Z-scan determination of two-photon absorption in Ag nano-fluid prepared using γ-radiation methods

E. SHAHRIARI[*], W. MAHMOOD YUNUS, K. NAGHAVI
Department of Physics, Universiti Putra Malaysia, 43400UPM, Serdang

In this work the two-photon absorption (TPA) coefficients of Ag nano-fluid was measured using Z-scan technique. The Ag nano-fluid sample was prepared at $4.71{\times}10^{-3}$ M concentrations and was radiated at different doses of γ-radiation. The conventional single beam Z-scan method with open aperture technique was used to measure the nonlinear absorption coefficient. A CW laser operated at 532 nm wavelength with output power of 40 mW was employed as the excitation source. The nonlinear absorption coefficient obtained was $5.8{\times}10^{-3}$, $4.5{\times}10^{-3}$ and $3.2{\times}10^{-3}$ cm/W. The TPA processes was confirmed by optical limiting experimental data for the same laser beam excitation. The results show that the particle size gives a significant effect to the TPA coefficient.

1. Introduction

The characterization of the nonlinear optical properties of materials is an active field of research because of its many potential applications such as optical signal processing, and optical devices [1-5]. One of the important fields of research in modern optics is multi-photon absorption because of its potential application in optical limiting, frequency conversion [6-11], two photon fluorescence microscopy [12], and three dimensional optical data storage [13]. Z-scan technique is one of the simple and effective tools for measuring the third order of nonlinear optics such as nonlinear coefficient refraction and absorption. Thus the technique has been widely used in characterizing nonlinear properties of material [14]. The technique can simultaneously provide the magnitude of real and imaginary parts of nonlinear refractive index and can also determine the sign of the real and imaginary part of susceptibility [15].

The nonlinear optical properties of metal Ag colloidal solution prepared by the chemical reaction method have been reported under CW laser (633 nm) excitation [1]. Their results showed that all the open aperture Z-scan measurements curves were linear. Thus the sample has only nonlinear refractive properties.

Most of the reported nonlinearities characterization of nanoparticles in dielectric materials was performed by the use of lasers with wavelengths close to the absorption maximum of the surface plasmon resonance of nanoparticles [16]. For Ag nanoparticles when the aggregation of nanoparticles increases, it will be broaden the plasmon resonance peak and will give rise to a spectral feature at longer wavelengths [17]. In the present work, a Z-scan method was applied for measuring two-photon absorption of Ag nano-fluid prepared using γ-radiation method. The objective of the works is to study the effect of particle size on the two-photon absorption processes.

We also investigated optical limiting of these samples since the need has rapidly increased for optical devices such as optical detectors and optical switching which protect sensitive components from intense optical radiation. Many investigations for nonlinear optics properties and optical limiting have been reported extensively [18, 19].

2. Material and methods

Nanoparticle samples were prepared using the similar procedure as described elsewhere [20]. Here a brief synthesis of the Ag nanofluids in PVP is presented. For preparing Ag nano-particle in polyvinylpyrrolidone (PVP), 40 mg of silver nitrate, ($AgNO_3$, Aldrish-99%), 4 g polyvinylpyrrolidone (PVP, MW 29,000 Aldrich), 1 ml isopropanol were used. The PVP and isopropanol were used as a colloidal stabilizer and radical scavenger of hydroxyl radical, respectively. The PVP solutions were made by dissolving PVP powder in 50 ml deionized water at room temperature. The solution was magnetically stirred for 2 hours and was bubbled with nitrogen gas (99.5%) in order to remove oxygen. The concentration of Ag nano-particle in PVP solution was calculated to be $4.71{\times}10^{-3}$ M. The γ-radiation (^{60}Co-rays) was used as an effective tool for polymerization process and reducing agent. Silver nitrate ($AgNO_3$) was added into PVP solution and isopropanol. Samples were then irradiated with γ-radiation at three different doses i.e. 40, 20 and 10 kGy. In this process, γ-irradiation produces hydrated electrons that reduce the silver ions to silver atoms, which then aggregated in the solution. The average diameters of Ag nano-particles were measured using nanophox machine

(Sympatec GmbH, D-38678). In this case three series of samples were referred as S1-40kGy, S2-20kGy and S3-10kGy, with respective nanoparticles size of 32.3, 38.5 and 41.8 nm respectively.

The Z-scan experiments were performed using a 532 nm laser beam (Coherent Compass SDL-532-150T). The beam was focused to a small spot using a lens and the sample was moved along the z-axis by a motorized translational stage. The transmitted light in the far field was recorded by detector D. The laser beam waist ω_0 at the focus length was measured to be 24.4 μm and the Rayleigh length was found to be satisfied the basic criteria of our Z-scan experiment. A quartz optical cell containing specimen solution was translated across the focal region along the z-axial direction. The linear absorption coefficient of the samples was measured by conventional method on the basis of $\alpha = -(1/L)ln(I_o/I)$ in the linear regime of the experiment. The schematic experimental set up is shown in Fig. 1.
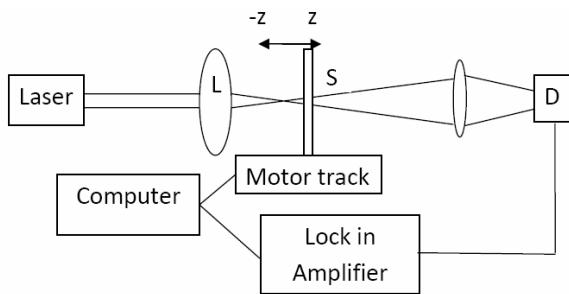
Fig. 1. Schematic diagram of Z-scan experiment setup: L, Lens; S, Sample; D₁, Detector

The optical power limiting property of the solution was measured with the same laser source as mentioned above. The laser excites the samples near the focal point. To obtain the optimum optical limiting characteristics, the sample was fixed at the position of the minimum value of the Z-scan curve (valley) where the input power and transmitted power were measured by a power meter. The incident beam intensity was varied from low to high and the signal was normalized with pure solvent of sample. The optical limiting phenomenon was observed by measuring the nonlinear power-dependent transmission.

3. Results and discussion

3.1. Nonlinear absorption

In this work we measured the optical beam propagation in Ag nano-particles solution by a single beam technique and the experimental data were analyzed by considering two-photon absorption (2PA) [21].

By using a well known two-photon absorption (2PA) equation proposed by Sheik Bahae, 1990 [14]

$$T(z) = \sum_{m=0}^{\infty} \left(\frac{\beta I_0 l_{eff}}{1 + z^2/z_0^2} \right)^m (m+1)^{-3/2} \qquad (1)$$

where $l_{eff} = [1 - \exp(-n\alpha_0 L)]/\alpha_0$ and, α_0 is the linear absorption of the medium, L is the thickness of the sample and $z_0 = \pi\omega_0^2/\lambda$ is the Rayleigh length.

Figs. 2(a)-(c) show the open aperture experimental data of Ag nanoparticle obtained for Ag nano-fluid at a concentration of 4.71×10^{-3} M using 532 nm excitation laser beam. The solid lines in the figures are theoretical fits based on the two-photon absorption phenomenon described by equation 1. Using the laser beam intensity at the focus point was as 4.27×10^3 W/cm², we calculated the two-photon absorption for three samples with different particles size. The values are listed in table 1. The results show that the two-photon absorption tends to decrease as the size of nanoparticles increases. This is due to large value of the volume fractions for small particles size thus, when the particles size become smaller, more particles are thermally agitated resulting in enhanced effect on two-photon absorption.

Table 1 The calculated values of two-photon coefficients.

Nano-Fluid samples	Concentration (M)	Average Particle size (nm)	β (2PA) (cm/W)	β(Optical limiting) (cm/W)	α (cm⁻¹)
S1	4.71×10^{-3}	32.3	5.8×10^{-3}	5.3×10^{-3}	9.01
S2	4.71×10^{-3}	38.5	4.5×10^{-3}	4.8×10^{-3}	8.22
S3	4.71×10^{-3}	41.8	3.2×10^{-3}	3.5×10^{-3}	8.02
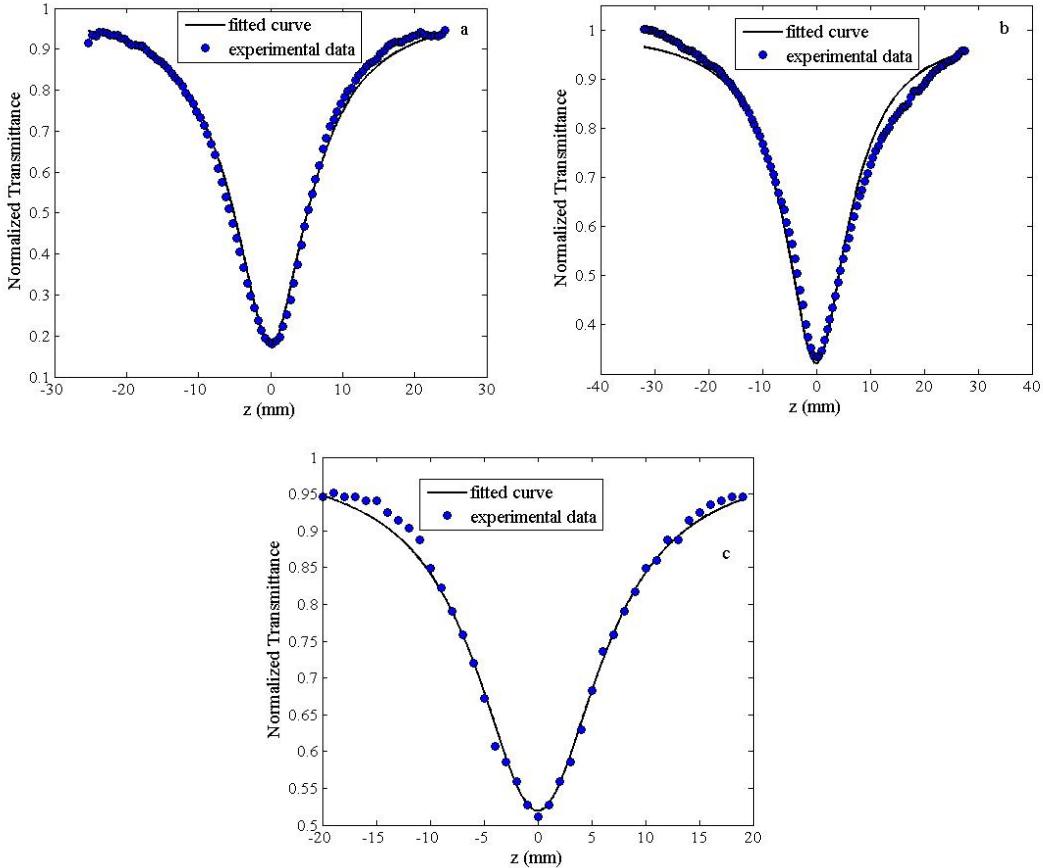
Fig. 2. Open aperture Z-scan experimental data obtained for Ag-PVP nano-fluid under 532nm excitation for sample concentration of 4.71×10^{-3} M radiated with a) 40 kGy b)20 kGy c) 10 kGy. The solid lines show the fitting lines for TPA.

3.2. Optical limiting

It is well known that TPA induced the optical limiting phenomenon which can be described theoretically by the following relation [22, 23]:

$$\frac{I_i}{I_0} = e^{\alpha_0 L} + \frac{(e^{\alpha_0 L} - 1)\beta I_i}{\pi \omega_0^2 \alpha_0} \qquad (2)$$

where I_i is the incident power, I_o is the output power and L is the thickness of the sample. Fig. 3 shows the measured transmitted power as a function of the incident power of Ag nanoparticles at a concentration of 4.71×10^{-3} M for three particles sizes; 32.3, 38.5 and 41.8 nm. At low power input I_i, the output power I_o, increases rapidly in the linear region then the output power deviates slowly from the linear indicated the nonlinear phenomenon. The experimental data show that the smaller particle size, the linear range becomes smaller. Obviously in work the Beer's law only applied for the input power less than 0.35, 0.45 and 0.5 mW for samples S1, S2 and S3 respectively. By fitting the experimental data to the equation the nonlinear absorption coefficient of samples S1, S2 and S3 due two-photon absorption process were obtained as

5.3×10^{-3}, 4.8×10^{-3} and 3.5×10^{-3} cm/W. These values agree well with the values obtained by measurements. Thus, it confirmed that the nonlinear absorption coefficient of our sample measured using method was attributed by to two-photon absorption processes.
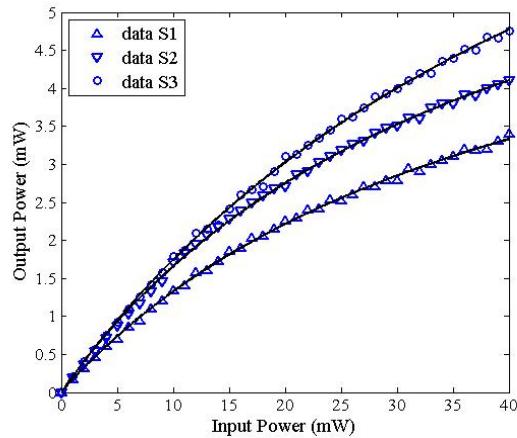
Fig. 3.JPG Measured transmitted power as a function of the incident power (at 532 nm) for nano-fluid samples S1, S2 and S3. The solid curves are fitted with Eq. (2).

4. Conclusion

We have studied two-photon absorption of silver nano-fluid prepared by γ radiation method. The Z-scan technique using 532 nm CW laser excitation source was used to measure the nonlinearity of absorption from two-photon absorption processes. Three samples of nanoparticles at different particle sizes were measured and the nonlinear absorption coefficients obtained were 5.8×10^{-3}, 4.5×10^{-3} and 3.2×10^{-3} cm/W. These values agreed very well with the values obtained from optical limiting experiments. In this work we observed that by increasing the radiation dose it make the particle size smaller but produced higher nonlinear absorption coefficient. This results show that the particle size plays important role in two-photon absorption processes. Since the particle size can be controlled by adjusting the irradiation dose this type sample has a potential for applications in nonlinear optical devices.

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[*]Corresponding author: esmaeil.phy@gmail.com