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Nonlinear optical absorption studies of sol–gel derived Yttrium Iron Garnet ($Y_3Fe_5O_{12}$) nanoparticles by Z-scan technique

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Abstract

Sol–gel synthesized Yttrium Iron Garnet ($Y_3Fe_5O_{12}$) nanoparticles were subjected to open aperture Z-scan studies in order to investigate the nonlinear optical (NLO) properties of these materials. The investigations were carried out using a Q-switched resonant Nd:YAG laser at a wavelength of 532 nm with different laser powers. Strong reverse saturable absorption (RSA) has been found when the sample is irradiated by the laser pulse of 10 Hz. The studies show that the material is highly nonlinear, which makes it useful for optical limiting applications.

Keywords: A. Sol–gel process; B. X-ray methods; C. Optical properties

1. Introduction

Nonlinear optics is attracting much interest because of their potential use in signal processing, data storage, image transmission and optical switching [1–3]. Advancement of research in this field depends on the development of new materials with strong nonlinear optical effects. Although several experimental techniques have been used to determine the nonlinear optical properties of materials, it is difficult to accurately measure the nonlinear optical parameters. In that respect, the Z-scan technique has been a very important technique to measure the nonlinearity because of its high sensitivity and simplicity [4]. Rare-earth iron garnets have been intensely studied in recent years, particularly in the light of magneto-optical storage and optoelectronics applications. Yttrium Iron Garnet (YIG) has been shown to increase the inductance in a device significantly. The optical properties of YIG are well studied on bulk single crystals and epitaxial films both in the fundamental absorption region $E > 2.6$ eV and in the region of a relatively high transparency (0.15–1.24 eV) [5–8]. The crystal lattice of the compound has two nonequivalent (octa and tetrahedral) positions occupied by Fe^{3+} ions in the ratio of 2:3; for this reason, the optical spectrum is complex. Many methods have been used to prepare $Y_3Fe_5O_{12}$ material. The conventional way of producing this material is by solid state reaction with oxide/carbonate and calcined at high temperature [9]. When YIG is prepared by ceramic methods, $YFeO_3$ and Fe_2O_3 are produced as intermediates and these phases remain as impurities unless heated to high temperatures [10,11]. Therefore, sol–gel methods have attracted the attention due to the lower synthesis temperature and finer and more homogeneous particles produced [12,13]. Most of the reported works have been devoted to the electrical and ferrimagnetic properties of YIG materials. However a survey of literature reveals that not much information exists on the nonlinear optical (NLO) properties of YIG nanomaterial. In the present work, the optical nonlinearities in YIG nanoparticles synthesized through sol–gel process have been studied using the Z-scan technique.

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2. Experimental procedure

The garnet nanoparticles were prepared by novel sol–gel technique. The starting solution was stoichiometric mixtures of iron nitrate $Fe(NO_3)_3\ 9H_2O$ and yttrium nitrate $Y(NO_3)_3\ 6H_2O$ dissolved in an aqueous solution of PVA. The PVA used here act as a chelating agent in making the gel. The mixtures were stirred continuously, at a temperature of 60 °C until the formation of a gel was observed. The gel was then dried in an oven for another 24 h at 110 °C. The dry gels were then heat treated in air at 800 °C for 3 h. The crystalline nature of the sample was characterized using Bruker AXS 5005 X-ray diffractometer. The morphology of the as prepared sample was investigated using a scanning electron microscope (JEOL Model JSM-6390LV).

The X-ray diffraction patterns of YIG nanoparticle synthesized by sol–gel method are shown in Fig. 1. The diffraction peaks of (4 0 0), (4 2 0) and (4 2 2) associated to garnet structure can be clearly observed (JCPDS card no. 43-0507). On the other hand, a broadening of the diffraction peaks is detected due to the nanoscale grain size. The Debye–Scherrer formula was used to calculate the average grain size of the sample and to be 61 nm.

The morphology of YIG powders was revealed by SEM as shown in Fig. 2. The micrograph exhibits inhomogeneous grain size distribution of small particles (<200 nm) and large agglomerate particles (∼200 nm–2 μm). It is noted that the grain size estimated by SEM are larger than those obtained from Scherrer's formula. This is could be attributed to the agglomeration of nanoparticles in the powder sample. The grain sizes for YIG sample prepared via sol–gel technique are found to be less than those prepared via conventional method, which is reported to be in the range 1–10 μm [14].

The open aperture Z-scan experiment was used to measure the nonlinear transmission of the powder samples suspended in ethanol. The sample is excited using a pulsed Nd:YAG laser (Quantaray GCR170) at a wave length of 532 nm, with pulse energy of 450 mJ. The pulse width is 6–7 ns and the pulse repetition frequency is 10 Hz with Gaussian spectral mode. The propagation direction of the laser beam is considered to be along the z-axis. The beam is focused using a convex lens and the focal point is taken as $z = 0$. The beam has maximum energy density (fluence) at the focus, which will symmetrically reduce towards either side of it on the z-axis. In the experiment, the sample is scanned along the z-axis. The light beam transmitted through the sample is detected by a high sensitive detector, and the measured signals are averaged over 10 pulses and recorded by an energy ratio meter as a function of the sample position z. The position-transmission curve thus obtained is the open aperture Z-scan curve and the nonlinear absorption coefficient of the sample can be calculated from the curve.

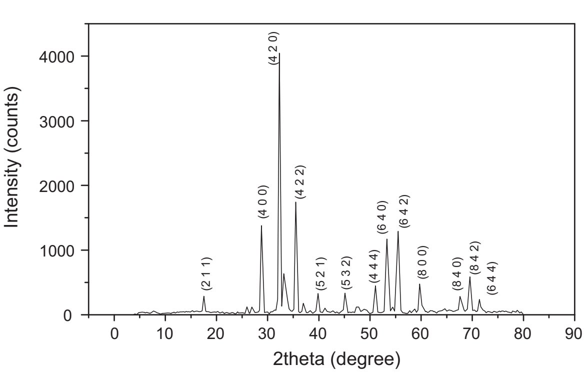

Fig. 1. XRD pattern of YIG nanoparticles calcined in air at 800 °C.

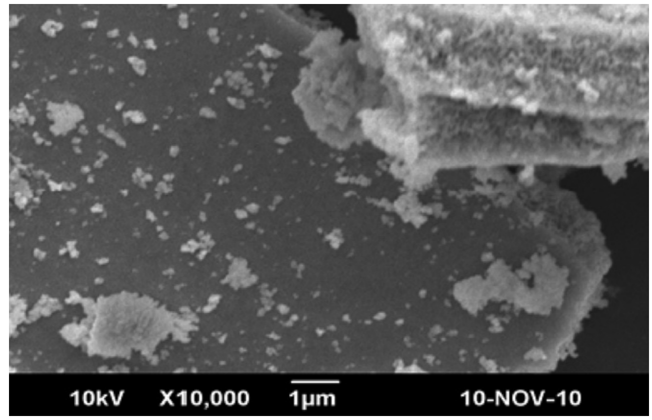

Fig. 2. Scanning Electron Micrograph of YIG powders.

3. Results and discussion

Fig. 3a and b shows the open aperture Z-scan curves obtained for YIG at two different laser powers of 152 and 364 MW cm^{-2}, respectively. Dotted lines represent the experimental data while the solid lines are theoretical fits to the open aperture scans using standard equations. It is well understood that the nonlinear absorption in such materials due to nanosecond pulses has contributions from both excited singlet and/or triplet states apart from two-photon absorption (simultaneous as well as instantaneous), depending on the excitation wavelength. A detailed five-level modeling along with the accurate knowledge of the excited state life times is essential to pin-point the exact contribution of each of these processes. However, for 532 nm excitation we can approximate the nonlinear absorption to an effective process and evaluate the nonlinear coefficient approximately [15,16]. The role of instantaneous two-photon absorption will be negligible and can be ignored due to the nanosecond pulse duration used combined with the excitation wavelength of 532 nm. We have fitted the open aperture data to a more general model and evaluated the nonlinear absorption coefficient (β expressed in cm/GW). The nonlinear absorption coefficient β can be obtained from a best fitting performed on the experimental data of the open-aperture measurement with the equation [17]

$$T_{OA} = \sum_{m=0}^{\infty} \frac{[-\beta I_0 L_{eff}/(1 + z^2/z_0^2)]^m}{(m+1)^{3/2}} \quad (1)$$

where T_{OA} is the normalized transmittance for the open-aperture, $L_{eff} = (1 - \exp(-\alpha_0 L))/\alpha_0$ is the effective thickness of the

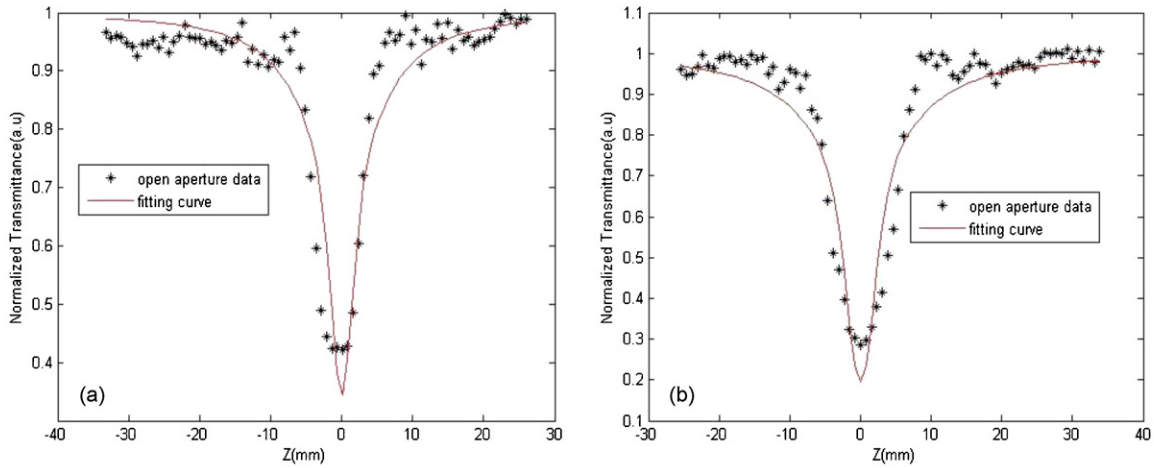

Fig. 3. Comparison of the theoretical and experimental open aperture Z-scan plots of YIG nanoparticles at 532 nm with different laser input powers: (a) 152 MW cm^{-2} and (b) 364 MW cm^{-2}.

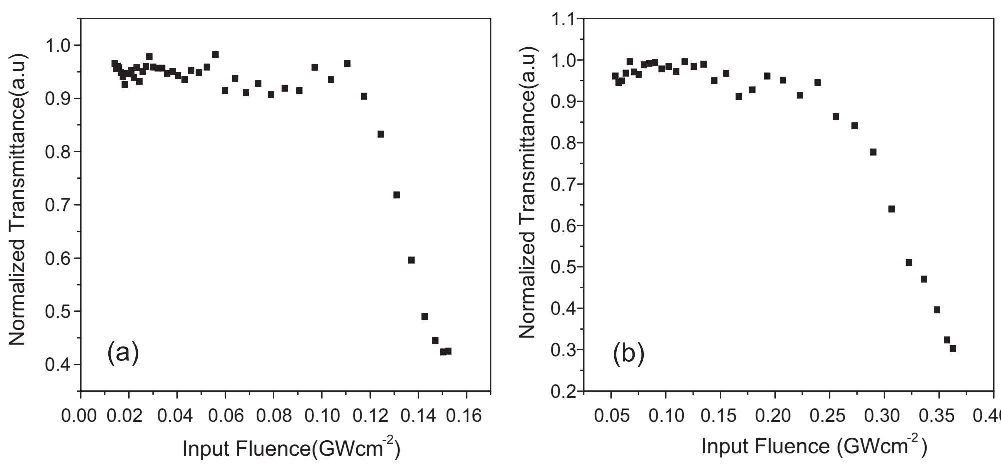

Fig. 4. Optical limiting response of YIG nanoparticles at 532 nm with different laser input powers: (a) 152 MW cm^{-2} and (b) 364 MW cm^{-2}.

sample (L denotes its thickness), α_0 is the linear absorption coefficient of the sample, and I_0 is the on-axis irradiance at focus. The open-aperture curve exhibits a normalized transmittance valley, indicating the presence of reverse saturable absorption (RSA) in the YIG nanoparticle. Normalized transmittance as a function of the position of the sample at wavelength of 532 nm with different laser powers (364, 152 MW cm^{-2}) is shown in Fig. 3. The calculated nonlinear absorption coefficients (β) are listed in Table 1 with an estimated error of 3%, which originates from the fluctuation of the laser pulse.

Z-scan data can be used to study the optical limiting property of nonlinear materials. Important application of RSA materials is in devices based on optical limiters. Candidates for optical limiting materials should have high transmittance for weak incident light, low transmittance for strong incident light, and instantaneous response over a broad spectral range [18]. YIG materials have relatively low linear absorption and strong RSA in the visible and near-infrared region and are suitable for optical limiting in this region. Fig. 4a and b illustrates the optical limiting response of YIG at an input laser intensity of 152 MW cm^{-2} and 364 MW cm^{-2} respectively. The limiting threshold at two laser intensities is tabulated in Table 1.

Table 1
Nonlinear optical properties of YIG nanoparticles at 532 nm.

Input laser power density (MW cm^{-2})	Nonlinear absorption coefficient β (cm/GW)	Optical limiting threshold (MW cm^{-2})
152	112.7	103
364	50.33	222

4. Conclusion

The NLO absorption properties of YIG nanoparticles were studied by using the open aperture Z-scan technique with different laser input powers and the nonlinear absorption coefficients were measured. The sample has shown strong RSA for 10 Hz and 7 ns laser pulses. The nonlinear absorption coefficients β and optical limiting threshold are obtained from the experimental results. The results reveal that YIG is a potential candidate optical limiting applications.

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