

Single Beam Z-Scan Measurements of Nonlinear Refraction and Nonlinear Absorption Coefficients in Silver Nano-Fluid

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Abstract: Problem statement: The nonlinear refractive index and nonlinear absorption are two important optical phenomena which are extensively used in optical switching, optical limiting and labeling. **Approach:** In the present research we had measured nonlinearity of Ag nano-fluid prepared using γ -irradiation method. The measurement was carried out using a single beam z-scan technique. Under a CW laser beam excitation operated at 532 nm with the power output of 40 mW, a closed aperture setup was used for thermal-induced nonlinear refractive index and an open aperture setup was applied for measuring nonlinearity absorption. **Results:** We measured the nonlinear refraction coefficient and nonlinear absorption coefficients for silver nano-fluid at concentration 6.475×10^{-3} M, the values obtained were $-6.173 \times 10^{-8} \text{ cm}^2 \text{ W}^{-1}$ and $7.994 \times 10^{-3} \text{ cm W}^{-1}$ respectively. **Conclusion:** The nonlinear refraction coefficient with the negative sign indicates the self-defocusing phenomenon. The experimental data from open aperture measurement showed that a two photon absorption phenomenon. These results showed that the Ag nano-fluid has significant values of nonlinear refractive index and nonlinear absorption, thus it could be good candidate for optical devices.

Key words: Self-defocusing, nonlinear optics, nano-metal, thermal effects

INTRODUCTION

The single beam Z-scan technique was developed by the Sheik-Bahae *et al.* (1989). This method is a simple and effective tool for determining the nonlinear properties. It has been used widely in material characterization because it provides not only the magnitudes of the real and imaginary parts of the nonlinear susceptibility, but also the sign of the real part (Sutherland *et al.*, 2003; Li *et al.*, 1994; Marder and Torruellas, 1997; He *et al.*, 1997). This method, 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 due to Nonlinear Absorption (NLA) and Nonlinear Refraction (NLR) are measured by an open aperture and closed aperture, respectively. In the open aperture technique, after the beam is passed through the sample, it is focused directly into a detector. As the sample travels through the focus of the initial 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 of sample position. For NLR, after the beam passing through the sample, it is attenuated by a semi-closed aperture that usually allows about 30% of the initial beam to be detected by 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) made changes in the refractive index. A pre-focal valley and post-focal peak is 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.

There has been a large necessity for nonlinear optical materials that can be used with low intensity lasers for applications such as phase conjugation, image processing and optical switching (Kramer *et al.*, 1986; Hernandez *et al.*, 1998; Yin *et al.*, 2000 Ryasnyansky and Palpant, 2005). 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 (Kovsh and Yang, 1999; Kovsh and Yang, 1999). In this study, we report the nonlinear refractive index and nonlinear absorption coefficient of Ag nano-fluid prepared using γ (^{60}Co -rays) radiation at 50 kGy level.

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MATERIALS AND METHOD

To prepare Ag nano-fluid sample, 55 mg of silver nitrate (AgNO_3 Aldrich-99%), 4 g of polyvinylpyrrolidone (PVP, MW 29,000 Aldrich) and 1 mL isopropanol were used. The PVP and isopropanol were used as a colloidal stabilizer and radical scavenger of hydroxyl radical respectively. The PVP solution was made by dissolving PVP powder in 50 ml of deionized water at room temperature. The solution was stirred for 2 h and was bubbled with nitrogen gas (99.5%) for removing the oxygen gas. The γ -radiation (^{60}Co -rays) source is an effective tool for polymerization process and reducing agent. Silver nitrate (AgNO_3) was added into PVP solution and the concentration of Ag nanoparticles in solution was calculated to be 6.475×10^{-3} M. Sample was 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 average diameter of Ag nanoparticles was measured using Nanophox Machine (Sympatec GmbH, D-38678) and the particle average size was recorded as 42.3 nm. The linear absorption spectra Ag nano-fluid sample was measured by conventional method on the basis of $\alpha = -(1/L)\ln(I_0/I)$ in the linear regime of the experiment.

Figure 1 shows the experimental set up of a single beam Z-scan experiment used in the present measurement. The experiment was carried out at room temperature 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 was measured to be 40 mW.

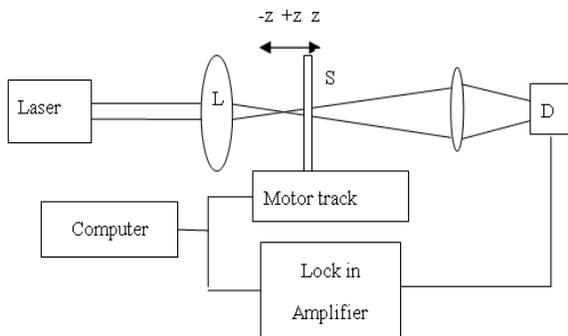


Fig. 1: Schematic diagram of a single beam Z-scan experiment setup: L: Lens; S: Sample; A: Aperture; D: Detector

The transmitted light in the far field passed through the aperture and the beam intensity was recorded by a photodiode detector, D. The laser beam waist ω_0 at the focus length was measured to be $24.4 \mu\text{m}$ 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.

RESULTS

Figure 2 shows the open aperture z-scan experimental data obtained for Ag nano-fluid with concentration of 6.475×10^{-3} M irradiated at 50 kGy. The nonlinear absorption coefficient β can be obtained from this open aperture Z-scan data by fitting the normalized transmittance data to the open aperture formula given as (Sheik-Bahae *et al.*, 1989; Sheik-Bahae *et al.*, 1990):

$$T(z, s = 1) = \sum_{m=0}^{\infty} \sum_{m=0}^{\infty} \frac{[-q_0(z)]^m}{(m+1)^{3/2}} |q_0(z)| < 1 \quad (1)$$

Where:

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

$z_0 = k\omega_0^2/2$ is the diffraction length of the beam

$k = 2\pi/\lambda$ is the wave factor

$\omega_0 =$ The beam waist radius at the focal point

$L_{\text{eff}} = (1 - \exp(-\alpha L))/\alpha$ is the effective thickness of the sample

The solid line in Fig. 2 is theoretical curve while the symbol is the experimental data. The nonlinear absorption coefficient obtained was $7.994 \times 10^{-3} \text{ cm W}^{-1}$.

The nonlinear refractive index can be calculated from the normalized transmittance data of closed aperture measurement which can be written as follows (Sheik-Bahae *et al.*, 1990):

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

where, $\Delta\phi_0$ is the on axis phase change at the focus point. This produces a symmetry ΔT_{p-v} (peak-valley) curve where the nonlinear refractive index, n_2 can be calculated using simple expression reported by Sheik-Bahae *et al.* (1989); Sheik-Bahae *et al.* (1990) that is:

$$n_2 = \frac{\Delta T_{p-v}}{0.406(1-s)^{0.25} k L_{\text{eff}} I_0} \quad (3)$$

where, s is the aperture in linear transmittance regime.

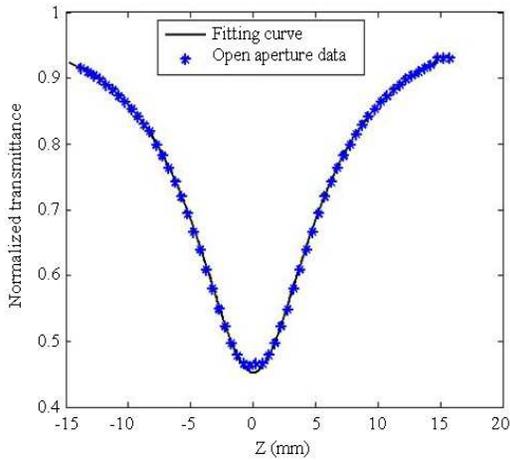


Fig. 2: Open aperture Z-scan curve for Ag nanoparticle measured at a concentration of 6.475×10^{-3} M irradiated at 50 kGy. Solid line is the fitted curve using Eq. 1

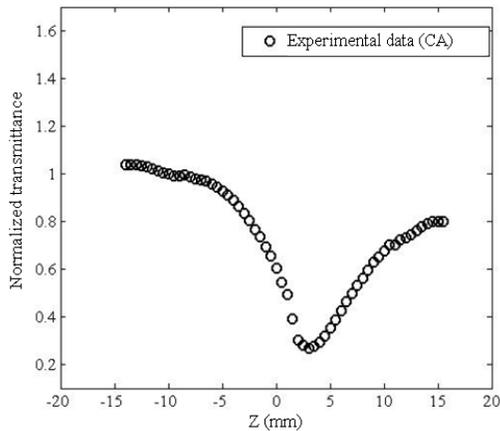


Fig. 3: Closed aperture experimental data of Ag nano-fluid measured at concentration of 6.475×10^{-3} M. The average particle size is 42.3 nm

Figure 3 shows the transmittance curves of a closed aperture z-scan obtained for Ag nano-fluid. The laser intensity measured was $I_0 = 4.27 \times 10^3 \text{ W cm}^{-2}$ and the linear aperture transmittance was 0.10. The curve is obviously asymmetry where there is a large suppressed in the transmittance peak and enhanced in the valley.

DISCUSSION

This behavior was due to the high nonlinear absorption phenomenon of the sample and can be solved by dividing the closed aperture (CA) experimental data of Fig. 3 over the open aperture (OA) experimental data of Fig. 2.

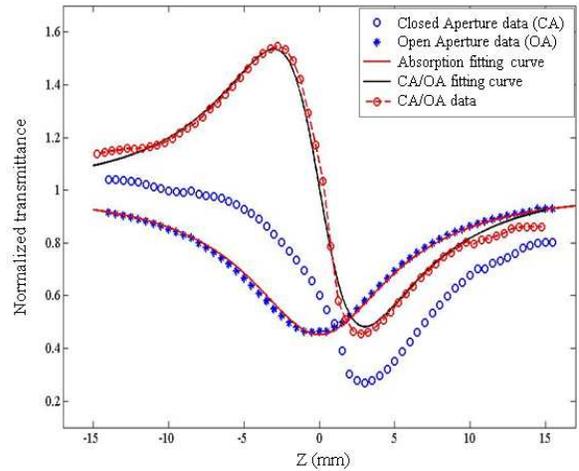


Fig. 4: Closed aperture Z-scan experimental data for Ag nano-fluid measured at concentration 6.475×10^{-3} M. The solid line is the theoretical curve calculated using Eq. 1 and 2 respectively

Figure 4 shows a perfect symmetry experimental curve was produced by dividing the closed aperture to open aperture experimental data of Fig. 3 and 2. By measuring the ΔT_{p-v} from CA/OA fitted curve in Fig. 4 we obtained the third order nonlinear refractive index, n_2 as $-6.173 \times 10^{-8} \text{ cm}^2 \text{ W}^{-1}$. The negative sign confirms the nonlinear phenomenon was due to the self-defocusing process.

This result shows that the Ag nano-fluid with the particle size of 42.3 nm gives significant values of 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 easily 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 for specific applications.

CONCLUSION

The third-order nonlinear refraction and two-photon absorption coefficient were measured for the Ag nano-fluid using a single beam 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 $-6.173 \times 10^{-8} \text{ cm}^2 \text{ W}^{-1}$ and $7.994 \times 10^{-3} \text{ cm W}^{-1}$ respectively. The experiment also confirmed that the nonlinear phenomenon was caused by self-defocusing process. These results show that the

Ag nano-fluid with a particle size of 42.3 nm is a promising material for optical devices.

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