

## Non-Linear Optical Properties of Nano Particle C<sub>60</sub> Fullerene Using Lasers

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### Abstract

The third order non-linear optical properties of Buckminster fullerene (C<sub>60</sub>) molecule has been studied using a Nd:YAG laser, in the visible and in the infrared region. The solvent using toluene was specifically used because of low threshold intensity for an optical limiter application. Closed aperture Z-scan technique was adopted to characterize the material due to its simplicity and high sensitivity in measuring the third-order optical nonlinearity. This allows computing the contributions of nonlinear absorption and nonlinear refraction towards nonlinearity. Saturable Absorption (SA) for C<sub>60</sub> nano particles is also established. RSA is not established. FT-IR studies is also carried out to characterize the sample and to correlate the NLO studies.

**Keywords:** Buckminster fullerene; Non-linear properties; Visible and infrared regions; Nd: YAG laser; Third order NLO property; Optical limiter; Non-linear refraction

### Introduction

Nanotechnology plays an important role in finding new nano scale materials for the benefit of renewable energies. Nano structured materials are mainly used in applications such as hydrogen and methane storage, fuel cells, solar cells, bio-fuel cells, rechargeable batteries, super capacitors, electrodes, catalysts, gas sensors and other applications. For developing new or novel materials, it is required to synthesize, fabricate, characterize and process these nano materials for any specific application. One such study is the non-linear optical property (NLO) of this C<sub>60</sub> Buckminsterfullerene nano particles. The C<sub>60</sub> molecule was discovered by Zhou et al. [1] during conduction of an experiment involving graphite under an inert atmosphere of helium. C<sub>60</sub> and other such similar structures are considered as promising non-linear optical materials due to its non-linear refraction and scattering process which is a necessary property for optical limitation.

An optical limiter is a device that will have high transmission of low input signal and for large input signal there will be constant output signal. For example, an intense laser or a light beam can damage the eye. So by using properly designed lenses, one can protect the eye. The other application of NLO property is in shaping of short laser pulses. Simplicity and a fast response time is the main reason for choosing NLO property of C<sub>60</sub> material. Saturable absorption, also known as SA is the well-known mechanism for optical limiting devices. SA is applicable for molecular systems because the excited state cross-section  $\sigma_{ex}$  is larger than the ground state cross-section  $\sigma_g$  which is an ideal situation for an optical limiter application. There are several characterization techniques available for measuring the third-order optical nonlinearities these includes degenerate four-wave mixing, nearly degenerate three-wave mixing, optical Kerr effect, ellipse rotation, interferometric methods, two beam coupling, beam self-bending and third harmonic generation [2]. Among the available techniques z-scan technique offers simplicity as well as very high sensitivity in measuring the third-order optical nonlinearity and also allows computing the contributions of nonlinear absorption and nonlinear refraction towards the nonlinearity. Z-scan technique is based on the principle of spatial beam distortion. It was originally proposed by Sheik-Bahae, has been since then implemented and applied to the study of third-order optical nonlinearity. Using z-scan technique, the magnitude of nonlinear absorption and the sign and magnitude of nonlinear refraction can be determined

simultaneously. When a high intensity laser beam propagates through a material, induced refractive index changes leads to self-focusing or defocusing of the laser beam. This enables to determine the third-order nonlinear optical properties of various materials in liquid, thin film or crystal forms. In this technique, the sample under investigation is moved along the tightly focused Gaussian laser beam. The intensity of the laser beam changes as the sample is moved. This is because the sample experiences different intensities, depending on the position of the sample relative to focus (z=0). The power transmitted through the sample is measured by translating the sample along the z-direction through the beam waist of a focused beam and hence the name z-scan.

### Experiment

We obtained a pure research grade 99.999% pure M/s. Merck Co.Ltd. for the analysis of C<sub>60</sub> nanoparticles. Initially we tried to record the second order optical non-linearity of the C<sub>60</sub> fullerene molecule with the help of a Nd: YAG pulsed laser with wavelength equal to 1.064 micrometre. But the sample did not show any absorption as the IR beam did not pass through it. The IR light was reflected back from the sample without any absorption and so it is reported that there is no second order optical non-linearity for this sample. Thereafter, we proceeded to do the Z-scan experiment to study the third order optical non-linearity using a Nd: YAG laser with a second harmonic output wavelength of 532 nm [3].

Since fullerene molecule shows high volatility it is dissolved in solution to perform NLO studies and so, the C<sub>60</sub> nano particles were dissolved in a solvent toluene with a transmittance of 64%. The reason to choose toluene is that it gives a low threshold intensity, to be used as an optical limiter, than any other solution like carbon-black, chloronaphthalene etc [4]. The sample cell was kept in front of a Nd:YAG laser using 532 nm second-harmonic generated beam in the visible

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region, for the study of third order NLO properties. The experimental set-up is shown here below in Figure 1 and the schematic of the z-scan experimental set up is shown in Figure 2 below.

The characterization of the sample was also done using the FT-IR for percentage of transmission of the C<sub>60</sub> molecule and it was found

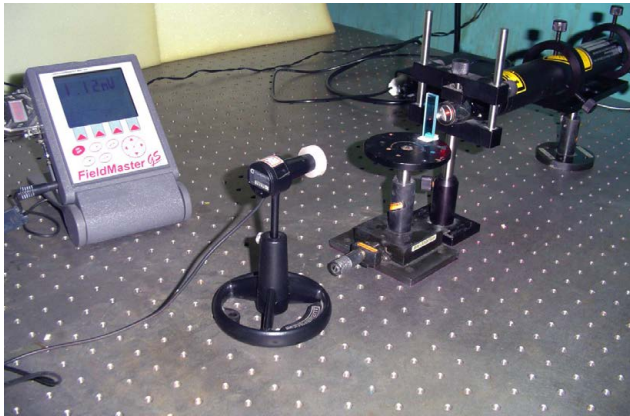


Figure 1: Z-scan set up using Nd: YAG laser as a source of light.

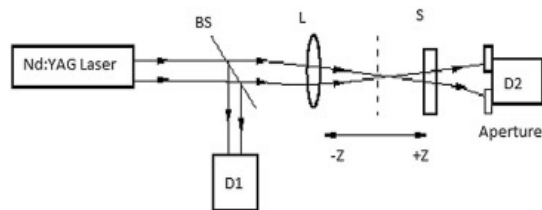


Figure 2: Schematic of the Z-scan experimental set-up using a Nd: YAG laser.

that the sample showed highest transmission (reference is 4000 cm<sup>-1</sup> at 100%), (97.8%) at 450 cm<sup>-1</sup> (0.002222 cm wavelength) and the lowest transmission percentage (5%) at 1426 cm<sup>-1</sup> (0.00070126 cm wavelength), indicating that at the lower wavelength, the sample has low percentage of transmittance and at higher wavelength it has high percentage of transmittance. Thus the third order nonlinear refractive index property can be observed at higher wavelengths due to high transmittance of light in the visible region [4]. The FT-IR spectrum recorded is shown below in Figure 3, (Table 1).

The recorded parameters for the C<sub>60</sub> fullerene molecules during our experiment are, namely, Kerr nonlinearity is found to be (n<sub>2</sub>)=5.38 x 10<sup>-8</sup> cm<sup>2</sup>/W,

Two-photon absorption coefficient TPA (β)=0.04 × 10<sup>-4</sup> cm/W,

Linear refractive index (n<sub>0</sub>)=1.13, third order electric susceptibility Re (χ<sup>3</sup>)=1.73 × 10<sup>-6</sup> esu,

Imaginary part of the third order electric susceptibility Im (χ<sup>3</sup>)=0.42 × 10<sup>-6</sup> esu, third order electric susceptibility (χ<sup>3</sup>)=1.79 × 10<sup>-6</sup> esu. The Table 2 gives the details as shown below.

It is seen from the Table 2 above that the contributions of nonlinear absorption (β) and nonlinear refraction (n<sub>2</sub>) towards nonlinearity is established [5-10]. The laser intensity dependent refractive index in the third order, namely, (χ<sup>3</sup>) has been prominent, showing negative signs for the absorptive nonlinearities. We attribute this negativity to saturable absorption [11,12]. This shows that C60 Fullerene nano particle is suitable for an optical limiting device, plasmon waveguide, sensor protection, medicine, and nano probes.

A graphical plot and a comparison between closed aperture and open aperture experiment and also the ratio between the two types is shown below.

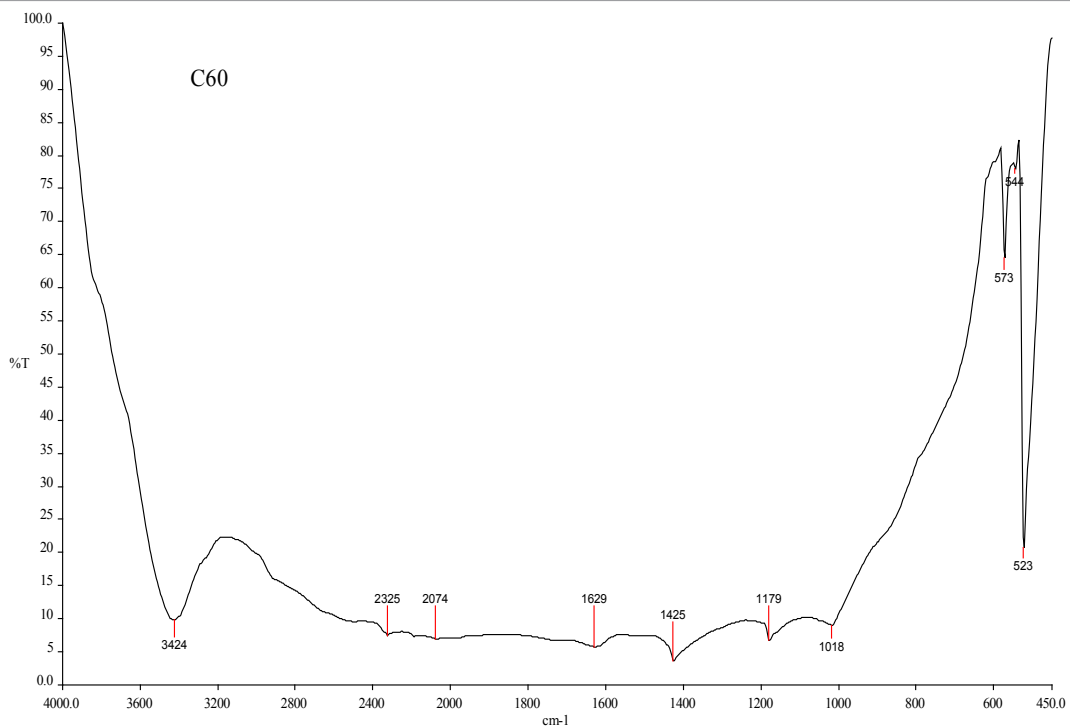


Figure 3: FT-IR transmission spectrum of C<sub>60</sub> recorded between 450 cm to 4000 cm<sup>-1</sup>.

1/λ(cm <sup>-1</sup> )	%Transmittance
492.000000	55.9774
491.000000	57.1641
490.000000	58.3859
489.000000	59.6555
488.000000	60.99971
487.000000	62.44777
486.000000	63.99795
485.000000	65.60268
484.000000	67.19168
483.000000	68.71644
482.000000	70.17152
481.000000	71.56383
480.000000	72.89073
479.000000	74.15598
478.000000	75.38279
477.000000	76.60604
476.000000	77.85104
475.000000	79.11794
474.000000	80.38951
473.000000	81.64969
472.000000	82.89004
471.000000	84.10621
470.000000	85.29494
469.000000	86.45373
468.000000	87.58083
467.000000	88.67436
466.000000	89.73207
465.000000	90.75125
464.000000	91.72814
463.000000	92.65752
462.000000	93.53251
461.000000	94.34452
460.000000	95.08398
459.000000	95.74079
458.000000	96.30546
457.000000	96.7717
456.000000	97.13885
455.000000	97.4127
454.000000	97.60391
453.000000	97.72556
452.000000	97.79342
451.000000	97.8248
450.000000	97.83732
1464.000000	7.169664
1463.000000	7.137567
1462.000000	7.102927
1461.000000	7.065651
1460.000000	7.025616
1459.000000	6.982962
1458.000000	6.938173
1457.000000	6.891164
1456.000000	6.841555
1455.000000	6.789933
1454.000000	6.736916
1453.000000	6.682667
1452.000000	6.627128
1451.000000	6.570048
1450.000000	6.511411
1449.000000	6.451523
1448.000000	6.390528

1447.000000	6.328339
1446.000000	6.265105
1445.000000	6.201242
1444.000000	6.137536
1443.000000	6.075523
1442.000000	6.016985
1441.000000	5.962674
1440.000000	5.911109
1439.000000	5.857932
1438.000000	5.795723
1437.000000	5.714539
1436.000000	5.603435
1435.000000	5.453477
1434.000000	5.260926
1433.000000	5.028564
1432.000000	4.765842
1431.000000	4.489079
1430.000000	4.220649
1429.000000	3.984898
1428.000000	3.801625
1427.000000	3.681865
1426.000000	3.627782
1425.000000	3.690331
1424.000000	3.690331
1423.000000	3.780523
1422.000000	3.888246
1421.000000	3.999439
1420.000000	4.104804
1419.000000	4.199532
1418.000000	4.282075
1417.000000	4.353808
1416.000000	4.418198
1415.000000	4.478963
1414.000000	4.538774
1413.000000	4.598907
1412.000000	4.659609
1411.000000	4.720651

**Table 1:** below gives data related to minimum and maximum percentage transmission of a FT-IR spectrum recorded for C<sub>60</sub> molecule.

$n_2 \times 10^{-8} \text{ cm}^2/\text{W}$	$\beta \times 10^4 \text{ cm/W}$	n0	$\text{Re } \chi^{(3)} \times 10^{-6} \text{ esu}$	$\text{Im } \chi^{(3)} \times 10^{-6} \text{ esu}$	$\chi^{(3)} \times 10^{-6} \text{ esu}$
5.38	0.07	1.13	1.73	0.42	1.79

**Table 2:** C<sub>60</sub> fullerene molecule- recorded parameters during z-scan experiment.

From the graph seen in Figures 4 and 5, it is seen that linear absorption coefficient has been steadily increasing from the centre on both sides, with the distance of z scan from the centre of the cell in which the solvent is kept. From the Figure 6, we can infer that the solubility in toluene is complete and the sample has shown more transmittance towards the centre of the cell and hence this non-linear refractive index property will be suitable to act as an optical limiter for C<sub>60</sub> nano particles. The data from which the graph has been plotted is shown in Table 3 below.

## Result and Discussion

For the C<sub>60</sub> nano particles, closed aperture Z-scan technique was adopted to characterize the material due to its simplicity and high sensitivity in measuring the third-order optical nonlinearity. This allows computing the contributions of nonlinear absorption and nonlinear refraction towards nonlinearity. The characterization of the

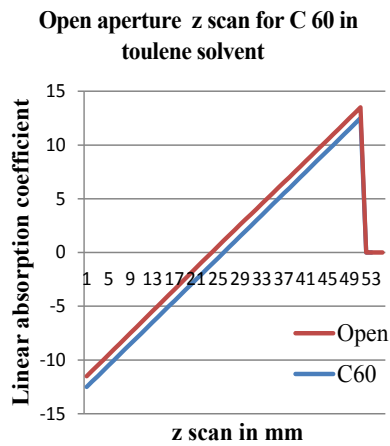


Figure 4: Graph plot for open aperture in C<sub>60</sub> z scan.

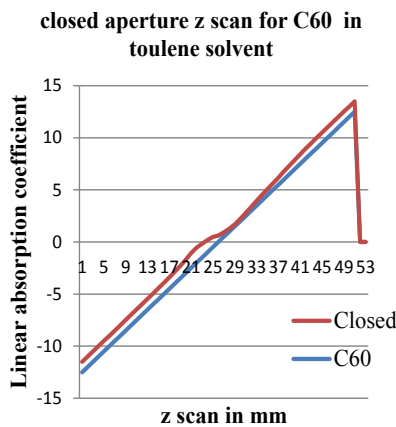


Figure 5: Graph plot for closed aperture in C<sub>60</sub> z scan.

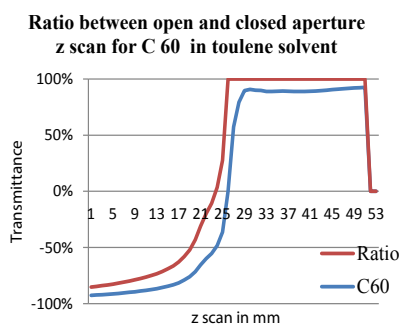


Figure 6: Ration between closed and open aperture for C<sub>60</sub> z scan.

sample was also done using the FT-IR for percentage of transmission of the C<sub>60</sub> molecule and it was found that the sample showed highest transmission (reference is 4000 cm<sup>-1</sup> at 100%), (97.8%) at 450 cm<sup>-1</sup> (0.002222 cm wavelength) and the lowest transmission percentage (5%) at 1426 cm<sup>-1</sup> (0.00070126 cm wavelength), indicating that at the lower wavelength, the sample has low percentage of transmittance and at higher wavelength it has high percentage of transmittance. It is seen from the experimentally recorded data above, that the contributions

Linear Abs. Co.	Closed	Open	Ratio
-12.5	0.9999	0.999999	0.999901
-12	0.9999	0.999999	0.999901
-11.5	0.9999	0.999999	0.999901
-11	0.9999	0.999999	0.999901
-10.5	0.9999	0.999999	0.999901
-10	0.9999	0.999999	0.999901
-9.5	1.00427	0.999999	1.004271
-9	1.00555	0.999999	1.005551
-8.5	1.012104	1.004621	1.007448
-8	1.016305	1.011863	1.00439
-7.5	1.021907	1.021484	1.000413
-7	1.03211	1.029306	1.002725
-6.5	1.031523	1.035471	0.996188
-6	1.044799	1.041777	1.0029
-5.5	1.057986	1.049689	1.007905
-5	1.065396	1.0559	1.008994
-4.5	1.086287	1.06259	1.022301
-4	1.133863	1.068322	1.061349
-3.5	1.184152	1.074534	1.102015
-3	1.285656	1.081074	1.189239
-2.5	1.431878	1.086574	1.317791
-2	1.459786	1.091766	1.337087
-1.5	1.326546	1.096317	1.210003
-1	1.18136	1.10076	1.073223
-0.5	0.972474	1.104559	0.880419
0	0.615292	1.104399	0.557129
0.5	0.4092	1.098236	0.372598
1	0.283165	1.08956	0.25989
1.5	0.188756	1.080745	0.174654
2	0.219205	1.072832	0.204324
2.5	0.297436	1.065832	0.279064
3	0.357654	1.0559	0.33872
3.5	0.459391	1.049689	0.437645
4	0.518974	1.043727	0.497232
4.5	0.572906	1.037267	0.552323
5	0.615473	1.031055	0.596935
5.5	0.686587	1.024844	0.669943
6	0.764568	1.018633	0.750582
6.5	0.815384	1.012422	0.80538
7	0.87584	1.006211	0.870434
7.5	0.917654	1.002987	0.914921
8	0.958749	0.999999	0.958749
8.5	0.976544	0.999999	0.976544
9	0.9999	0.999999	0.999901
9.5	0.9999	0.999999	0.999901
10	0.9999	0.999999	0.999901
10.5	0.9999	0.999999	0.999901
11	0.9999	0.999999	0.999901
11.5	0.9999	0.999999	0.999901
12	0.9999	0.999999	0.999901
12.5	0.9999	0.999999	0.999901

Table 3: Data on linear absorption coefficient, closed, open apertures and their ratio.

of nonlinear absorption ( $\beta$ ) and nonlinear refraction ( $n_2$ ) towards nonlinearity is established. The laser intensity dependent refractive index in the third order, namely, ( $\chi^3$ ) has been prominent, showing negative signs for the absorptive nonlinearities. The non-linear refractive index property of C<sub>60</sub> nano particles is thus suitable to act as an optical limiter, plasmon waveguide, sensor protection, medicine, and nano probes.

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