively measure NLA [1–3].Compared to other techniques such as degenerate four-wave mixing and third-harmonic generation, Z-scan offers simplicity and sensitivity, and requires only a single laser beam with minimal alignment [4,5].

The Z-scan technique has been extensively applied to a wide range of materials, including semiconductors, nanocrystals, semiconductor-doped glasses, liquid crystals, organic compounds, and biomaterials [6–8]. Both continuous-wave (CW) and pulsed lasers are employed in Z-scan experiments, with the choice of laser depending on the material's response and the specific nonlinearities under investigation. CW lasers are typically used for studying thermal nonlinearities, whereas pulsed lasers —such as femtosecond or picosecond sources—are better suited for investigating ultrafast electronic nonlinearities [9–11]. The origin of optical nonlinearity in materials is strongly influenced by the laser's characteristics, particularly intensity and pulse duration. With ultrashort pulses (femtoseconds to picoseconds), high peak

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intensities can induce nonlinear processes like multi-photon absorption and Kerr effect, leading to instantaneous electronic nonlinearities [12–14]. In contrast, longer pulses or CW lasers can cause thermal effects due to cumulative heating, leading to thermal lensing or refractive index change [15,16]. Therefore, the observed nonlinearity is a complex interplay between the material's intrinsic properties and the laser parameters, necessitating careful selection of experimental conditions to isolate specific nonlinear effects.

The nonlinear optical properties of olive oil can be effectively studied using the Z-scan technique, providing insights into its nonlinear refraction and absorption characteristics. Due to its complex molecular composition, olive oil exhibits significant third-order optical nonlinearity, making it a promising medium for optical applications [17]. Studies using continuous-wave (CW) lasers, such as a 532 nm CW Nd:YAG laser, have revealed a nonlinear refractive index (n_2) of approximately 3.99×10^{-6} cm^2/W and a nonlinear absorption coefficient (β) of -0.0017 m/W, indicating strong self-defocusing behavior. Olive oil has also demonstrated significant third- and fifth-order nonlinear refractive indices, with a negative third-order index around 10^{-7} cm^2/W and a positive fifth-order index around 10^{-9} cm^4/W^2, making it a promising natural nonlinear medium [18]. These findings underscore the potential of olive oil as a natural nonlinear optical medium for applications in biophotonics, optical limiting, and optical switching.

In this study, we performed a comprehensive optical characterization of an olive oil sample using a 532 nm continuous-wave (CW) laser. The nonlinear optical properties were evaluated through the Z-scan technique, allowing the determination of both nonlinear absorption and refraction coefficients. Beam profiling was carried out using the knife-edge method, providing precise measurements of beam waist and divergence. Additionally, linear absorption characteristics were assessed via UV-Visible spectroscopy to obtain the absorption spectrum of the sample. This integrated approach offers a detailed understanding of the sample's linear and nonlinear optical behaviors, which is essential for potential applications in photonic devices.

Experimental methods.

The schematic of the experimental setup is shown in figure (1). The measurements of both closed and open aperture are carried out within the same experiment. Here, the output laser beam has a diameter of 2 mm. To

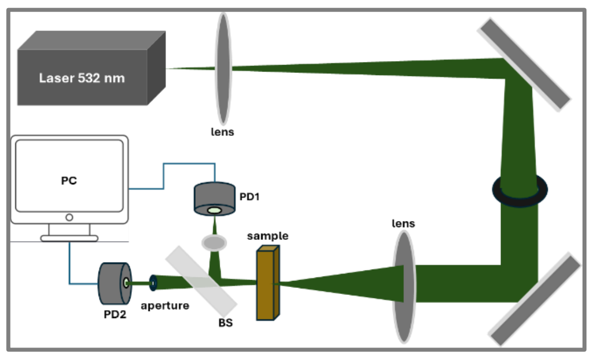

Figure1: Schematic of experimental setup of z-scan experiment, for both open and closed apertures. PD1, PD2 are photodiodes 1,2 respectively. BS: is the beam splitter

extend the beam, a 10 cm focal length lens was used. The beam was spatially filtered using an iris of 14 mm to obtain a semi-columned beam with a diameter of 14 mm. A convex lens with a focal length (f) of 200 mm was then used to focus the beam onto the olive oil sample. The olive oil sample was poured into a cuvette with a 1 mm optical path length and was mounted on a translation phase that can be moved in steps of 100 m.

After the sample, a beam splitter that reflects the beam at 45^o was used to reflect the beam toward a photodiode PD1. The transmitted beam from the beam splitting unit is directed at an i1.75 mm, which serves as the closed aperture, and was then directed to another photodiode PD2. Both photodiodes: PPD2,and PD2 are connected to computers through an interface for data acquisition.

Results and discussions

We performed UV-visible absorption measurements for virgin and refined olive oil, both produced by the same company.

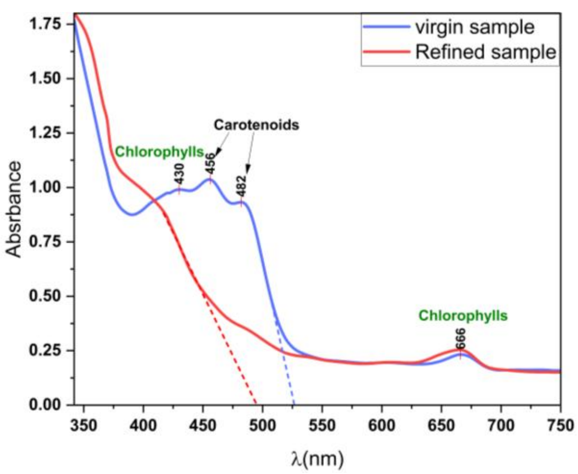

Figure2: Absorption Spectra for virgin olive oil (blue) and refined olive oil(red)

Olive oil primarily consists of saturated and unsaturated fatty acids, triglycerides, and smaller quantities of other components such as glycerol, phosphate, pigments, flavor compounds, and sterol. Among the unsaturated fatty acids, oleic acid ($C_{18}H_34O_2$) and linoleic acid ($C_{18}H_{32}O_2$) are the most abundant. additionally, olive oil contains various organic pigments in the form of carotenoids ($C_{40}H_{56}$), lutein ($C_{40}H_{56}O_2$), chlorophyll A ($C_{55}H_{72}O_5N_4Mg$) and B ($C_{55}H_{70}MgN_4O_6$). In general, the composition of olive oil can vary depending on geographical origin, climate, growing conditions, and harvest time.

As shown in Figure 2, the virgin olive oil sample displays distinct absorption peaks at 430 nm, 456 nm, 482 nm, and 666 nm. The peaks at 430 nm and 666 nm are associated with chlorophylls, while those at 456 nm and 482 nm are attributed to carotenoids. In contrast, in the refined olive oil sample, the carotenoid peaks are completely absent. Moreover, the 430 nm chlorophyll peak is no longer visible, with only a weak shoulder appearing in its place. This indicates a significant loss of chlorophyll during the refining process. These findings confirm that absorption of spectroscopy is a reliable technique for characterizing olive oil and detecting adulteration in the oil market.

Moreover, the absorption edge was observed at approximately 525 nm for virgin olive oil, and around 495 nm for the refined sample. These results indicate that both samples exhibit two-photon absorption when exposed to a 532 nm laser to determine their nonlinear absorption properties.

To further characterize the nonlinearity of both virgin and refined olive oil samples, Z-scan experiment was conducted with closed and open apertures. The closed aperture transmission curves for virgin and refined olive oil samples are shown in figure3, (a) and (b), respectively(22).

The difference between the peak and valley (ΔT_{p-v}) in the normalized transmission is directly related to the induced phase shift $\Delta\phi$ and is given by the following equation:

$$\Delta T_{p-v} = 0.406(1-S)^{0.25}\Delta\phi \qquad (1)$$

Where S is the linear transmission of the aperture and here is given by,

$$S = 1 - \exp\left(\frac{-2r_o^2}{\omega_o^2}\right) \qquad (2)$$

where r_o is the radius of the aperture, ω_o is the radius of the beam at the aperture. The phase shift can be calculated using the closed-aperture transmission data and the following equation [19],

$$T(z) = 1 + \frac{4\Delta\phi}{(X^2+1)(X^2+9)} \qquad (3)$$

Where $X = \frac{Z}{Z_o}$, Z is the position of the sample and Z_o is the Rayleigh range.

The third order nonlinear refractive index n_2 of virgin and refined olive oil were determined using this equation [19],

$$n_2 = \frac{\Delta\phi}{kI_oL_{eff}} \qquad (4)$$

k is the wave vector and $k = \frac{2\pi}{\lambda}$, λ is the wavelength. The L_{eff} is the effective thickness of the sample and can be calculated by:

$$L_{eff} = \frac{1 - e^{-\alpha L}}{\alpha} \qquad (5)$$

Here, α represents the linear absorption coefficient, which can be calculated from the absorbance data shown in figure2 using this relation

$$\alpha = \frac{2.303 * A}{L} \qquad (6)$$

Where A is the absorbance, and L is the thickness of the sample. Here, the absorption coefficient for pure and refined olive oil samples are $\alpha_v = 0.556/\text{cm}$, $\alpha_{re} = 0.667/\text{cm}$, respectively. The values of nonlinear refractive index for both virgin and refined samples were calculated to be $1.92 \times 10^{-11}\ \text{m}^2/\text{W}$ for virgin and $2.54 \times 10^{-11}\ \text{m}^2/\text{W}$ for the refined sample.

These values agree with those values reported in literature [20].

The real part of $\chi^{(3)}$ is related to the second-order nonlinear refractive index by,

$$\chi_R^{(3)} = (4/3)n_o^2\epsilon_o c n_2\left(\frac{m^2}{V^2}\right) \qquad (7)$$

Based on this relation, the $\chi_R^{(3)}$ for virgin and refined olive oil are $1.46 \times 10^{-13}\ \text{m}^2/\text{V}^2$, and $1.935 \times 10^{-13}\ \text{m}^2/\text{V}^2$. The Imaginary part of $\chi^{(3)}$) which is related to the nonlinear absorption coefficient (β), is given by,

$$\chi_I^{(3)} = \left(\frac{n_o^2\epsilon_o c\lambda}{3\pi}\right)\beta\left(\frac{m^2}{V^2}\right) \qquad (8)$$

Where ϵ_0, n_o and λ are, respectively, the permittivity of free space (8.85×10^{-12} F/m), linear refractive index of the material, and the wavelength. To determine χ^3, and n_2, the normalized transmission of the closed aperture was plotted as a function of z and was fitted to equation (3) to calculate the $\Delta\phi$.

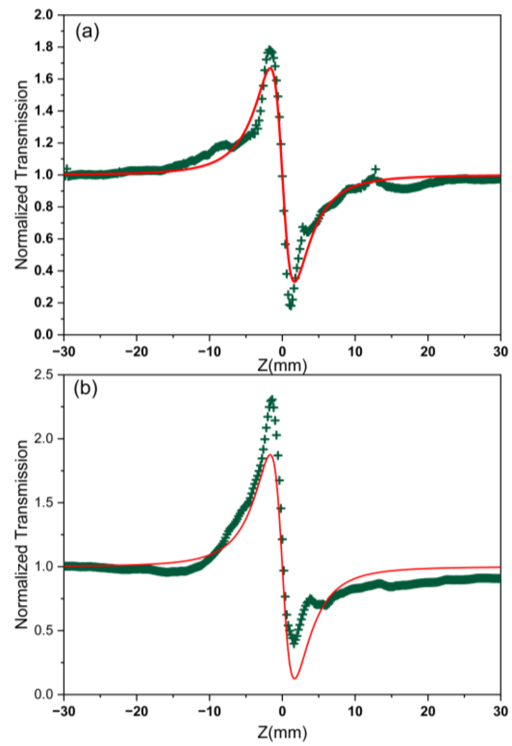

Figure 3: The Closed aperture normalized transmission as a function of axial distance Z(mm) for both virgin , and refined olive oil samples, (a), (b) respectively

To investigate nonlinear absorption, we have conducted open aperture experiments. The results for virgin and refined olive oil are shown in figure4. So, the normalized transmission is plotted as a function of the sample position along the Z-axis (axial distance). The data fitted to the following equation [1–3],

$$T(Z) = 1 - \frac{\beta I_o L_{eff}}{2\sqrt{2}\left(1 + \frac{Z^2}{Z_o^2}\right)} \qquad (9)$$

β here is the nonlinear absorption coefficient. I_o is the intensity at the focues.it was calculted using $I_o = 2P/\pi\omega^2$. The intensity of the laser at the focues was claculted to be $1.2 \times 10^8\ \mathrm{W/m^2}$. The normalized transmission valley represents the nonlinear absorption lead to reverse saturation

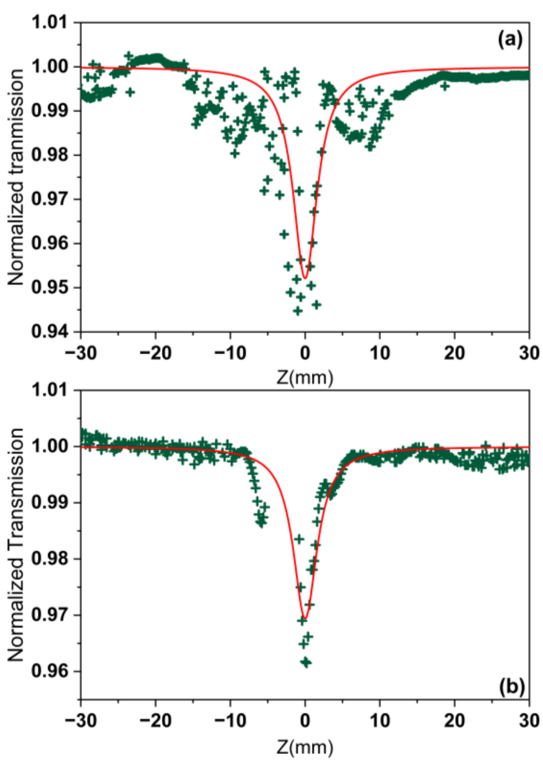

Figure4: Open aperture data for the nonlinear absorption of virgin(a) and refined(b) olive oil

absorption with $\beta > 0$. The nonlinear absorption coefficient was determined to be $1.26 \times 10^{-6}\ \mathrm{m/W}$ for virgin olive oil and $7.84 \times 10^{-7}\ \mathrm{m/W}$ for the

The imaginary part of the third order nonlinear susceptibility was calculated using equation (8). The $\chi_I^{(3)}$ for the virgin olive oil sample is $\chi_I^{(3)}$ is $4.06 \times 10^{-16}\left(\frac{m^2}{V^2}\right)$, and for the refined sample is $2.53 \times 10^{-16}\left(\frac{m^2}{V^2}\right)$. These values are significantly lower than those corresponding to the real part of the third-order susceptibility.

To estimate the third order nonlinear susceptibilities of the virgin and refined olive oil samples, the following equation was used [24].

$$\chi^{(3)} = \sqrt{\left(\chi_R^{(3)}\right)^2 + \left(\chi_I^{(3)}\right)^2} \qquad (10)$$

The calculated values of $\chi^{(3)}$ for both virgin and refined olive oil samples are $1.46 \times 10^{-13}\left(\frac{m^2}{V^2}\right)$, and $1.94 \times 10^{-13}\left(\frac{m^2}{V^2}\right)$ The results reveal slightly higher nonlinear absorption in the virgin olive compared to that for the refined sample.

Measuring the laser spot size is pivotal for the determination of Raleigh range at the focal. The spot size is defined as the radius at which the power has fallen to $1/e^2$ of its peak value at the center of the beam. The beam waist w(z) at any point along Z axis is given by [22]:

$$\omega(z) = \omega_o = \sqrt{1 + \left(\frac{M^2 \lambda z}{\pi \omega_o^2}\right)^2} \qquad (11)$$

where ω_o is the smallest beam waist (spot size), which is obtained at z=0, λ is the wavelength, and M^2 is the quality factor for the Gaussian beam.

Table 1: nonlinear parameters for both virgin and refined olive oil sample

sample	$n_2\left(\frac{m^2}{W}\right)$	$\beta\left(\frac{m}{W}\right)$	$\chi_R^{(3)}\left(\frac{m^2}{V^2}\right)$	$\chi_I^{(3)}\left(\frac{m^2}{V^2}\right)$	$\chi^{(3)}\left(\frac{m^2}{V^2}\right)$
Virgin olive oil	1.92×10^{-11}	1.26×10^{-6}	1.46×10^{-13}	4.06×10^{-16}	1.46×10^{-13}
Refined olive oil	2.54×10^{-11}	7.84×10^{-7}	1.935×10^{-13}	2.53×10^{-16}	1.94×10^{-13}

The axial distance from the smallest beam waist to the point at which the beam waist is increased by a factor of √ 2 is called the Rayleigh length Z_R, which given by [23]:

$$Z_R = \frac{\pi \omega_o^2}{M^2 \lambda} \quad (12)$$

The spot size here was measured using the knife edge technique. In this technique, the beam is scanned spatially along X or Y, and the spot size is determined by measuring the transmitted power. To ensure accuracy, the transmission was collected after each fine move -100μm along X-axis`

$$P = P_o + \frac{P_{max}}{2}\left[1 - erf\left(\frac{\sqrt{2}(x - x_o)}{\omega}\right)\right] (13)$$

To determine the beam waist, the data shown in Figure 5(a) were fitted using an error function in equation (13). This fitting process was repeated at multiple positions along the z-axis. The resulting beam waist values were fitted to Equation (11) to calculate the minimum beam waist. The minimum measured value is 17 μm, as shown in Figure 5(b). this value was then substituted in equation (9) to calculate the nonlinear absorption coefficient, β.

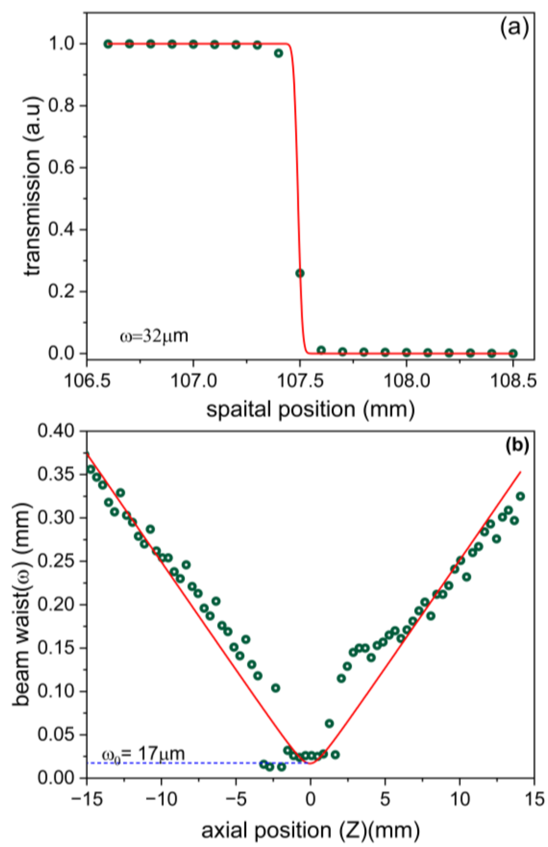

Figure 5: knife edge measurements to determine the beam waist

Conclusion

This study comprehensively examined the linear and nonlinear optical properties of virgin and refined olive oil using a 532 nm continuous-wave laser. The Z-scan technique was used to determine nonlinear absorption and nonlinear refractive indices of both samples. UV-visible spectroscopy was employed to analyze the linear absorption behavior, revealing the absorption edge for each oil type. The analysis revealed that two-photon absorption is the dominant nonlinear mechanism in both oil types. These findings confirm the effectiveness of optical techniques for characterizing olive oil and differentiating between virgin and refined samples

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