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Z-scan measurements of the third order optical nonlinearity of C_{60} doped poly(ethylacetylenecarboxylate) under CW regime

M.D. Zidan[a,*], M.B. Alsous[a], A.W. Allaf[b], A. Allahham[a], A. AL-Zier[b], H. Rihawi[a]

[a] Department of Physics, Atomic Energy Commission, P.O. Box 6091 Damascus, Syria
[b] Department of Chemistry, Atomic Energy Commission, P.O. Box 6091 Damascus, Syria

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ABSTRACT

The third order nonlinear optical properties of C_{60}/poly(ethylacetylenecarboxylate) have been studied using Z-scan technique. Experiments are performed using a CW Diode laser at 635 nm wavelength and 20 mW power. The nonlinear absorption coefficient β, nonlinear refractive index n_2, Re χ^3, and Im χ^3 in C_{60} poly(ethylacetylenecarboxylate) are measured using Z-scan data. Our results show that doping C_{60} into poly(ethylacetylenecarboxylate) has enhanced and contributed to the increase of the NLO property of the polymer.

1. Introduction

Since the lasers have been used in various applications, it was recognized that intense laser beams can damage the human eyes and solid-state optical sensors [1–3]. Thus, the search for new nonlinear optical materials to serve as passive optical limiters is very important for potential public safety issue. Up to now, a number of optical limiting materials that exhibit nonlinear optical effect, including organic molecules [4,5], organic dyes [6], conjugated compounds [7], and fullerenes molecules and their derivatives [8,9], have been found to exhibit a strong nonlinear optical effect at high-intensity laser beam and hence could serve as candidates for practical optical limiters. The optical limiting properties of fullerenes and their modified derivations have been widely investigated by doping into solid matrices [10–12] or solutions [13,14].

Z-scan technique proposed by Sheik-Bahae [15,16] based on the spatial distortion of a laser beam, passed through a nonlinear optical material, is widely used in material characterization because of its simplicity and high sensitivity. It can be used to determine both the nonlinear absorption coefficient β and the nonlinear refractive index n_2 of optical nonlinear materials. Early work reported on open-aperture Z-scan on C_{60} solution are performed over the visible region (440–660 nm) using OPO [17], and C_{60} in toluene [18]. The nonlinear absorption coefficients of C_{60} in toluene solution was measured using 532 and 1064 nm [19], also the open aperture Z-scan of C_{60}, C_{60}/polymer (C_{60}/PSVPY32 and C_{60}/PS) was measured [20].

Following our previous investigations on C_{60} doped acetylenedicarboxylic acid polymer [21], and investigation of optical nonlinearity of C_{60} doped polymer [22], this paper reports the Z-scan measurements of C_{60} doped poly(ethylacetylenecarboxylate) using a CW diode laser at 635 nm wavelength and a power of 20 mW. To our knowledge, there is no report published before on the investigation of the Z-scan measurements of C_{60} doped poly(ethylacetylenecarboxylate).

2. Experimental techniques

C_{60} was purchased from Fluka and used as received without any further purification.

Poly(ethylacetylenecarboxylate) material has been prepared and identified [23]. The method of preparing the sample of C_{60}/poly(ethylacetylenecarboxylate) was mentioned in details in previous publication [24]. The concentrations samples of C_{60} dissolved in toluene, C_{60}/poly(ethylacetylenecarboxylate) and poly(ethylacetylenecarboxylate) are 5.0×10^{-4}, 4.40×10^{-4} and 10^{-3} M/l, respectively.

The Z-scan experimental setup was analogous to that described in Ref. [25]. The measurements were done with linearly polarized TEM_{00} Gaussian beam of a CW diode laser at 20 mW (λ = 635 nm). The experimental parameters during the measurements of the samples were used as follows: the laser beam is focused by a 5 cm focal length lens to a waist radius ω_0 of 22 µm at the focal point. The diffraction length z_0 is larger than 2.39 mm, the radius of the

* Corresponding author. Tel.: +963 11 2132580; fax: +963 11 6112289.
E-mail address: PScientific8@aec.org.sy (M.D. Zidan).

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aperture is $r_a = 0.4$ mm and $\omega_a = 1.75$ mm is the radius beam waist on the aperture. The sample cell used in this work is a 2 mm thick quartz cell. The cell was hold on an optical rail and translated across the focal region along the axial direction that is in the direction of the propagation of the laser beam. The transmitted power through the sample is measured as a function of the sample distance z from the waist plane of the Gaussian beam. The transmission of the beam through an aperture placed in the far field is measured with a power meter (Thorlabs PM300E).

3. Results and discussions

Z-scan measurements, in the cases of open and closed aperture, allow us for the determination of the nonlinear absorption coefficient β and the nonlinear refractive index n_2, respectively.

Fig. 1 displays the open aperture Z-scan results of C_{60}, poly(ethylacetylenecarboxylate), and C_{60}/poly (ethylacetylenecarboxylate) in toluene. The nonlinear absorption coefficient β can be obtained from a best fitting performed on the experimental data of open aperture (OA) measurement with equation [15,16]:

$$T(z) = \sum_{m=0}^{\infty} \frac{(-q_0)^m}{(m+1)^{3/2}} \tag{1}$$

For $q_0 < 1$, where $q_0\,(z)$ is a function of I_0, $L_{\rm eff}$ and β:

$$q_0(z) = I_0\, L_{\rm eff}\, \beta/(1 + z^2/z_0^2) \tag{2}$$

Solving the summation and for $\alpha_0 \ll 1$;

$$T\,(z) = 1 - (I_0\;\; L_{\rm eff}\, \beta)/[2^{3/2}(1 + z^2/z_0^2)] \tag{3}$$

where $L_{\rm eff} = (1 - \exp(-\alpha_0 L))/\alpha_0$ is the effective thickness of the sample, L is the thickness of the sample, α_0 is the linear absorption coefficient, $z_0 = \pi\omega^2{}_0/\lambda$ is the diffraction length of the beam, λ is the laser wavelength, and $I_0 = 1315\ \mathrm{W/cm^2}$ is the intensity of the laser beam at the focus $z = 0$. The solid line in Fig. 1 is the fitting curve while the symbols are the experimental data.

To determine the sign and magnitude of the nonlinear refractive index (n_2) of the studied samples, a closed aperture (CA) Z-scan was performed by placing a small diameter aperture in front of the detector. Since the closed aperture transmittance is affected by the nonlinear refraction and nonlinear absorption components, it is necessary to separate the effect of the nonlinear refraction components 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 Z-scan. The data obtained in this way reflect purely the effect of nonlinear refraction.

Fig. 2 shows the pure nonlinear refraction curves of C_{60}, poly(ethylacetylendicarboxylate), and C_{60}/poly(ethylacetylendicarboxylate) in toluene, obtained by division method.

To calculate the nonlinear refractive index (n_2), the on-axis nonlinear phase shift at the focus, $\Delta\phi_0$, was obtained through fitting the CA/OA curve (Fig. 2) with the following approximate equation [15,16]:

$$T(z,\,\Delta\phi) = 1 - \frac{4\Delta\phi_0 X}{(X^2+9)(X^2+1)} \tag{4}$$

where $X = (Z/Z_0)$, and T is the normalized transmittance for CA/CO curve. Then, the nonlinear refractive index n_2 can be obtained through the given equation [15,16]:

$$n_2 = \frac{\lambda\, \Delta\phi_0}{2\pi I_0 L_{\rm eff}} \tag{5}$$

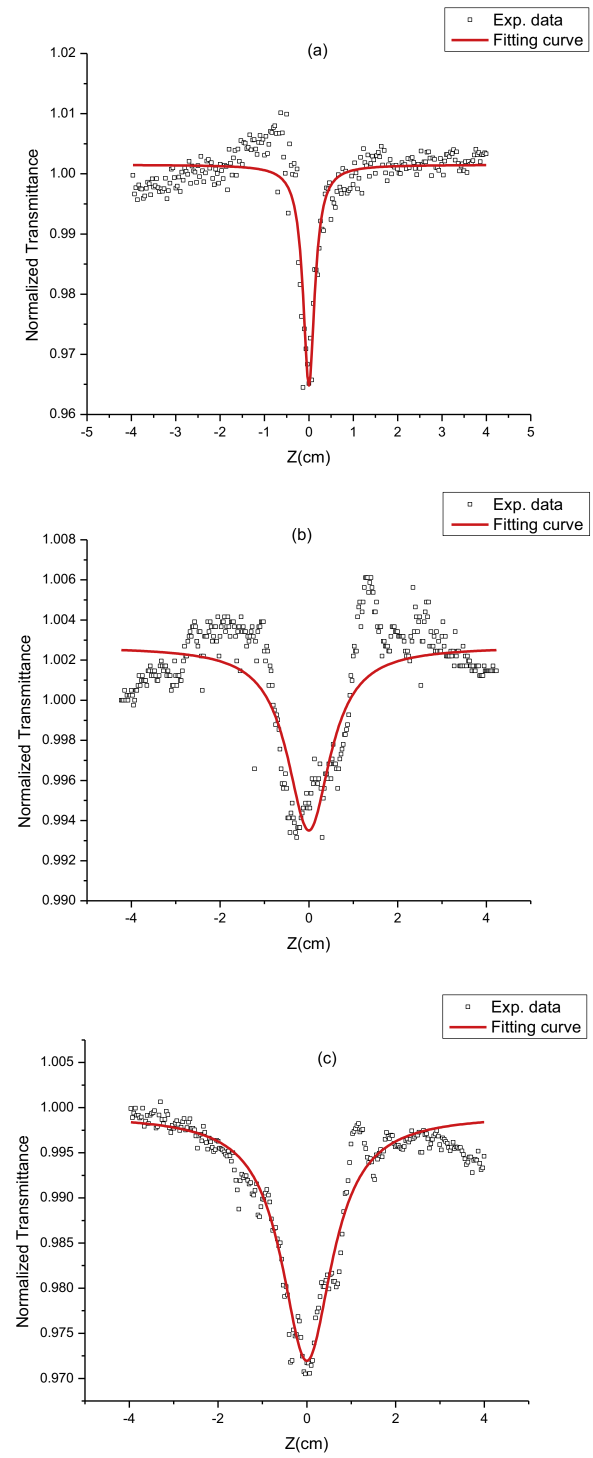

Fig. 1. Open – aperture Z-scan data (a) C_{60} in toluene, (b) poly(ethylacetylenecarboxylate) and (c) C_{60}/poly(ethylacetylenecarboxylate).

The solid line in Fig. 2 is the fitting curve while the symbols are the experimental data.

The obtained values of the nonlinear refractive index n_2 and nonlinear absorption coefficient β can be used to determine the real and imaginary parts of the third-order nonlinear optical susceptibility (χ^3) according to the following relations [15,16]:

$$\mathrm{Re}\,\chi^3(\mathrm{esu}) = (10^{-4}\,\varepsilon_0 c^2 n_0^2/\pi)\;n_2\,(\mathrm{cm^2/w}) \tag{6}$$

$$Im\,\chi^3(\mathrm{esu}) = (10^{-2}\,\varepsilon_0 c^2 n_0^2 \lambda/4\pi^2)\,\beta\,(\mathrm{cm/w}) \tag{7}$$