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# Optical Nonlinearity Properties of Ag Nanofluid Measurements using a Single Beam Z-scan Technique

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**Abstract:** Nonlinear refractive index and nonlinear absorption of Ag nano-fluid prepared by γ-radiation method were investigated using a single beam Z-scan technique. Under a CW laser beam excitation operated at 532 nm with the power output of 40 mW, the Ag nano-fluid shows a large thermal-induced nonlinear refractive index and nonlinearity absorption. We measured the nonlinear refraction coefficient of silver nano-fluid for concentrations, 1.170 x $10^{-3}$ and 5.297 x $10^{-3}$ M. respectively. We found the nonlinear refractive index is in the order of $10^{-8}$ $cm^2/W$ with the negative sign indicates the self-defocusing phenomenon. However for the Ag nano-fluid sample with 1.170 x $10^{-3}$ M, the result shows that the nano-fluid sample has only a nonlinear refractive index property.

**Key words:** Self-defocusing, Nanoparticles, Nonlinear refractive index, Thermal effects

## INTRODUCTION

The field of nonlinear optics has enlarged a new frontier in science and technology. Metal nano-fluids has demonstrated a vast range of applications such as, the labeling of biological molecules, surface enhanced Raman scattering, optical limiter and optical photonics devices (Sheik-Bahae *et al*. 1990; Liao *et al*. 1997; Sutherland, 1996). Z-scan method is one of the simple and accurate methods for measuring the nonlinear optical properties such as nonlinear refraction and nonlinear absorption of solid and liquid samples (Yang *et al*. 2002; Marder *et al*. 2004; Ganeev *et al*. 1997). This method also provides the magnitudes of real and imaginary part of nonlinear susceptibility and the sign of the real part can be simultaneously determined (Mukherjee, 1993; He *et al*. 1997, 2003; Sheik-Bahaei *et al*. 1989).

In 1990, Sheik-Bahae *et al*. presented a method of studying the nonlinear absorption (NLA) and nonlinear refraction (NLR) of materials using a single laser beam (Sheik-Bahae *et al*. 1990). This method, known as Z-scan, utilizes a tightly focused laser beam that is intense enough to access nonlinearities in a sample. As the sample passes through the focal point of the beam, changes in its transmittance for nonlinear absorption (NLA) and nonlinear refraction (NLR) are measured using an open aperture configuration and closed aperture configuration, respectively. In the open aperture technique, after the beam passed through the sample, it is focused directly into a detector. As the sample travels through the focus of the beam, the transmittance either increases or decreases (depending on the nonlinearity of the sample) and the detector receives more or less light than the linear transmittance, yielding a hump or dip in the curve of transmittance as a function sample position. For the case of nonlinear refraction, after passing through the sample, the beam is attenuated by a semi-closed aperture that usually allows about 30% of the beam to pass through to the detector. With this, due to converging and diverging of the beam (allowing more and less of the beam to pass through the aperture, respectively) with changes in the refractive index, a pre-focal valley and post-focal peak are observed for a positive change in refraction and a pre-focal peak and a post-focal valley is observed for a negative change in refraction.

In the present work we report the nonlinear refractive index and nonlinear absorption coefficient of Ag nano-fluids prepared using γ ($^{60}$Co-rays) radiation at 50 kGy level.

### *Experimental:*

For preparing Ag nano-fluid sample at 1.170 x $10^{-3}$ M, 10 mg of silver nitrate (AgNO3 Aldrish-99%), 3 g of polyvinylpyrrolidone (PVP, MW 29,000 Aldrish) and 1 ml isopropanol were used. The PVP and isopropanol were used as a colloidal stabilizer and radical scavenger of hydroxyl radical respectively. The solution was stirred for 2 hours and was bubbled with nitrogen gas (99.5%) in order to remove oxygen. Silver

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nitrate ($AgNO3$) was added into PVP solution and isopropanol, which acted as a hydroxyl radical scavenger. Samples were then irradiated with γ-radiation at a dose of 50 kGy. In this process, γ-irradiation produces hydrated electrons that reduce the silver ions to silver atoms, which then aggregated in the solution. The γ-radiation ($^{60}$Co-rays) source is an effective tool for polymerization process and reducing agent. The similar method was applied for preparation of high concentration of Ag nano-fluid (5.297 x $10^{-3}$ M).

The average particle diameters of 1.170 x 10 $^{-3}$ and 5.297 x $10^{-3}$ M nanofluid samples were measured using nanophox machine (Sympatec GmbH, D-38678) and the average sizes were recorded as 17.9 nm and 41.6 nm, respectively. The linear absorption spectrum for sample with 1.170 x 10 $^{-3}$ M concentration was measured using UV-Vis spectrophotometer (Shimadzu-UV1650PC) but for sample with higher concentration the linear absorption coefficient was measured using Z-scan set up with 2 mm sample cell. For this case the transmission signal was measured in the linear regime of the experiment and the absorption coefficient was calculated using simple equation as $\alpha = -(1/L)Ln(I_O / I)$.

Fig. 1 shows the schematic diagram of a single beam Z-scan experiment used in the present measurement. The experiments were performed using a CW beam diode laser operated at 532 nm wavelength (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. At the focus point the power output of the laser beam measured was 40 mW. The transmitted light at the far field passed through the aperture and the beam intensity was recorded by a photodiode detector, D. The laser beam waist $w_0$ at the focus length was measured to be 24.4 µm and the Rayleigh length was found to satisfy the basic criteria of a Z-scan experiment. A quartz optical cell containing specimen solution was translated across the focal region along the z-axis direction.

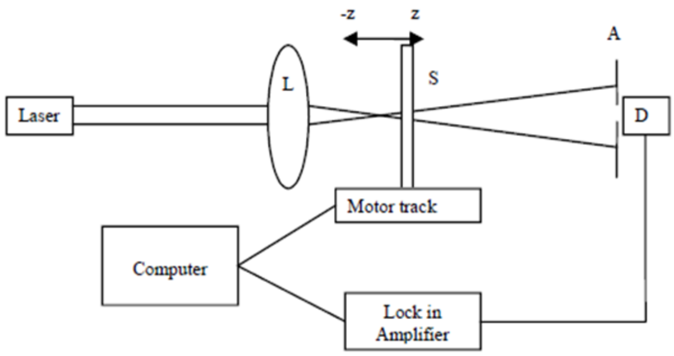


**Fig. 1:** Schematic diagram of a single beam Z-scan experiment setup: L, Lens; S, Sample; A, Aperture; D, Detector

## RESULTS AND DISCUSSION

Fig. 2 shows typical absorption spectrum of Ag nano-fluid with the surface plasmon absorption peak located at 413 nm, while Fig. 3 shows the transmittance curves obtained for the same sample using 532 nm laser excitation beam. The laser beam intensity, $I_0$ was measured to be $4.27\times10^3$ W/cm$^2$ and the aperture linear transmittance was 0.15. The peak-valley curve indicates that the self-defocusing effect and nonlinear refractive index of the medium is negative. The third order nonlinear refractive index, $n_2$ was calculated using expression reported by Sheik-Bahae *et al.*(1990), Moran *et al.* (1975) and Brueck *et al.* (1980) i.e.

$$n_2 = \frac{\Delta T_{p-v}}{0.406(1-s)^{0.25} k L_{eff} I_o} \tag{1}$$

where $k = 2\pi / \lambda$ . $I_0$ is the beam intensity at focus point, s is the aperture linear transmittance, $L_{eff} = \left(1-\exp(-\alpha L)\right)/\alpha$ is the effective thickness of the sample and $\Delta T_{p-v}$ is measured from the experimental data of normalized peak to valley transmittance. In the present work the linear absorption coefficients, $\alpha$ for two samples were obtained from the absorption spectra of Fig. 2 and linear regime measurement as described in experimental section.

The solid line in Fig.3 is the calculated values using analytical equation proposed by Liao *et al*. (1997) and Dmitrity *et al*. (1999).

$$T(z,\Delta\phi)=1-\frac{4\Delta\phi_o x}{(x^2+1)(x^2+9)} \quad (2)$$

The curves calculated using Equation (2) is generally in good agreement with the experimental data obtained for Ag nano-fluid sample. Using the values of $I_0$ and s measured in the present work, we obtained the value of nonlinear refractive index $n_2$ for Ag nano-fluid sample as $-4.180\times10^{-8}$ $cm^2/W$. However no nonlinear absorption phenomenon occurs for this sample. Knowing $n_2$, the change in nonlinear refractive index at the focus can be calculated as $\Delta n_0 = n_2 I_0$ where $I_0$ being the on-axis irradiance at the focus. This gives the values of $\Delta n_0$ for sample S1 as $-1.768\times10^{-4}$.

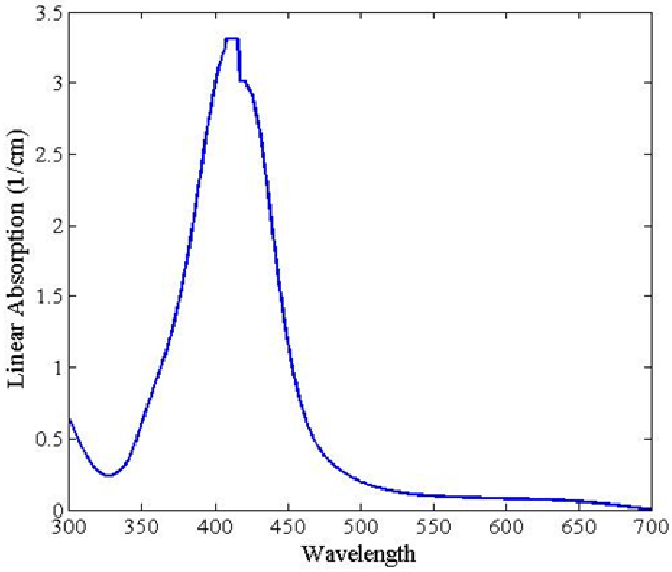


**Fig. 2:** Absorption spectrum of Ag nano-fluid at concentration of $1.170\times10^{-3}$ M. The average particle size is 17.9 nm

Figure.4 shows the open aperture Z-scan experimental data of Ag nanoparticle obtained for Ag nano fluid with concentration of $5.297\times10^{-3}$ M (S2). The solid line is theoretical fitting by considering the two-photon absorption process calculated using a well know normalized transmittance for the open aperture Z-scan given as:

$$T = q_0^{-1}(z)\ln(1+q_0(z)),\ |q_0(z)|<1 \quad (3)$$

where $q_0(z)=I_0\ \beta\ L_{eff}/(1+z^2/z_0^2)$ . The nonlinear absorption coefficient obtained by fitting the experimental data to Equation 3 was $6.91\times10^{-3}$ cm/W, while the nonlinear refraction coefficient obtained from closed aperture measurement was $-5.85\times10^{-8}$ $cm^2/W$, which is higher than the value obtained for sample S1.

The values of $n_2$ and β for both samples (S1 and S2) are listed in Table 1. In the present work, for low concentration sample (S1) and small particle size the result shows that there is nonlinear absorption phenomenon occur. The nonlinear behavior of the medium was only described by nonlinear refractive index value of $-4.180\times10^{-8}$ ($cm^2/W$). However for high concentration sample and larger particle size (S2), the nonlinear absorption and nonlinear refractive index obtained were $6.91\times10^{-3}$(cm/W) and $-5.85\times10^{-8}$ ($cm^2/W$), respectively. In this study, it is shown that the sample concentration and particle size are the main parameters that affect the thermal-induced nonlinearity phenomenon in liquid. We have displayed that the nano-particle size will give a significant effect to the nonlinearity properties such as nonlinear refractive index and nonlinear absorption. Thus sample preparation technique will be an important aspect in studying nonlinear optical properties of metal nano-particle solution. Since the γ- radiation technique can control the particle size of metal nano-particle in solution, a specific particle size can be produced for specific nonlinear optical devices. This will offer a further nonlinearity study in metal nano-fluids field.

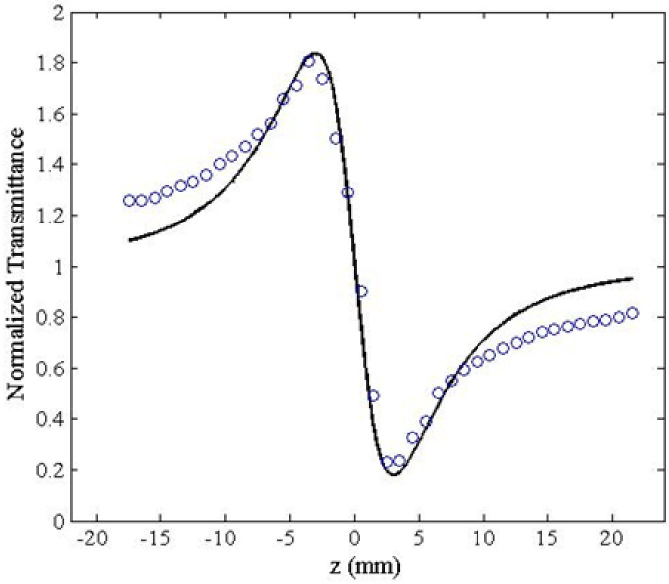


**Fig. 3:** Closed aperture Z-scan curve for Ag nanoparticles measured at a concentration of $1.17\times10^{-3}$ M irradiated at 50 kGy. Solid line is the fitted curve using Equation (2)

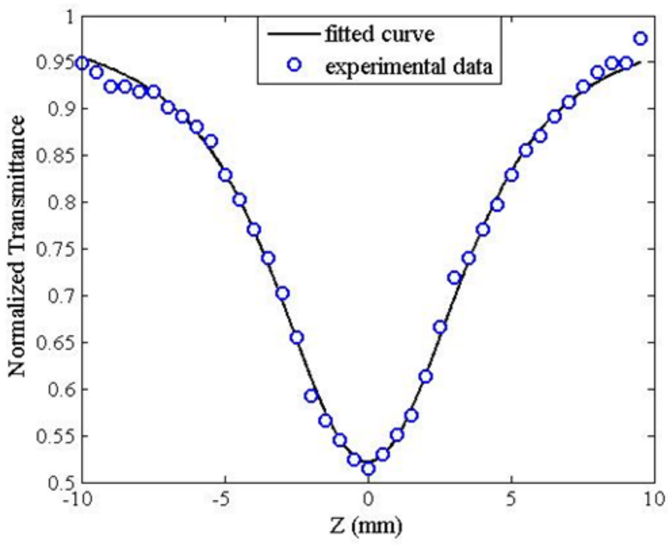


**Fig. 4:** Closed aperture Z-scan experimental data for Ag nano-fluid measured at concentration $5.297\times10^{-3}$ M. The solid line is the theoretical curve calculated using Equation (3)

**Table 1:** Nonlinear refractive index and nonlinear absorption coefficient of Ag nano-fluids prepared by γ radiation technique

| sample | S1 | S2 |
|---|---|---|
| Concentration (M) | $1.170\times10^{-3}$ | $5.297\times10^{-3}$ |
| Particle size (nm) | 17.9 | 41.6 |
| $n_2$ ($cm^2$/W) | $-4.180\times10^{-8}$ | $-5.85\times10^{-8}$ |
| β (cm/W) | - | $6.91\times10^{-3}$ |

***Conclusions:***

The third-order nonlinear refraction and two-photon absorption coefficient were measured for the Ag nano-fluid nanoparticles using a single bean Z-scan technique. The values of nonlinear refractive index, $n_2$ and nonlinear absorption coefficient for Ag nano-fluid measured at 532 nm laser excitation were in order of $10^{-8}$ $cm^2/W$ and $10^{-3}$ cm/W, respectively. The experiment also confirmed that the nonlinear phenomenon was caused by self-defocusing process. These results show that the Ag solution nanoparticles are promising materials for optical devices.

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