



Influence of technological factors on conductivity and dielectric dispersion in ZnO nanocrystalline thin films

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ABSTRACT

Influence of the technological factors such as annealing of a substrate on the dielectric dispersion of the zinc oxide thin films with nanograins was investigated. In the first stage the dielectric dispersion of such nanostructured zinc oxide films at frequency range of 4 Hz–100 kHz and temperature range of 120–380 K was studied. The two types of films were prepared by the RF-magnetron sputtering in argon atmosphere using ZnO targets. The first sample was obtained by sputtering on the substrate heated to 573 K, whereas the second sample was manufactured without substrate heating. We observed the relaxation process in these materials at the temperature range of 220–350 K within the above mentioned frequency window. The second relaxation process was recognized in the frequency range of 100 kHz–2 MHz in vicinity of 350 K. The specific dispersion connected with a piezoelectric effect in the first sample was observed in vicinity of 215 K. The value of dielectric permittivity reaches 1100 at frequencies of few Hertz and decreases with increase of measuring electric field frequency. The nature of the low-frequency relaxation process is connected with space-charge polarization on the grain boundary. It is realized due to the high defect concentration within the interfaces of the ZnO nanograins in comparison with a bulk material. The high-frequency relaxation process corresponds to the thermal dipole polarization involving electrons localized at oxygen vacancies.

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1. Introduction

ZnO has many advantages, as compared to other wide band gap semiconductors, such as high-thermal/chemical stabilities; the possibility of wet chemical etching and existence of large area wafers. They have all led to the demonstration of ZnO as an alternative material to nitride semiconductors [1,2]. Zinc oxide is a technologically important material exhibiting multifunctional properties for various applications in optoelectronic devices applications, such as solar cell, transparent conducting electrodes and heat mirror [3–5]. Transparent conducting ZnO films have been extensively studied in recent year. There are numerous methods to obtain zinc oxide thin films, including reactive and ion-assisted evaporation, laser ablation, spray pyrolysis, sol–gel processing, chemical vapor deposition and sputtering [6–9].

This work belongs to a series of articles, testifying that the dielectric properties of nanostructured semiconductor such as zinc oxide can be modified by different technological factors, for example, by changing of the nanocrystals' size or manner in which these

materials are synthesized [10]. Modification of the crystallites' sizes leads to the change of the arbitrary interfaces volume and its drastic influence on the dielectric properties, conductivity of a sample especially in a radio-frequency range. One of the most powerful methods for studying of the electrical properties of such materials is dielectric measurements within the wide frequency range. Investigations of the dielectric dispersion offer an opportunity to throw light on the different very complicated electronic processes in the nanostructured semiconductors. In this paper we study the dielectric properties of the zinc oxide polycrystalline films over a wide range of frequencies and temperatures as well as consider the impact of certain technological factors on the dielectric properties of these materials.

2. Material and methods

The thin films were deposited on the glass substrates with ITO buffer layers by the standard RF-magnetron sputtering using ZnO targets in the argon atmosphere at the gas pressure of 10^{-3} Torr. The RF-power was 100 W. The target-to-substrate distance was 60 mm, the magnet field strength 0.1 T. The substrate temperature of the sample ZnO-1 at sputtering was chosen to be 300 °C. The sample ZnO-2 was obtained without substrate heating. Prior to deposition, the target was pre-sputtered for 10 min in order to remove any contamination. According to the ellipsometric studies the film thickness was approximately 1.2 mkm.

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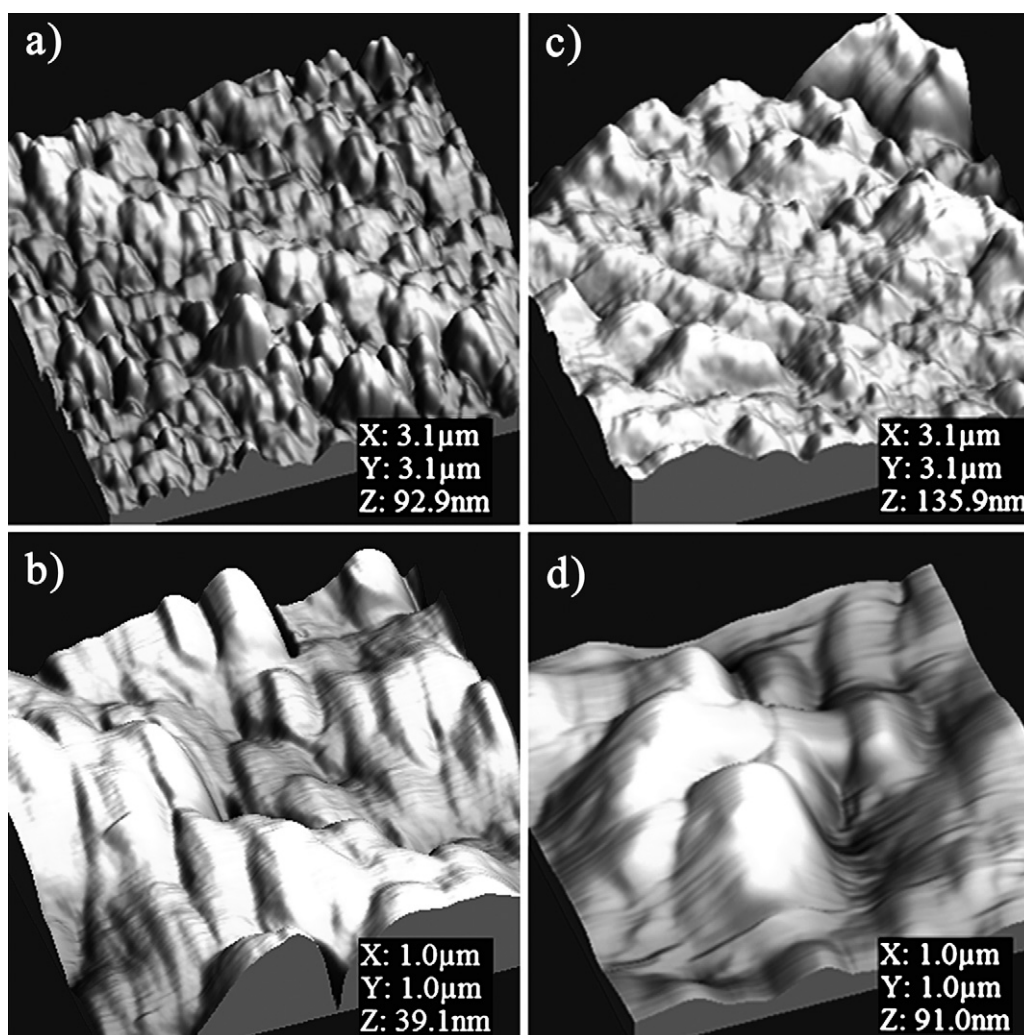


Fig. 1. AFM micrographs of the sample ZnO-1: (a), (b) and sample ZnO-2: (c), (d) in different scale.

The ITO films were prepared by RF-magnetron sputtering method on glass substrates using ITO target (10 wt.% SnO₂) in the argon atmosphere at the gas pressure of 10^{-3} Torr. The RF-power was 100 W. The target-to-substrate distance was 60 mm, the magnet field strength 0.1 T. The substrate temperature was 300 °C. According to ellipsometry measurements the thickness of ITO film was 300 nm. Aluminum electrodes were deposited by thermal evaporation of metallic aluminum of 99.9% purity in vacuum (10^{-5} Torr). The sequence of the layers on the glass substrate in both cases was ITO/ZnO/Al respectively. According to the microinterferometric studies thickness of the aluminum electrode was 600 nm.

The crystalline structure of the films was studied using the full-profile data obtained by automated HZG-4A diffractometer intended for examination of polycrystals. On the basis of X-ray diffraction study one can conclude that both films are textured along the crystallographic directions [001]. However, the sample ZnO-1 possesses better crystal quality and larger average crystallite size in comparison with the sample ZnO-2, since the peak (002) in the first case is more intense and sharper, and in addition it is observed also reflex on the plane (004).

The surface morphology of ZnO films was monitored by atomic force microscopy (AFM) (Digital Instruments – USA with Nanoscope E controller) working in the contact mode and equipped with OTR8 probe (Veeco NanoProbe™). The length and the spring constant of the applied V-shaped cantilever were 200 μm and 0.15 N/m respectively. The used constant forces were about 10 nN. All measurements were performed in air.

The dielectric measurements were performed on the samples of rectangular form, using automated setup. The measurements of the real part of dielectric permittivity and conductivity were carried out using traditional method of capacitor capacitance measurement. The capacitance was measured using LCR-meter HIOKI 3522-50 LCF HiTester at the frequency range of 4 Hz–100 kHz. Additionally, the dielectric parameters of ZnO films were measured at temperature 350 K and frequency range of 100 kHz–2 MHz. The nitrogen cryostat with UNIPAN 680 temperature control system was used for the dielectric measurements. The sample was placed into the cryostat. It was blown by the nitrogen fume. The

temperature was measured using a copper–constantan thermocouple with the margin of error 0.1 K.

3. Results and discussion

The micrographs of surfaces of the sample ZnO-1 and ZnO-2 are presented in Fig. 1. The image of each grain is contrast and well resolved. The samples surface, which was sputtered with heating of the substrate (ZnO-1), in most cases, is composed of the crystalline grain with a more regular structure in comparison with the sample ZnO-2. Indeed, the micrograph of surface of the second sample reveals the strong irregular structure with the separate well resolved islands on the background of weak variable surface. One can expect that the difference in the crystalline grain sizes should strongly influence the dielectric properties of both samples. The crystallite size obtained from AFM data for the sample ZnO-2 and sample ZnO-1 are equal 210 ± 120 nm and 140 ± 40 nm respectively. This means that the obtained films would be considered as the nanostructured (nanocrystalline) materials.

On the basis of the AFM study one can assume the interesting dielectric properties of the nanostructured semiconductor sample ZnO-1 and ZnO-2, especially at low radio-frequencies range. The temperature dependences of the real part of dielectric permittivity for the sample ZnO-1 measured in the range of 4 Hz–10 kHz are presented in Fig. 2. The wide asymmetric maximum of the

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