

Effect of vacuumization on the photoluminescence and photoresponse decay of the zinc oxide nanostructures grown by different methods



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ABSTRACT

Influence of vacuumization on the photoluminescence (PL) spectra and photoresponse decay of ZnO nanostructures fabricated by different methods was investigated. The visible band of photoluminescence and ultraviolet (UV) photosensitivity of the samples grown from a vapor phase was associated with the intrinsic defects such as doubly charged zinc vacancies, and for the samples grown by hydrothermal method – with presence of the oxygen vacancies. The experimental results show that ZnO nanostructures grown from the vapor phase would be promising for producing of the low cost and effective UV detecting devices.

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1. Introduction

Because of low cost and ease of manufacturing, ZnO is emerging as a potential alternative to GaN in the optoelectronic applications [1]. In recent years, the extraordinary multifunctional properties of ZnO nanostructures were demonstrated, in particular on the example of based on them field effect transistors, nanogenerators, solar cells, light-emitting diodes [2], optically pumped lasers [3], chemical and biological sensors [4], detectors of ultraviolet and optical switches [1].

Despite of the quite long history of the industrial applications of ZnO, there is no clear understanding of some of the fundamental properties of this material [1,4]. In particular, the emission bands of the zinc oxide have not been completely interpreted. A precise understanding of the nature of these bands is necessary for creating of light emitting diodes or lasers based on ZnO. Thus, the nature of the green luminescence band of the undoped ZnO has been controversial for decades. In early papers the defect responsible for the green luminescence was uniquely considered to be Cu²⁺ ions replacing the zinc ions at the crystal lattice sites [5]. But now, the relevant defect is considered to be either oxygen (V_O) [6] or zinc (V_{Zn}) [7] vacancies, or interstitial zinc atoms (Zn_i) [8].

UV photodetectors have been widely used in the various commercial and military applications including but not limited to secure space-to-space communications, pollution monitoring, water sterilization, flame sensing and early missile plume detection [9]. The most common UV detectors are based on *p-i-n* Si photodiodes or UV-filtered photomultiplier tubes. Application of the nitride semiconductor UV detectors has advantages in terms of more precise detection window, lower background currents due to solar flux and a wider range of operating temperatures [10]. In recent years, ZnO was found to be one of the promising materials for UV photodetectors due to its wide direct bandgap (~3.37 eV), high radiation durability, low cost, non-toxicity and biological compatibility as well. ZnO-based UV photodetectors in recent years were fabricated on the basis of the single crystals, thin films and nanostructures [9].

In this paper we present the data concerning the impact of the surface defects on the luminescent properties, photodetection and photoconductivity of the ZnO nanostructures grown by different methods.

2. Experimental

ZnO nanostructures were grown on the sapphire substrates with pre-deposited Au electrodes by two different methods. Distance between Au electrodes did not exceed 100 μm. ZnO nanowires (Fig. 1) were produced by the method of gas-transport

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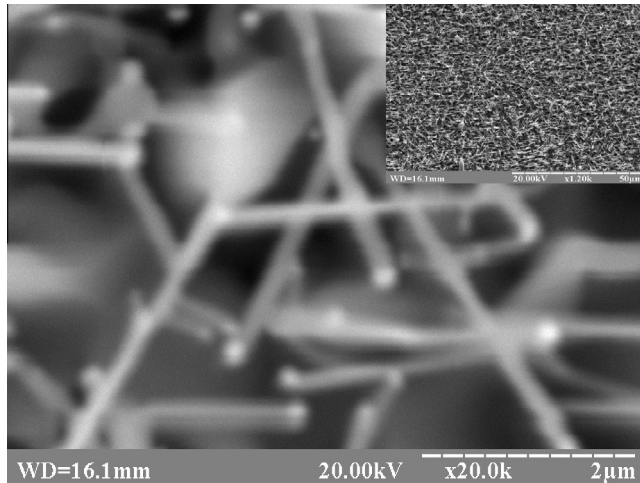


Fig. 1. Microphotographs of ZnO nanowires.

reactions [11]. The mixture of the powdered high purity zinc oxide and graphite in the proportion of 1:2 was taken as the initial material for vaporization. This material and a sapphire substrate with a (0112) crystallographic orientation were placed into a quartz tube. The mixture of powders was placed in a sealed end of the tube whereas the substrate - near the open end. The quartz tube was placed into a horizontal oven. The powder mixture was heated to the temperature of about 1050 °C and the substrate was located into a zone with the temperatures of 850–900 °C. This temperature distribution was maintained for 2 h. Afterwards the oven was shut off and cooled spontaneously to room temperature. This yielded a uniformly deposited, white layer of zinc oxide on the substrates.

ZnO nanorods (Fig. 2) were grown by hydrothermal method [12]. For seed-supported growth of ZnO nanorods, the nanocrystalline ZnO seed particles were prepared in a solution of 0.005 mol/L zinc acetate $[\text{Zn}(\text{CH}_3\text{CO}_2)_2]$ dissolved in ethanol at 90 °C for 15 min. The seed solution was then spin-coated onto $\text{Au}/\text{Al}_2\text{O}_3$ substrate. Subsequently, the seed-coated substrates were thermally annealed at 300 °C for 5 min to remove the residual solvent. The procedures of spin coating and thermal annealing were repeated twice for ZnO seed growth. On the next stage the $\text{Au}/\text{Al}_2\text{O}_3$ substrate with ZnO seeds were immersed in the same main growth solution consisting of 0.05 mol/L zinc nitrate hexahydrate $(\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O})$, 0.025 mol/L hexamethylenetetramine

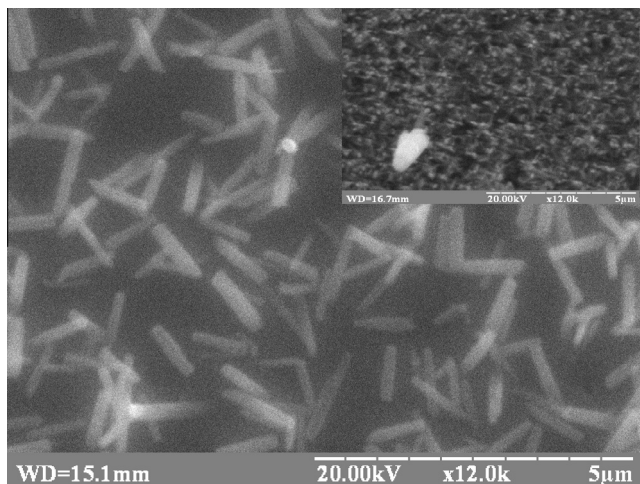


Fig. 2. Microphotographs of ZnO nanorods.

$(\text{C}_6\text{H}_{12}\text{N}_4)$ and distilled water. ZnO nanorods were grown at 90 °C for 90 min.

Morphology of the samples was examined using REMMA-102-02 Scanning Electron Microscope-Analyzer (JCS SELMI, Ukraine).

The room-temperature photoluminescence spectra (PL) were measured using automated monochromator/spectrograph M266 (SolarLS JSC, Minsk, Belarus) connected with CCD camera, based on Hamamatsu S7030-1006S sensor. The samples were excited by FQSS266-Q2 Nd:YAG laser (266 nm).

The measurements of the current-voltage (*I-V*) characteristics were carried out using Keithley Model 2401 Low Voltage Source-Meter (Keithley Instruments Inc., Ohio, USA) multimeter.

As a light source for the photoresponse decay measurements there was used 360-nm wavelength line obtained from a Xe lamp (1 kW) using a monochromator.

All experimental measurements were performed in air as well as in vacuum at a pressure of 0.5 Pa.

3. Results and discussion

The morphology of ZnO nanowires and nanorods is shown in Figs. 1 and 2, respectively. ZnO nanowires were found to possess a diameter of 150–200 nm and a length of 5 µm. ZnO nanorods are characterized by the uniform length of about 1 µm with an average diameter of about 200 nm.

The room-temperature PL spectra of ZnO nanowires (Fig. 3) and nanorods (Fig. 4) obtained in vacuum and air consists of the two bands in the UV and visible regions.

The band at 388 nm is typical for ZnO and arises due to recombination of the free excitons, bound excitons and transitions in the donor-acceptor pairs [4]. The wide band in the range from approximately 450 nm to 750 nm is caused by defects, first of all, by uncontrolled impurities and stoichiometry defects [4]. For both types of the samples after evacuating of air till the pressure of 0.5 Pa one could observe a slight increase of the ultraviolet band intensity, whereas the intensity of the visible band was changed in a different way. In the case of ZnO nanowires in vacuum the integrated intensity of the visible band increased by about 6%, whereas in the case of nanorods – vice versa, decreased by 17%.

According to [13] the emission of ZnO nanorods peaking near 600 nm is originating from the hydroxyl groups attached to the ZnO nanorods. These groups are also found to be removed upon evacuation, suggesting that there is a physical adsorption on a surface of the nanorods. When the PL spectrum is recorded again at the atmospheric condition, the visible intensity recovers almost

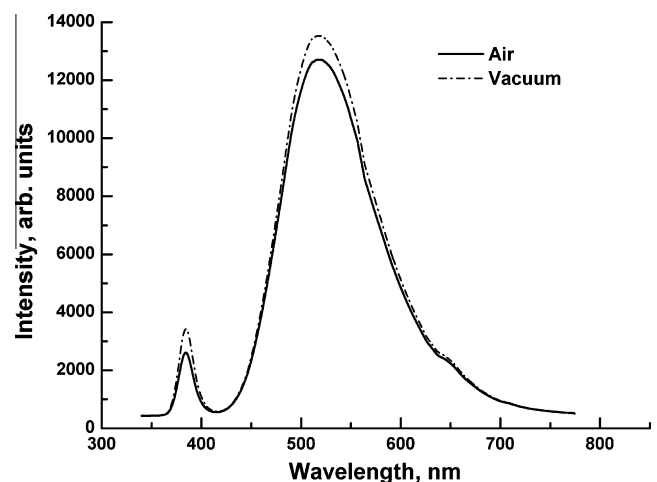


Fig. 3. PL spectra of the ZnO nanowires in vacuum and air.

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