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Femtosecond laser excited second harmonic and multiphoton absorption induced UV luminescence generation behaviour of ZnO nanofibers

Rudrashish Panda[a], Avanendra Singh[b], Rudranarayan Samal[a], Sayantan Bhattacharya[c], Pratap K. Sahoo[b], Prasanta K. Datta[c], Susanta K. Das[a,*]

[a] Dept. of Physics, School of Applied Sciences, KIIT University, Bhubaneswar, Odisha, 751024, India
[b] School of Physical Sciences, NISER, HBNI, Bhubaneswar, Odisha, 752050, India
[c] Dept. of Physics, Indian Institute of Technology, Kharagpur, West Bengal 721302, India

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ABSTRACT

Femtosecond (fs) laser excited second harmonic (SH) and multiphoton absorption (MPA) induced UV luminescence generation behaviour of Zinc oxide (ZnO) nanofibers is reported here. The used nanofibers are grown by seed assisted chemical bath deposition technology over glass substrates and are excited by a near-infrared (NIR) Ti: Sapphire laser emitting pulses of wavelength 812 nm and duration 100 fs. The peak wavelength of SH and MPA induced UV luminescence signals are found to be 406 nm and 398 nm respectively. Due to good optical quality, no defect level related MPA induced visible luminescence is observed in nanofibers. In comparison to this, ZnO nanostructures grown over plane substrate without seed layer shows very less SH and MPA induced UV luminescence. However in this case, the defect level related MPA induced visible luminescence was found to be very high. Applications like ultrafast pulse diagnostics, NIR induced photodynamic therapy etc. require high SH and MPA induced UV luminescence and low defect level related visible luminescence. So the ZnO nanofibers can be a potential candidate for these applications.

1. Introduction

The nonlinear optical effects are significantly enhanced in nanostructures due to surface in-homogeneities, higher charge densities and electrostatic potential gradient caused by grain boundaries [1,2]. Compared to other frequency conversion and short wavelength laser schemes, ultrafast multiphoton induced luminescence of semiconductors by NIR photons has become an efficient way to generate highly energetic radiation. The systematic study on such excitation channels of wide bandgap semiconductors using ultrafast lasers are of growing importance. ZnO nanostructures are found to be promising candidates for this purpose because of their direct wide band-gap (3.37 eV), intrinsically high nonlinear coefficients [3,4] and transparency at the laser irradiation wavelengths in the NIR spectral region. In this aspect several forms of ZnO nanostructures like nanocrystals [5], nanorods [6] and nanowires [7] has been extensively studied. Recently ZnO nanofibers have drawn much research attention. The surface to volume ratio of such nanostructures is very high and hence these structures are found to be very promising for various applications like photocatalysis [8], gas sensor [9], dye-sensitized solar cell [10],

* Corresponding author.
E-mail addresses: skdasfpy@kiit.ac.in, susant2@rediffmail.com (S.K. Das).

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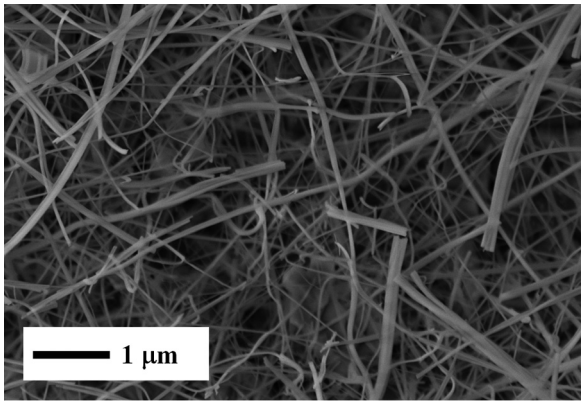

Fig. 1. FESEM image of ZnO nanofibers grown on glass substrate with seed layer.

glucose biosensors [11], photonics [12,13] etc. In powder second harmonic generation (SHG) experiment using nanosecond (Nd:YAG) laser Vijayakumar et al. reported that ZnO nanofibers grown by electrospinning and sol-gel technique can show 11.1 times higher efficiency than that of standard nonlinear optical material Potassium Dihydrogen Phosphate (KDP) [14]. In this work, we did the systematic studies on SH and MPA induced luminescence generation behaviour of ZnO nanofibers excited with NIR fs laser. Comparison is made for the nanostructures grown with and without seed layer. The potential applications are discussed.

2. Experimental

2.1. Material synthesis and characterization

The ZnO nanofibers are grown on glass substrate by seed assisted chemical bath deposition method. For this, firstly, a solution of 5 mM concentration is prepared by mixing Zinc acetate dihydrate with absolute ethanol followed by slow stirring. Then this solution is drop coated four times on a glass substrate. Finally, the sample is calcinated in air medium at 132 °C for 120 min by using an indigenously built induction heater based heating system to get the seed layer. This substrate is then dipped into a chemical bath containing a mixture of aqueous solution of Zinc nitrate (20 mM) and NaOH (0.8 M). The solution is heated and maintained at 70 °C with constant stirring for 120 min to get the film containing ZnO nanofibers. The film is then washed with deionized water and dried with hot air. The formation of ZnO nanofibers are verified by field emission scanning electron microscopy (FESEM) and X-ray diffraction analysis.

2.2. SH and MPA induced luminescence generation

For SH and MPA induced luminescence generation study the samples are excited by an amplified Ti: Sapphire laser of pulse duration 100 fs at 1 kHz repetition rate and central wavelength 812 nm. A BG40 color glass filter is used to separate the generated radiation from the fundamental/incident radiation. The generated signals are detected by using a fiber coupled spectrometer (Ocean Optics-USB4000).

3. Results and discussion

Fig. 1 shows the FESEM image of the ZnO nanofibers grown on the substrate with seed layer. The average diameter and length of the nanofibers are found to be 86 nm and 20 μm respectively.

The generated radiation from the nanofibers within the wavelength range of 375 nm to 700 nm is shown in Fig. 2(a). The fundamental radiation is shown in the inset of this figure. The generated signal in more specific wavelength range (390 nm to 420 nm) is shown in Fig. 2(b). Here it is found that the generated spectrum can be resolved into two peaks at 398 nm and 406 nm respectively. The peak at 398 nm is due to MPA bandedge luminescence which is 230 meV below the bandgap, 3.37 eV (368 nm) of ZnO at room temperature and is a result of bandgap renormalization [15,16]. The peak at 406 nm is the SH signal as because the central wavelength of the fundamental radiation is 812 nm (Ref. inset of Fig. 2a). As can be seen from Fig. 2(a); except the SH and MPA induced UV luminescence no other signal is found from ZnO nanofibers.

For comparison, ZnO nanostructures are grown on the substrate without any seed layer. The FESEM images of the nanostructures grown in this condition are shown in Fig. 3. Here the 1D ZnO nanostructures are less dense compared to ZnO nanofibers grown on substrate with seed layer (Refer Fig. 1).

Fig. 4(a) shows the generated signal from the nanostructures grown without any seed assistance whereas Fig. 4(b) shows the Gaussian fits for the SH and MPA induced luminescence signal. Here we also got signal in UV range which can be resolved