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# Size effect on memristive properties of nanocrystalline ZnO film for resistive synaptic devices

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**Abstract.** Size effect on memristive properties of nanocrystalline ZnO film was investigated. It was shown, ZnO film thickness increase from  $6.23 \pm 1.54$  nm to  $47.60 \pm 8.12$  nm leads to high-resistance state (HRS) increase from  $3.26 \pm 2.14$  M $\Omega$  to  $700.32 \pm 300.83$  M $\Omega$  and low-resistance state (LRS) from  $0.03 \pm 0.02$  M $\Omega$  to  $0.09 \pm 0.03$  M $\Omega$ , respectively. The HRS/LRS ratio increases from 108 to 7742. The results can be useful for based on nanocrystalline ZnO films resistive synaptic devices manufacturing.

## 1. Introduction

A resistive switching effect in metal/oxide/metal structures is attractive for resistive synaptic devices (RSD) manufacturing [1-9]. RSD offer significant advantages over conventional computers, such as an effective processing of unstructured data, high-density storage, low voltage operation [10, 11]. RSD technology based on nanocrystalline thin oxide films has great prospects for synaptic computer systems manufacturing [12-20]. ZnO is the one of the promising oxides, which widely used in electronic element developments, sensors and microsystem technology [5, 13]. Also ZnO demonstrates effect of resistive switching and it is compatible with semiconductor technology. To fabricate ZnO based resistive synaptic elements, it is necessary to study resistive switching in ZnO films and today there are insufficiently experimental results about. Also it is important to get reliable information about morphology and thickness of ZnO films during size effect investigations. Atomic Force Microscopy (AFM) is one of the promising techniques for surface diagnostics and analysis. Reliability of AFM images defines by many factors, most of which are related to the shape and quality of the preparation of probes [21]. So, it is important to prepare probe tip, which allows getting reliable information about morphology and thickness of ZnO films.

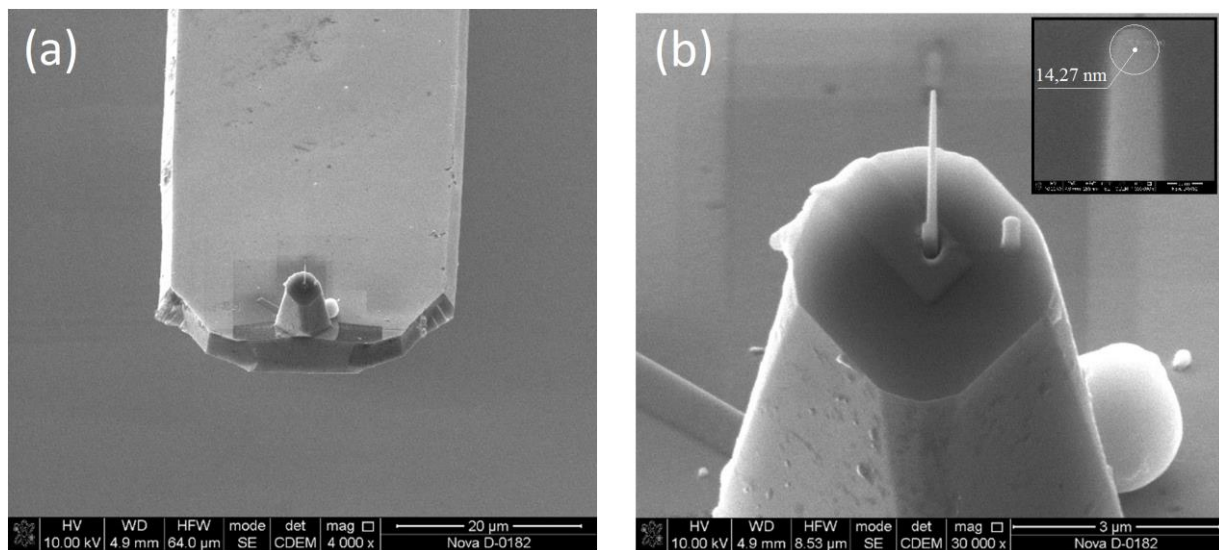
Thus, investigation of resistive switching dependence on the thickness of nanocrystalline ZnO films is the goal of this work.



## 2. Experiment details

To carry out experimental studies of the size effect on memristive properties of nanocrystalline ZnO film, five samples were prepared. Sapphire substrate  $\text{Al}_2\text{O}_3$  with a crystallographic orientation (0001) as a wafer was used. As the bottom electrode titanium nitride (TiN) was used. TiN was deposited by pulsed laser deposition technique for the following regimes: temperature 600 °C, number of pulses: 10000, frequency: 10 Hz, argon pressure: 1 mTorr. TiN film thickness was about  $30.3 \pm 5.1$  nm. Nanocrystalline ZnO films were grown also by pulsed laser deposition technique. To provide electrical contact to the bottom TiN electrode, the ZnO films were deposited through a special mask pattern. So, on a template-protected surface area of the TiN films, the ZnO did not precipitate.

To investigate ZnO films morphology, advanced probe tip for AFM was fabricated using the Focused Ion Beam (FIB) local milling. The fabrication of probe tip was performed with a FEI Company DualBeam system Nova NanoLab 600, combining a Ga<sup>+</sup> FIB and a field emission scanning electron microscope. At the first step, the probe was built by FIB-induced deposition of tungsten on a commercial Si cantilever NSG 10 with broken tip (resonant frequency: 287 kHz, force constant: 38 N/md) and then sharpening it using a focused ion beam milling. The following FIB parameters were used: the accelerating voltage of the ion beam – 28 keV; the ion beam current – 32 pA; and the dwell time of the ion beam — 0.9  $\mu\text{s}$ . The chamber pressure after introducing  $\text{W}(\text{CO})_6$  gas was  $1 \times 10^{-4}$  Pa. A bitmap of the desired probe structure was created by using Unigen 3.2 software, and then uploaded into the FIB software. Figure 1 shows secondary electron image (SEM) of the FIB-fabricated probe tip.



**Figure 1(a, b).** SEM of the FIB-fabricated AFM probe: (a) cantilever; (b) tip.

AFM images of surface of ZnO films were obtained using Probe Nanolaboratory Ntegra (NT-MDT, Russia) (figure 2). Electrical measurements were carried out using semiconductor characterization system Keithley 4200-SCS (Keithley, USA) with W probes. During experiment, TiN film was grounded. Current-voltage (IV) characteristics of the bottom electrodes of all the samples were obtained to confirm Ohmic behavior of TiN.

Initially, ZnO films of all five samples exhibited dielectric properties. So, current-voltage characteristics measurement took place in two steps. At the first step, the electroforming of the samples was carried out at the point after which the films began to exhibit a memristor effect at this point.

The electroforming voltage of the sample was determined as follows: after the probe was landed to the surface of the ZnO film, a continuous symmetrical linear voltage signal with an initial amplitude of 1V was fed to it at a point. At the same time, the values of the currents flowing through the film were

investigated. If an electrical breakdown of the film occurred, it was assumed that the given amplitude of the voltage is the electroforming voltage. If the electrical breakdown did not occur, the amplitude of the sweep increased by 1 V, and the process was repeated anew until the electroforming voltage was reached. It should be noted, that at voltages above the electroforming voltage, irreversible breakdown of the film occurred, and at voltages below the electroforming voltage, the film remains an insulator. The results of the electroforming voltage investigations of all five samples are shown in Table 1.

At the second step, the current-voltage characteristics were measured at the point (figure 3 a). For each sample, the average statistical IV characteristics at different points on the surface of the ZnO films for different number of electroforming cycles (0, 1, 2, 4, 6, 8, 10, 12) were received. IV curves were obtained depending on the thickness of ZnO films at the range from -1 V and from -5 to +5 V voltage sweep. 0.5 V was used as the read voltage. Curves analysis was implemented using Origin 8.1 software.

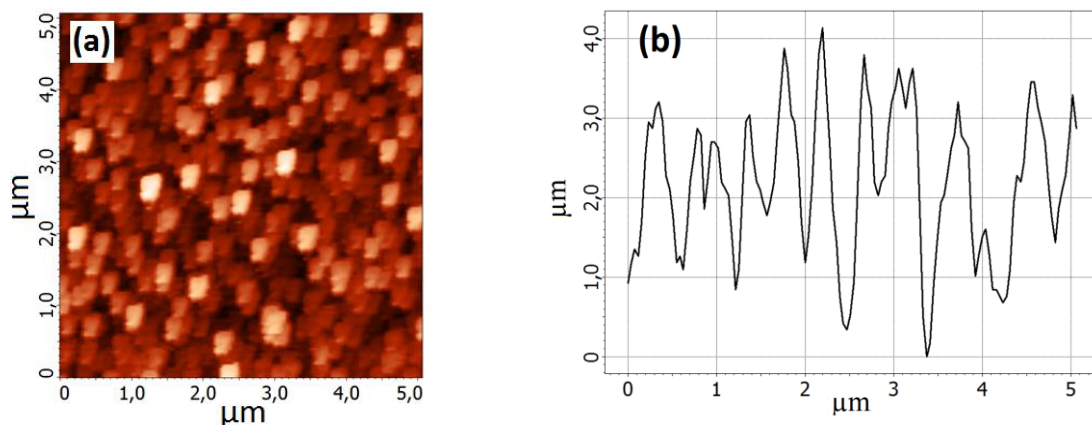
**Table 1.** Electroforming voltages of nanocrystalline ZnO films.

ZnO film thickness (nm)	6±1	20±3	40±6	45±8	47±8
Electroforming voltage (V)	3	9	14	15	16

### 3. Results

The analysis of SEM image (fig. 1) shows, that a probe with a tip radius of about 14.27 nm, cone angle 1° and aspect ratio 1:30 was obtained after fabrication. Figure 2 shows experimental investigation of ZnO film morphology. It is shown that ZnO film surface has a granular structure (figure 2 a) with  $0.7 \pm 0.3 \mu\text{m}^2$  grain size and  $3.2 \pm 1.1 \text{ nm}$  grain height (figure 2 b). The ZnO film thicknesses were investigated using AFM by scanning bottom TiN/ZnO film boundary, and were in range from  $6 \pm 1$  to  $47 \pm 8 \text{ nm}$  (table 1).

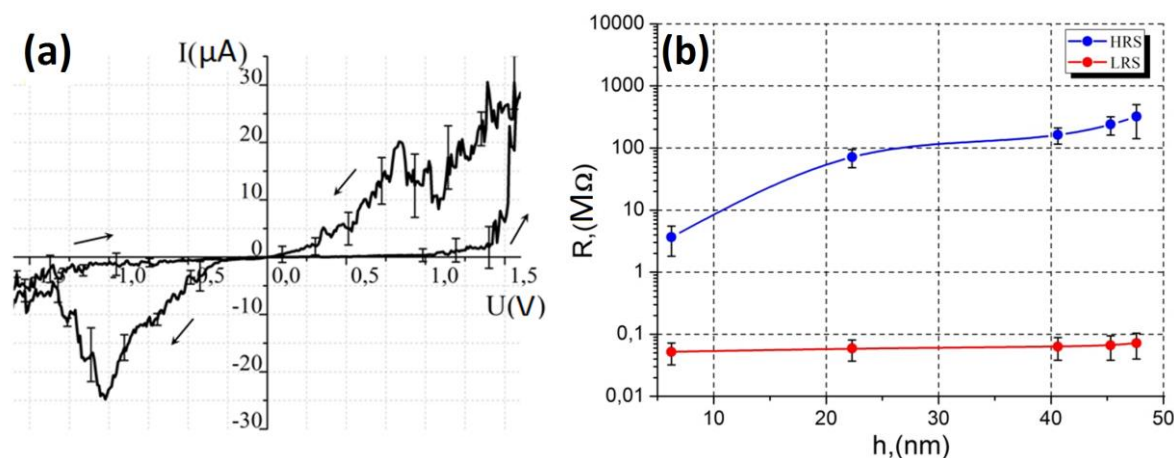
The number of electroforming cycles was chosen such that the maximum HRS/LRS ratio for the given sample was observed. Based on the results obtained, the dependences of the HRS and LRS on the thickness of the ZnO films (figure 3 b) were built.



**Figure 2(a, b).** Investigation of nanocrystalline ZnO film surface: **(a)** AFM image of nanocrystalline ZnO film and; **(b)** average profilogram of (a).

It was shown, ZnO film thickness increase from  $6.23 \pm 1.54 \text{ nm}$  to  $47.60 \pm 8.12 \text{ nm}$  leads to high-resistance state increase from  $3.26 \pm 2.14 \text{ M}\Omega$  to  $700.32 \pm 300.83 \text{ M}\Omega$  and low-resistance state from  $0.03 \pm 0.02 \text{ M}\Omega$  to  $0.09 \pm 0.03 \text{ M}\Omega$ , respectively. The HRS/LRS ratio increases from 108 to 7742. It can be explained by

increasing of the oxygen nanofilament dissolution length on metal/ZnO interface due to increasing of ZnO film thickness.



**Figure 3(a, b).** Size effect on memristive properties of nanocrystalline ZnO film investigation: **(a)** Average IV curve of TiN/ZnO/W structure; **(b)** HRS and LRS dependence from ZnO film thickness.

The obtained results can be used for development of technological processes of nanocrystalline ZnO film based resistive synaptic devices.

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