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Memristive effect in hydrothermal ZnO structures

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Abstract. In this paper, we investigate the memristor properties of ZnO microstructures synthesized by hydrothermal method. ZnO is a promising material for obtaining energy-efficient memory elements due to its compatibility with CMOS technologies, low cost, and good scalability. In this study, hexagonal ZnO microprisms with a diameter of ~10 μm and a thickness of ~2 μm were obtained. The study of the current-voltage characteristics revealed a unipolar memristor effect with switching between the high (HRS) and low (LRS) resistance states when applying both positive and negative voltage. The switching ratio I_{ON}/I_{OFF} was 10^2 for forward bias and 10^4 for reverse bias. The obtained results demonstrate the potential of ZnO structures for application in non-volatile memory with low power consumption.

Keywords: zinc oxide, hydrothermal, memristor

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Мемристорный эффект в гидротермальных структурах ZnO

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Аннотация. В данной работе исследуются мемристорные свойства микропризм ZnO, синтезированных гидротермальным методом. ZnO является перспективным материалом для создания энергоэффективных элементов памяти благодаря своей совместимости с КМОП-технологиями, низкой стоимости и хорошей масштабируемости. В ходе исследования получены гексагональные микропризмы ZnO диаметром ~10 мкм и толщиной ~2 мкм. Исследование вольт-амперных характеристик выявило униполярный мемристорный эффект с переключением между состояниями с высоким (HRS) и низким (LRS) сопротивлением при подаче как положительного, так и отрицательного напряжения. Коэффициент переключения I_{ON}/I_{OFF} составил 10^2 для прямого смещения и 10^4 для обратного смещения. Полученные результаты демонстрируют потенциал микроструктур ZnO для применения в энергонезависимой памяти с низким энергопотреблением.

Ключевые слова: оксид цинка, гидротермальный, мемристор

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Introduction

Memristors have become one of the most promising candidates in the field of fabricating miniature non-volatile memory elements [1]. A memristor is a nonlinear resistor, which conductivity depends on the electric charge flowing through it. Among their advantages are low power consumption, fast resistive switching between a high-resistance state (HRS) and a low-resistance state (LRS), good scalability.

Zinc oxide (ZnO) is a proven wide-bandgap semiconductor material [2] known for its piezoelectric properties [3], which allows it to be used as a material for resistive switching. Its advantages include its high content in nature, which determines its low cost, compatibility with CMOS technologies in terms of process integration, as well as the scalability of the resulting devices, down to nanometer sizes.

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Despite the fact that the memristor effect was previously demonstrated in ZnO nanostructures of various geometries [4, 5], the prospects for their application in memory device fabrication technologies were determined, additional system studies are required to determine the optimal geometry of ZnO-based nanostructures to observe a stable memristor effect with low switching power consumption and fast response.

Methodology

Hydrothermal synthesis is a method of growing various materials and compounds, based on the use of physical and chemical processes that take place in aqueous solutions at slightly elevated temperatures often used to obtain ZnO nanostructures [6].

In our study, we use a silicon substrate Si (100) coated with a 100 nm thick Ag layer for the hydrothermal synthesis of ZnO nanostructures. An aqueous solution of zinc acetate was used as a seed layer. The seed layers were deposited by the spin coating method. A solution of zinc acetate was dropwise applied to the substrate using a Pasteur pipette. Then, the substrate with the solution was spun in a centrifuge at 4000 rpm for 30 s. After the centrifuge, the sample was placed on a furnace and annealed at 350 °C for 3 minutes. A total of 3 such layers were deposited.

To study the memristor properties of zinc oxide, hexagonal microprisms were synthesized on the prepared substrate. The growth solution consists of equimolar aqueous solutions of $Zn(NO_3)_2$ and hexamethylenetetramine (HMTA) with a concentration of 100 $mmol \cdot L^{-1}$. Additionally, sodium citrate at a concentration of 5 $mmol \cdot L^{-1}$ was added to the growth solution to suppress vertical growth and stimulate lateral growth. Citrate ions are negatively charged, so they selectively bind to Zn^{2+} ions on the (0001) surface, block growth along the c axis and stimulate it in the $[01\bar{1}0]$ and $[21\bar{1}0]$ directions. This process leads to pronounced lateral growth and a decrease in the aspect ratio of the structures, which leads to the formation of hexagonal microprisms. The following reactions occur during the synthesis:

$$C_6H_{12}N_4 + 6H_2O \rightarrow 6HCHO + 4NH_3, \quad (1)$$

$$NH_3 + H_2O \rightarrow NH^{4+} + OH^-, \quad (2)$$

$$Zn(NO_3)_2 \leftrightarrow Zn^{2+} + 2NO_3, \quad (3)$$

$$2OH^- + Zn^{2+} \leftrightarrow Zn(OH)_2, \quad (4)$$

$$Zn(OH)_2 \leftrightarrow ZnO + H_2O. \quad (5)$$

During the synthesis, a constant temperature of 85°C was maintained. The synthesis duration was 3 hours. After synthesis, the samples were washed with deionized water and annealed in a furnace at a temperature of 350 °C for 5 minutes. The fabrication scheme of the experimental sample is shown in Fig. 1.

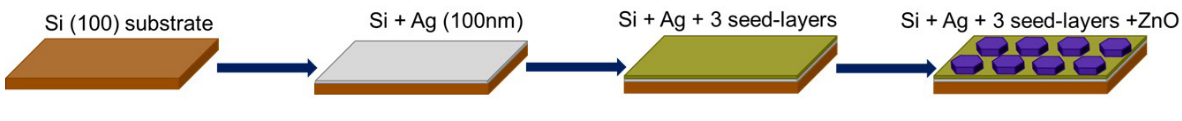

Fig. 1. Fabrication scheme

The study of the current-voltage characteristics in order to detect the memristive effect in the synthesized ZnO structures was carried out using the NTegra Aura, NT-MDT SI setup. The TipsNano HA_FM/W_2C probe was used. To measure the current-voltage characteristics, two probes were applied to the Ag/ZnO structure. One of the probes served as the upper (positive) contact to ZnO, the second as the lower (negative) contact to Ag.

Results

ZnO structures in the geometry of hexagonal prisms with a ~10 μm diameter and a ~2 μm thickness were synthesized (Fig. 2).

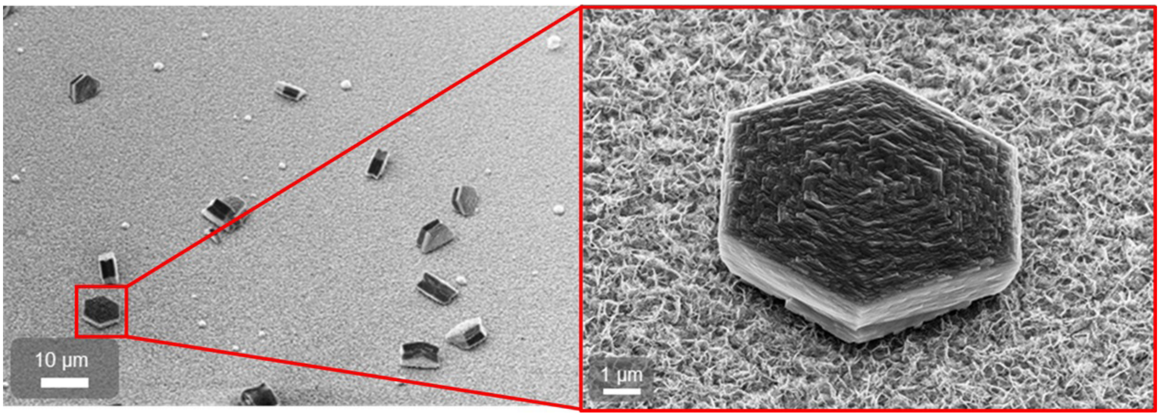

Fig. 2. SEM images of the synthesized sample

The I–V characteristic is divided into 4 stages (Fig. 3). At stage I, a positive voltage is applied to the sample (forward bias) transition from the high-resistance state to the low-resistance state. During the initial application of voltage (blue curve in Fig. 3, *a*), positively charged oxygen vacancies V_O^{++} drift toward the negatively charged electrode, forming a conducting channel in the ZnO semiconductor, significantly reducing the resistance of the Ag/ZnO/probe structure. This process is referred to as SET (turn-on). Thus, for a given polarity of the applied voltage, the Ag/ZnO/probe structure is in the low-resistance state (LRS). The "turn-on" voltage, V_{SET}, was 5.6 V. Stage II corresponds to LRS (red curve in Fig. 4, *a*). At stage III, a negative voltage is applied to the sample (reverse bias). A transition from the high-resistance state (HRS) to LRS is observed (blue curve in Fig. 3, *b*), as with the application of positive voltage. V_{SET} was 7.2 V. Stage IV corresponds to LRS (red curve in Fig. 3, *b*).

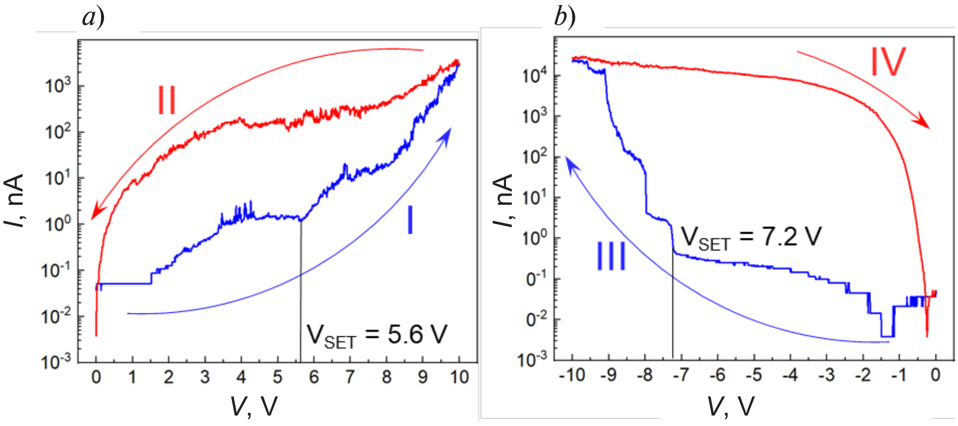

Fig. 3. Current–voltage characteristics of the synthesized sample:
forward bias (*a*); reverse bias (*b*)

The transition from HRS to LRS with the application of both positive and negative voltage indicates the unipolar nature of the obtained memristor. When applying forward voltage, the ratio of the "on" current to the "off" current $I_{ON}/I_{OFF} = 10^2$. In the case of applying reverse voltage $I_{ON}/I_{OFF} = 10^4$

Conclusion

The results confirmed occurrence of the memristor effect in hexagonal ZnO microprisms synthesized by the hydrothermal method. The unipolar nature of resistance switching, as well as high values I_{ON}/I_{OFF} indicate the prospects of using ZnO as an active material for next-generation memory elements. Further research should be aimed at obtaining methods for the ordered synthesis of zinc oxide in order to fabricate contacts to its surface.

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