



## Full length article

# Coherent source interaction, third-order nonlinear response of synthesized PEG coated magnetite nanoparticles in polyethylene glycol and its application



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## ABSTRACT

Third-order nonlinear response of synthesized polyethylene glycol coated  $\text{Fe}_3\text{O}_4$  nanoparticles dispersed in a suitable solvent, polyethylene glycol has been studied. The structural characterization of the synthesized magnetite nanoparticles were carried out. The linear optical property of the synthesized magnetite nanoparticles was investigated using UV-visible technique. Both closed and open aperture Z-scan techniques have been performed at 532 nm with pulse width 5 ns and repetition rate 10 Hz. It was found that polyethylene glycol coated magnetite exhibits reverse saturable absorption, with significant nonlinear absorption coefficient. Two-photon absorption intensity dependent positive nonlinear refraction coefficients indicate self focusing phenomena. Results show that higher concentration gives better nonlinear and optical limiting properties.

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## 1. Introduction

The nonlinear optical properties of different metallic nanoparticles have been investigated extensively [1–4]. They have been encouraged as they exhibit a stronger and faster optical response, good control over shape and easy fabrication, durability and thermo mechanical stability [5]. The change in transmittance of a medium as a function of intensity of the incoming light or fluence, which evoke the nonlinear terms in the dipole oscillation, modifies the properties of medium is referred to as nonlinear absorption or nonlinear transmission of the medium. Especially the third-order optical response described by the  $\chi^{(3)}$  susceptibility, is responsible for effects such as third harmonics generation, self focusing, Raman scattering, Brillouin scattering and phase conjugation. Usually the refractive index (RI) of an optical material is expressed as,  $n = n_0 + n_2 I$ , where  $n_0$  represents the linear refractive index of a weak field. 'I' is the intensity of exposed light and ' $n_2$ ' is the nonlinear refractive index. Also the intensity dependent absorption coefficient is defined as  $\alpha(I) = \alpha_0 + \beta I$ , ' $\alpha_0$ ' and ' $\beta$ ' represents the linear and nonlinear absorption respectively. The real part of  $\chi^{(3)}$  is proportional to  $n_2$  (nonlinear refractive index, RI) and imaginary part is related to nonlinear absorption [6].

There are different types of experimental setup to measure the optical nonlinearity of materials. Among them Z-scan technique, is a simple and highly sensitive single beam method, that works on spatial beam distortion in the medium to measure both the sign and magnitude of the third-order optical nonlinearity [7]. Materials exhibiting nonlinear optical properties were differentiated depending on mechanisms like saturable absorption (SA) or reverse saturable absorption (RSA). Ferrofluid have been widely used in loud-speaker coils and pressure sensors. However, only a very few study were reported for ferrofluid so far in nonlinear optic field. Magnetic nano materials have good nonlinear optical absorption characteristics [8]. This interest has been motivated for large applications such as synthesis of material nano composites [9] for data storage [10], spintronics [11], magnetic resonance imaging (MRI) [12] and other bio medical applications [13].

The present study investigates the third-order nonlinear optical properties of ferrofluid, in which magnetite nanoparticles were suspended in polyethylene glycol (PEG), which is a suitable base fluid [14]. The study also lines out the open and closed aperture Z-scan of the magnetite-PEG composite.

Nanomaterials have invited significant attention as optical limiters. Optical limiting (OL) property can be analysed by the open aperture nonlinear optical processes of nanomaterials [15]. For a material to be an ideal optical limiter, it should be transparent to low energy laser pulse while opaque at high energies. It can protect eyes and optical sensors from the intense laser radiation. The present study also checks the possibility of the sample under study to

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be used as an optical limiter. Ferrofluid are also widely used, due to their very high saturation magnetization, good thermal stability and are also free from agglomeration [16]. The physical and chemical stability of ferrofluid, which is an inevitable attribute for an optical limiter, encourage us to investigate their optical limiting properties with change in concentration of the sample in PEG [17–19].

## 2. Experimental

PEG coated  $\text{Fe}_3\text{O}_4$  nanoparticles were synthesized using chemical coprecipitation method [20]. The structural analysis was carried out by X-ray diffraction technique (XRD), transmission electron microscopy (TEM) and high resolution transmission electron microscopy (HRTEM). The XRD patterns of the present sample were recorded by X-ray diffractometer (3 kW X'pert PRO X-ray diffractometer) using  $\text{Cu K}\alpha$  radiation ( $\lambda = 0.1540 \text{ nm}$ ) to determine the crystal structure of the samples. The particle size and morphology was confirmed by TEM (Jeol/JEM 2100). HRTEM was performed to confirm the phase of synthesized  $\text{Fe}_3\text{O}_4$  nanoparticles. The linear optical property of synthesized magnetite nanoparticles were analysed by UV-vis absorption spectra (Perkin-Elmer Lambda-34). To understand the interaction between PEG and Iron oxide nanoparticles by FT-IR Spectrometer (Perkin Elmer) is used. The presence of PEG over the magnetite nanoparticles was confirmed by Thermogravimetry - Differential thermal analysis (TG-DTA) using Perkin Elmer Diamond TG/DTA.

The third-order nonlinear optical measurements of the sample were obtained from Z-scan technique [21] with the aid of neodymium-doped yttrium aluminium garnet (Nd:YAG) pulsed laser ( $\lambda = 532 \text{ nm}$ ) with pulse width 5 ns and repetition rate 10 Hz. The Z-scan method implemented in this study is Sheikh-Bahae et al. [22]. The experimental arrangement consists of a lens, sample and a large area detector. An aperture is used at the output of the laser to obtain a smooth profile in the far field. Energy after the aperture can be varied from 1 to 2 mJ/pulse. 1mm quartz cuvettes were used for exposing the sample. The whole Z-scan set up is shown in the Fig. 1. The beam from the laser source is focused on the sample, by means of a convex lens of focal length 100 mm, leading to a measured beam waist of  $35.5 \mu\text{m}$ . Each point is average of 4 pulses. PEG coated magnetite nanoparticles at different concentration in PEG such as 1 mM, 2 mM and 3 mM of solution have been prepared. The solution is then subjected to ultrasonification, overnight so as to obtain magnetite-PEG colloidal solution for the investigation of their nonlinear optical response. The transmitted beam energy, the reference beam energy, and ratio of them were measured using an energy ratiometer.

The closed and open aperture Z-scan technique is used to investigate the nonlinear optical property of the sample under study. The closed aperture setup yields the nonlinear refractive index ( $n_2$ ) and open aperture yields the nonlinear absorption coefficient ( $\beta$ ), measured after removing the aperture

## 3. Results and discussion

### 3.1. X-ray diffraction study

The crystallographic analysis of  $\text{Fe}_3\text{O}_4$  nanoparticles were performed by powder X-ray diffraction (XRD). A continuous scan mode was used to collect  $2\theta$  ranging from  $10^\circ \text{C}$  to  $70^\circ \text{C}$  (Fig. 2). The XRD pattern displays nine characteristic peaks, (220), (311), (222), (400), (422), (511), (440), (620) and (533) corresponding to,  $2\theta = 30.0^\circ \text{C}$ ,  $35.4^\circ \text{C}$ ,  $37.06^\circ \text{C}$ ,  $43.1^\circ \text{C}$ ,  $53.6^\circ \text{C}$ ,  $56.9^\circ \text{C}$ ,  $62.5^\circ \text{C}$ ,  $65.7^\circ \text{C}$ ,  $70.9^\circ \text{C}$  and  $74.1^\circ \text{C}$  respectively [23] indexed in Fig. 2 have been compared to the JCPDS file (PDF #89-2355). The pattern is found identical to the peaks of  $\text{Fe}_3\text{O}_4$  crystalline cubic spinel structure. No other peaks corresponding to other phases of  $\text{Fe}_3\text{O}_4$  are found which shows the mono phase of magnetite. The strong and sharp peaks reveal the high purity and crystalline nature of  $\text{Fe}_3\text{O}_4$ .

The average particle size is calculated by Scherer's formula.  $D = 0.9\lambda/\beta\cos\theta$ , where  $\lambda$  is the wavelength used,  $\beta$  is the full width half maximum (FWHM) and  $D$  is the grain size. From the measured line broadening, the average grain size is found to be 26 nm. The lattice parameter, 'a' is found to be 8.3 Å.

### 3.2. Transmission electron microscopy analysis

TEM images and high resolution transmission electron microscopy (HRTEM) show the nanostructure of the synthesized PEG coated  $\text{Fe}_3\text{O}_4$  nanoparticles, (Fig. 3). Nanoparticles are found to be in polyhedral shape, with a little agglomeration [24] (Fig. 3 (a) and (b)). Information supporting the structure and phase purity of the nanoparticles was obtained through the detailed examination of high resolution TEM (Fig. 3(d)). The interplanar distance 4.8 Å was assigned to the crystallographic plane of Miller indices (111) [JCPDS standard file PDF #89-2355], which is the characteristic peak of  $\text{Fe}_3\text{O}_4$  cubic inverse spinel structure, supported by XRD pattern. The particle size distribution of magnetite nanoparticles is embedded Fig. 3(d), by counting 120 particles and was found to be  $23 \pm 3 \text{ nm}$ , which is in good agreement with the XRD result. A slight coating of polyethylene glycol can be viewed over the synthesized magnetite nanoparticles (Fig. 3(c)).

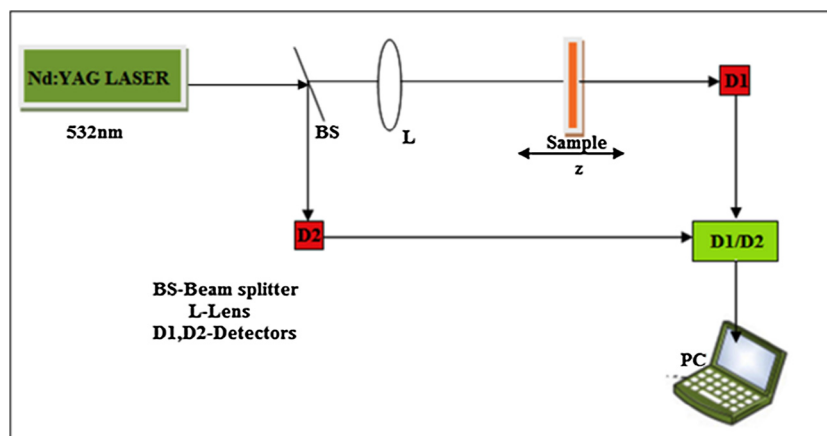


Fig. 1. Experimental set up of Z-scan technique.

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