Optics and Laser Technology 109 (2019) 131–138

Contents lists available at ScienceDirect

Optics and Laser Technology

journal homepage: www.elsevier.com/locate/optlastec

Full length article

High nonlinear optical response of Lanthanum-doped TiO_2 nanorod arrays under pulsed laser irradiation at 532 nm

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HIGHLIGHTS

- This paper opens a new avenue to further development for nonlinear optical devices.
- Nonlinear optical properties of La-doped TiO_2 nanorods reported for the first time.
- Preparation and characterization of La-doped TiO_2 nanorod arrays were reported.
- Samples exhibited the strong nonlinearity which measured by using Z-scan technique.

ARTICLE INFO

Keywords:
La-doped TiO_2 nanorods
Hydrothermal method
Nonlinear optics
Z-scan technique

ABSTRACT

La-doped TiO_2 nanorod arrays were prepared using the hydrothermal method and the effects of doping on the structural and linear optical properties were considered by using UV–Vis diffused reflectance spectroscopy (DRS), field emission scanning electron microscopy (FESEM), X-ray diffraction (XRD), energy dispersive X-ray spectroscopy (EDS), and X-ray photoelectron spectroscopy (XPS) analyses. The XPS analysis confirmed the presence of La doping in TiO_2 nanorod arrays. According to XRD patterns, La doping leads to an enhancement of lattice parameter, c, as well as a reduction in the other lattice parameter, a, which indicates an expansion along the c direction of the lattice in La-doped TiO_2 nanorods. From UV–Vis DRS analysis, the band gap energy of undoped and doped samples were calculated by using Kubelka- Munk method that exhibited a slight decreasing with adding of La doping. The measurements of nonlinear optical (NLO) properties of undoped and La-doped TiO_2 nanorod arrays carried out using a nanosecond Nd: YAG pulse laser by using the Z-scan technique. Both samples showed a negative NLO refractive index at 532 nm. The NLO absorption of undoped and La-doped nanorod arrays is attributed to two-photon absorption. The nonlinear susceptibility, $\chi^{(3)}$, of undoped and La-doped TiO_2 nanorods was determined by the Z-scan technique of the order of 10^{-5} esu which is remarkably large value. The consequences suggest that undoped and La-doped TiO_2 nanorod arrays may be a promising candidate for the NLO applications.

1. Introduction

Search for the finding of new materials with high NLO capabilities has been one of the great challenges facing scientists in recent years. Nonlinear optics, study the interaction of high-intensity light with the matter which leads to NLO effects such as self-focusing, solitons and high-harmonic generation. Despite the large number of substances that are candidates for nonlinear optics, there are some limitations in their usage in the optoelectronic devices that include: the difficulty in the production process, the high cost of raw materials, toxicity, and so on.

Semiconductors are the most important materials with widespread applications in optoelectronics technology. Among them, titanium dioxide (TiO_2) is a wide band gap (3.2 eV for anatase and 3.0 eV for rutile phases) functional semiconductor material which has an extensive scientific and industrial applications in photovoltaic and photochromic devices, photocatalysis, water splitting, sensors, and lithium -ion batteries [1,2]. It is believed that single crystal one-dimensional nanomaterials compared to the random network of nanoparticles should increase the electron transport rate and improve the performance of optoelectronic devices [3]. Doping is a kind of modification method that can change the electronic properties of the material in order to alter its optical and electrical properties. Modification of TiO_2

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https://doi.org/10.1016/j.optlastec.2018.07.075
Received 12 April 2018; Received in revised form 8 July 2018; Accepted 30 July 2018
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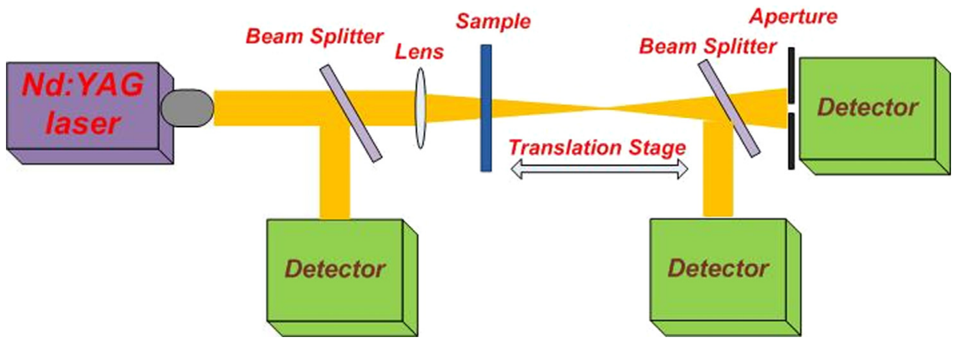

Fig. 1. Schematic diagram of the experimental Z-scan setup.

nanomaterials by doping could extend the light absorption toward the visible wavelength region. Due to superior properties of TiO_2 nanorods, doping of it with metal elements (Eu, Sn, Nb, Mg, and La) by the hydrothermal method is investigated for enhanced charge transport in solar cells [4–8]. Sadhu and coworker investigated the optoelectronic properties of La-doped TiO_2 nanorod arrays for photoelectrochemical application [8]. In addition, Meksi et al. considered the photocatalytic properties of La-doped 1D-TiO_2 nanorods and nanotubes [9]. Also, TiO_2 and its composites are considered as functional semiconductors that have shown good nonlinearity at various wavelengths [10–18]. The nonlinear absorption coefficient of La-doped Bi_3TiNbO_9 thin films reported for two-photon absorption (2PA) and three-photon absorption (3PA) cases [19].

In the present work, Z-scan technique is used for consideration of NLO properties of undoped and La-doped TiO_2 nanorods grown on FTO coating glass substrates. To the best of our knowledge, it is the first report on the NLO properties of La-doped TiO_2 structures. The undoped and La-doped TiO_2 nanorod arrays were grown by using the hydrothermal synthesis method on FTO coated glass substrates. The influence of the La doping on morphology, crystal structure, and optical nonlinearity was reported.

2. Experiments

2.1. Preparation of sample

La-doped TiO_2 nanorods are synthesized on fluorine-doped tin-oxide (FTO) coated glass substrates (15 Ω/g; Solaronix) by a one-step hydrothermal synthesis method. The FTO glasses are cleaned ultrasonically with acetone, ethanol, and deionized water, respectively. The reaction solution is prepared by mixing HCl, deionized water, titanium butoxide, and lanthanum nitrate hexahydrate (La $(NO_3)_3{\cdot}6H_2O$). The La/Ti molar ratio in doped samples is approximately equal to 0.4%. The substrates are placed horizontally with the conducting side up, into the reaction solution in a Teflon lined autoclave. The reaction temperature during hydrothermal processes is kept constant at 180 °C for 4 h. After synthesis, the autoclave is cooled down to room temperature naturally for 1 h. Then, the samples rinse with deionized water and allowed to dry in ambient air. The samples are annealed for 30 min at 450 °C.

2.2. Characterization

The morphology of nanorods was characterized by using a field emission scanning electron microscope (FESEM; Hitachi S-4166). The crystal structure of undoped and La-doped TiO_2 nanorods is examined by using an X-ray diffraction (XRD; X' Pert ProMPD) equipped with a Cu Kα radiation (λ = 0.15460 nm). The surface composition and chemical states of the samples are examined by X-ray photoelectron spectrometer (XPS; BESTEC). The base pressure was maintained at 0.75×10^{-10} Torr. Monochromatic Al Kα X-rays (1486.6 eV) was employed as the excitation source operated at 10 kV. The binding energies were calibrated with respect to the C1s peak (285 eV). EDS analysis was carried out by a FESEM; Mira2 TESCAN device.

2.3. Z-scan technique

The Z-scan technique is known as a very convenient and fast experimental method to apply for consideration of NLO properties of materials. The Z-scan technique is performed by translating a sample through the beam waist of a focused beam and then measuring the power transmitted through the sample [20].

In the Z-scan measurement setup which used in this work, the light source was an Ekspla NL640 model SHG Q-switched Nd: YAG laser with pulses of 10 ns duration that operating at 532 nm. The laser beam with a repetition rate of 200 Hz was focused by a lens to a beam waist radius 20 μm. These operating conditions produced an intensity of 113 MW cm^{-2} at the focal point. This configuration was applied previously for the calculation of the NLO parameters of Al-doped ZnO thin films grown on the FTO coating glass substrate [21].

Fig. 1 illustrates the experimental Z-scan apparatus, schematically. In this configuration, it is possible to measure simultaneously both NLO refractive index (n_2) and NLO absorption coefficient (β) by using only one measurement.

As can be seen in Fig. 1, an intense laser beam was sent through a focal length lens and was divided by a beam splitter. The beam splitter used to split the input beam and a fraction of the beam is sent to the first detector. Undoped and La-doped TiO_2 nanorods grown on the FTO coated glass substrate were translated separately through the beam waist using a motorized translation stage. The samples are moved in the Z-direction from the −Z to +Z, along the light propagation path. The remainder of the beam focused using a lens and then was split by using another beam splitter. The second detector records this beam after passing the sample which named open-aperture signal. The remainder of the beam was sent through an aperture and then recorded by using the third detector and named closed-aperture signal. A PC was used to collect and process the data that received from all three detectors. Multiple software was applied to calculate NLO parameters of undoped and La-doped TiO_2 nanorod arrays.

3. Results and discussion

3.1. Structural and morphological properties

Fig. 2(a) and (b) show the top view and cross-section FESEM images of La-doped TiO_2 nanorods grown on FTO coated glass substrates. The average diameter of La-doped nanorods is 101 ± 28 nm, while the diameter of the undoped TiO_2 nanorods is 99 ± 13 nm. Also, the length of doped nanorods is approximately equal to 1.5 μm. Thus, according to these images well aligned La-doped TiO_2 nanorods are grown on FTO substrates and doping has no significant effect on the morphology of TiO_2 nanorods.

The elemental mapping of doped sample by using FESEM with

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