Conference materials
UDC 621.38-022.532
DOI: https://doi.org/10.18721/JPM.161.238

Multilevel resistive switching in forming-free nanocrystalline ZnO films for neuromorphic applications

R.V. Tominov[1]✉, I.A. Shikhovtsov[1], D.A. Khakhulin[1], Z.E. Vakulov[2], V.A. Smirnov[1]

[1] Southern Federal University, Taganrog, Russia

[2] Southern Scientific Centre of the Russian Academy of Sciences (SSC RAS), Rostov-on-Don, Russia

✉ tominov@sfedu.ru

Abstract. We have experimentally studied the multilevel resistive switching in forming-free nanocrystalline ZnO films. It was shown that potentiation and depression at 0.5 V and –0.5 V for 3000 cycles led to the film resistance increasing by 3 orders of magnitude. In addition, it was shown that ZnO films successfully mimic biological memory through increased pulse number stimulation. Fixing the amplitude of the training pulses makes it possible to achieve different resistive states such as synaptic weight levels of the biological brain. The obtained results can be used for ReRAM elements of neuromorphic artificial intelligence systems fabrication based on forming-free nanocrystalline ZnO films.

Keywords: neuromorphic systems, memristor; ReRAM, multilevel resistive switching, forming-free nanocrystalline ZnO, pulsed laser deposition

Funding: The reported study was funded by the Russian Foundation for Basic Research according to research project 19-29-03041_mk and project 19-38-60052 (Electrical measurements in forming-free nanocrystalline ZnO film). ZnO thin film samples fabrication was carried out with the financial support of the by grant of the President of the Russian Federation MK6252.2021.4-. Potentiation and depression test was carried out with the financial support of the Russian Federation Government (Agreement 075-15-2022-1123). Investigation of multilevel resistive switching in forming-free nanocrystalline ZnO film was supported by a grant of the Russian Science Foundation 21-79-00216, https://rscf.ru/project/21-79-00216/, in Southern Federal University.

Citation: Tominov R.V., Shihovtsov I.A., Khakhulin D.A., Vakulov Z.E., Smirnov V.A., Multilevel resistive switching in forming-free nanocrystalline ZnO films for neuromorphic applications. St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 16 (1.2) (2023) 247–252. DOI: https://doi.org/10.18721/JPM.161.238

Материалы конференции
УДК 621.38-022.532
DOI: https://doi.org/10.18721/JPM.161.238

Многоуровневое резистивное переключение в forming-free нанокристаллических пленках ZnO для нейроморфных приложений

Р.В. Томинов [1]✉, И.А. Шиховцов[1], Д.А. Хахулин[1], З.Е. Вакулов[2], В.А. Смирнов[1]

[1] Южный федеральный университет, Таганрог, Россия

[2] Южный научный центр Российской академии наук (ЮНЦ РАН), Ростов-на-Дону, Россия

✉ tominov@sfedu.ru

Аннотация. Мы экспериментально исследовали многоуровневое резистивное переключение в forming-free нанокристаллических пленках ZnO. Было показано, что потенцирование и депрессия при 0,5 В и -0,5 В в течение 3000 циклов привели к увеличению сопротивления пленки на 3 порядка. Также было показано, что пленки ZnO успешно имитируют биологическую память путем увеличения числа импульсов стимуляции. Фиксация амплитуды обучающих импульсов позволяет достичь различных резистивных состояний, таких как уровни синаптического веса биологического мозга. Полученные результаты могут быть использованы для изготовления элементов ReRAM нейроморфных систем искусственного интеллекта на основе forming-free нанокристаллических пленок ZnO.

Ключевые слова: нейроморфные системы, мемристор; ReRAM, многоуровневое резистивное переключение, forming-free нанокристаллический ZnO, импульсное лазерное осаждение

Финансирование: Работа выполнена при финансовой поддержке Российского фонда фундаментальных исследований в рамках научно-исследовательского проекта № 19-29-03041_мк и проекта № 19-38-60052 (Электрические измерения в forming-free нанокристаллической пленке ZnO). Изготовление образцов тонкой пленки ZnO проводилось при финансовой поддержке гранта Президента РФ № МК-6252.2021.4. Потенцирование и депрессия пленок ZnO проводились при финансовой поддержке Правительства Российской Федерации (соглашение № 075-15-2022-1123). Исследование многоуровневого резистивного переключения в бесформенной нанокристаллической пленке ZnO проводилось при поддержке гранта Российского научного фонда № 21-79-00216, https://rscf.ru/project/21-79-00216/, в Южном федеральном университете.

Ссылка при цитировании: Томинов Р.В., Шиховцов И.А., Хахулин Д.А., Вакулов З.Е., Смирнов В.А. Многоуровневое резистивное переключение в forming-free нанокристаллических пленках ZnO для нейроморфных приложений // Научно-технические ведомости СПбГПУ. Физико-математические науки. 2023. Т. 16. № 1.2. С. 247–252. DOI: https://doi.org/10.18721/JPM.161.238

Introduction

In recent years, tasks related to perception, recognition, learning and memory functions have become increasingly important [1–3]. The concept of neuromorphic computing, which mimics the computational primitives of the biological brain, is replacing conventional von Neumann calculations [4–6]. Here, the main functional component is the synapse, which enables the transfer of information between neurons. It is worth noting the extreme complexity of this kind of system, where parallel data processing and information storage take place in a single device consisting of an array of nonvolatile and energy efficient memory elements operating in analog mode [7, 8]. It is important to ensure that any artificial synaptic device correctly recreates the computational primitives of biological synapses before using it in neuromorphic applications. One promising candidate as an element base for neuromorphic systems is non-volatile resistive memory (ReRAM), as it satisfies several important requirements, such as non-volatility, low power consumption and the presence of multiple stable resistive states between the high-resistance and low-resistance states (R_{HRS} and R_{LRS}) due to redistribution of oxygen ions and oxygen vacancies in the oxide volume [9, 10]. It is important to note that ReRAM can mimic various biological brain characteristics such as short-term plasticity (STP), long-term plasticity (LTP), spike-rate-dependent plasticity (SRDP), and spike-timing-dependent plasticity (STDP). In addition, ReRAM is bipolar, has a simple metal/dielectric/metal structure, which means it is compatible with conventional semiconductor technology, and offers a high integration density [12, 13].

Various classes of materials such as perovskites, chalcogenides, solid electrolytes, organic compounds, and metal oxides exhibit the memristor effect [14–16]. Among the latter, oxides such as ZnO, TiO_2, HfO_2, ZrO_2, etc. are of particular interest. Literary analysis showed that many scientific publications are devoted to the study of the creation of ReRAM memory elements based on forming-free nanocrystalline ZnO films, largely because this material has only one phase, which increases the stability of the neuromorphic systems created. There are many methods for producing metal oxide films, the main ones being atomic layer deposition (ALD), magnetron sputtering, pulsed laser deposition (PLD), etc. [17, 18]. A few studies have shown that ZnO nanocrystalline films grown using PLD exhibit impressive memristor effect parameters, with higher R_{HRS}/R_{LRS} ratios and low operating voltages [19—21]. However, there is still a lack of knowledge in various aspects of the fabrication of ReRAM neuromorphic elements based on this material, such as potentiation and depression processes as well as multi-bit properties, which is the aim of this paper. The results obtained can be used to create ReRAM elements of neuromorphic artificial intelligence systems based on forming-free nanocrystalline ZnO films.

Materials and Methods

Forming-free nanocrystalline ZnO films were fabricated using a Pioneer 180 pulsed laser deposition system (Neocera LCC, USA) equipped with a KrF excimer laser with a wavelength of 248 nm and an energy of 200 mJ. Silicon wafers with (100) crystallographic orientation was used as substrates. Titanium nitride (TiN) 42.3±5.2 nm films were formed using PLD under the following conditions: substrate temperature 600 °C, number of pulses 30,000, frequency 10 Hz, nitrogen pressure 1 mTorr. Forming-free nanocrystalline ZnO films were prepared on Si/TiN structure under the following conditions: substrate temperature 300 °C, laser frequency 10 Hz, number of pulses: 17,000.

The multilevel resistive switching in forming-free nanocrystalline ZnO films was studied using a Keithley 4200-SCS semiconductor parameter analyser (Keithley Instruments, USA), an EM-6070A submicrometer probe system (Planar, Republic of Belarus), Keithley electrometer/high resistance meter Series 6517B, and Keithley picoammeter/voltage source 6487. MathWorks MATLAB v9.12 (MathWorks, USA) was used for data processing. TiN bottom contact was grounded, and a tungsten probe with diameter about 80 nm was used as the top contact. The compliance current was set to 3 mA to avoid thermal breakdown of the investigated films. As a result, current-voltage curves (CVC) were obtained for the sweep voltage amplitude from 0.5 V to 1.5 V, a potentiation/depression study was performed at 3000 cycles at 0.5 V and –0.5 V, respectively. The resistance dependences on the cycle number were performed at 1000 cycles for the pulse voltage (up) 0.8 V and the read voltage 0.1 V.

Results and Discussion

Fig. 1 shows the results of experimental studies of electrical measurements in forming-free nanocrystalline zinc oxide films. Analysis of the results obtained showed that an increase in voltage amplitude from 0.5 V to 1.5 V leads to a decrease in the R_{HRS}/R_{LRS} ratio from 100 to 35 (Fig. 1,*a*), to a decrease in R_{HRS} and R_{LRS} from 1.92×10^6 Ω to 0.61×10^6 Ω and from 19.23×10^3 Ω to 16.09×10^3 Ω, respectively (Fig. 1,*b*).

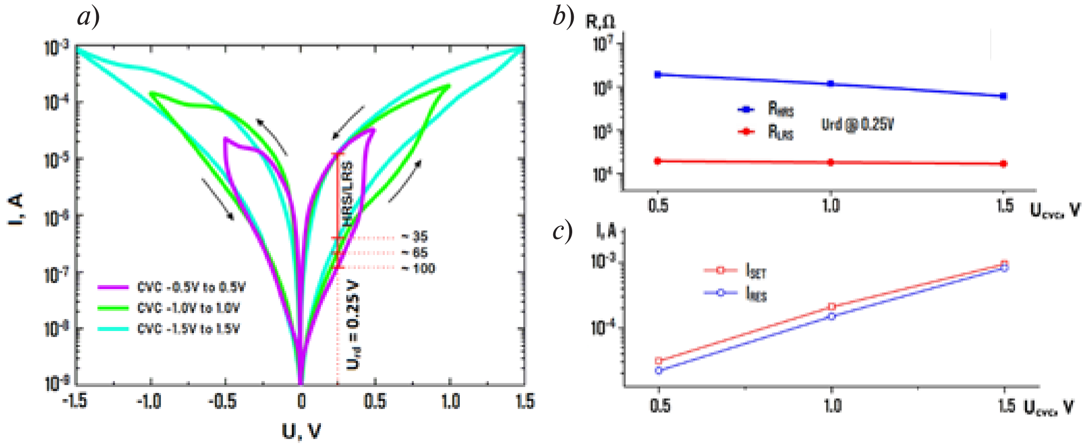

Fig. 1. Electrical measurements in the forming-free nanocrystalline ZnO film: current-voltage characteristic (*a*); dependences of R_{HRS} and R_{LRS} on the CVC voltage amplitude (*b*); dependences of I_{SET} and I_{RES} on the CVC voltage amplitude (*c*)

Additionally, an increase in the amplitude of the CVC voltage from 0.5 V to 1.5 V leads to an increase in I_{SET} and I_{RES} from 0.03×10^{-3} A to 0.92×10^{-3} A and 0.02×10^{-3} A to 0.81×10^{-3} A, respectively. The obtained result can be explained by an increase in the number of electrons due to an increase in the concentration of oxygen vacancies in the bulk of the ZnO film with an increase in the voltage amplitude.

An analysis of the experimental results of the multilevel resistive effect in a forming-free nanocrystalline zinc oxide film showed that the potentiation from 1200 cycles to 1500 cycles led to the film resistance increasing by 3 orders of magnitude. When the polarity of the applied voltage of the training pulse is reversed, the film resistance returns to the initial state from the 1800 cycle to the 1900 cycle. It was shown that potentiation occurs faster than depression because of the different free energies of the metal and the oxide.

Thus, it was shown that the forming-free nanocrystalline zinc oxide film successfully mimics biological memory through increased pulse number stimulation. Fig. 2,*b* shows that fixing the amplitude of the training pulses makes it possible to achieve different resistive states (synaptic weight levels), which is like the biological brain, in which memory changes as the stimulation increases (increasing the number of pulses).

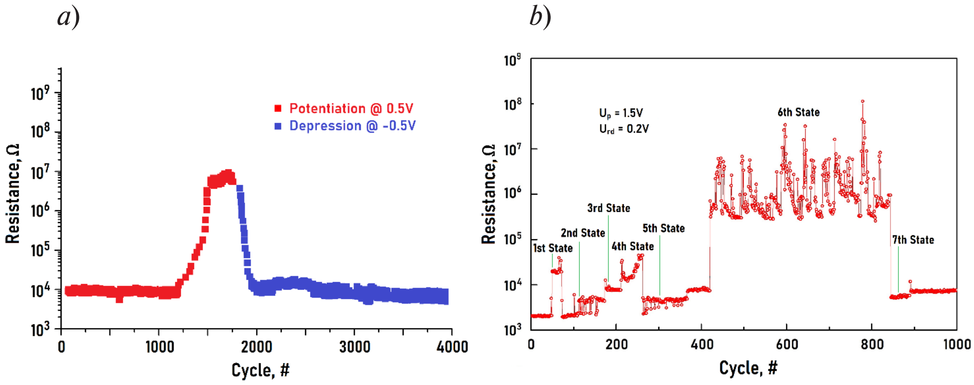

Fig. 2. Resistive switching in forming-free nanocrystalline ZnO film:
potentiation and depression (*a*); ZnO film stimulation by training pulses (*b*)

This analog-like behavior with multilevel states is desirable for improving the performance and reliability of a neuromorphic computing systems. The results obtained can be used to create ReRAM elements of neuromorphic artificial intelligence systems based on forming-free nanocrystalline ZnO films.

Conclusion

In this work, multilevel resistive switching in forming-free nanocrystalline ZnO films has been studied. Potentiation and depression at 0.5 V and –0.5 V for 3000 cycles led to the film resistance increasing by 3 orders of magnitude. It was shown that ZnO films successfully mimic biological memory through increased pulse number stimulation. Fixing the amplitude of the training pulses makes it possible to achieve different resistive states such as synaptic weight levels of the biological brain. The obtained results can be used for ReRAM elements of neuromorphic artificial intelligence systems fabrication based on forming-free nanocrystalline ZnO films.

REFERENCES

1. **Ismail M., Mahata C., Kim S.,** Forming-free Pt/Al_2O_3/HfO_2/HfA_lO_x/TiN memristor with controllable multilevel resistive switching and neuromorphic characteristics for artificial synapse, Journal of Alloys and Compounds. (892) (2022) 162141.

2. **Dongale T.D., Khot, A.C., Takaloo, A.V., Kim, T.G.,** Facile synthesis of nickel cobaltite quasi-hexagonal nanosheets for multilevel resistive switching and synaptic learning applications, NPG Asia Materials. 13(1) (2021) 1–12.

3. **Das B., Samanta M., Sarkar P., Ghorai U.K., Mallik A., Chattopadhyay K.K.,** Phthalocyanine (CuPc) Based Optoelectronic Memory Device with Multilevel Resistive Switching for Neuromorphic Application, Advanced Electronic Materials. 7(4) (2021) 2001079.

4. **Zhong T., Qin Y., Lv F., Qin H.,** Tian X. Light-activated multilevel resistive switching storage in Pt/Cs2AgBiBr6/ITO/glass devices, Nanoscale research letters. 16(1) (2021) 1–9.

5. **Saini M., Kumar M., Mandal R., Mitra A., Som T.,** Light-activated multilevel resistive switching storage in Pt/Cs2AgBiBr6/ITO/glass devices, Nanoscale research letters. 16(1) (2021) 1–9.

6. **Tominov R.V., Vakulov Z.E., Avilov V.I., Khakhulin D.A., Polupanov N.V., Smirnov V.A., Ageev O.A.,** The effect of growth parameters on electrophysical and memristive properties of vanadium oxide thin films, Molecules. 26(1) (2020) 118.

7. **Tominov R.V., Vakulov Z.E., Polupanov N.V., Saenko A.V., Avilov V.I., Ageev O.A., Smirnov V.A.,** The effect of growth parameters on electrophysical and memristive properties of vanadium oxide thin films, Molecules. 26(1) (2020) 118.

8. **Zou L.,** Multilevel resistive switching performance of TiO2-based flexible memory prepared by low-temperature sol-gel method with UV irradiation, Current Applied Physics. (24) (2021) 32–38.

9. **Swathi S.P., Angappane S.,** Low power multilevel resistive switching in titanium oxide-based RRAM devices by interface engineering, Journal of Science: Advanced Materials and Devices. 6(4) (2021) 601–610.

10. **Patil S.R., Mullani N.B., Kamble B.B., Tayade S.N., Kamat R.K., Park T.J., Dongale T.D.,** Forming-free and multilevel resistive switching properties of hydrothermally synthesized hexagonal molybdenum oxide microrods, Journal of Materials Science: Materials in Electronics. 32(9) (2021) 12490–12502.

11. **Tominov R., Avilov V., Vakulov Z., Khakhulin D., Ageev O., Valov I., Smirnov V.** Forming-Free Resistive Switching of Electrochemical Titanium Oxide Localized Nanostructures: Anodization, Chemical Composition, Nanoscale Size Effects, and Memristive Storage, Advanced Electronic Materials. (2022) 2200215.

12. **Singh R., Kumar M., Iqbal S., Kang H., Kim U., Park J. Y., Seo H.**, Electric Field-Induced Area Scalability toward the Multilevel Resistive Switching, Advanced Materials Interfaces. 8(17) (2021) 2100664.

13. **Shvetsov B.S., Minnekhanov A.A., Emelyanov A.V., Ilyasov A.I., Grishchenko Y.V., Zanaveskin M.L., Demin V.A.**, Parylene-based memristive crossbar structures with multilevel resistive switching for neuromorphic computing, Nanotechnology. 33(25) (2022) 255201.

14. **Ray R., Pal S.K.**, Solution-Processed Photoinduced Multilevel Resistive Switching Devices Based on Lead-Free All-Inorganic Perovskite, IEEE Electron Device Letters. 42(9) (2021) 1284–1287.

15. **Mahata C., Kim S.**, Modified resistive switching performance by increasing Al concentration in HfO_2 on transparent indium tin oxide electrode, Ceramics International. 47(1) (2021) 1199–1207.

16. **Fang S.L., Han C.Y., Liu W.H., Li X., Wang X.L., Huang X.D., Geng L.**, Multilevel resistive random access memory achieved by $MoO_3/Hf/MoO_3$ stack and its application in tunable high-pass filter, Nanotechnology. 32(38) (2021) 385203.

17. **Yun H.J., Ryu S.Y., Lee H.Y., Park W.Y., Kim S.G., Choi B.J.**, Multilevel operation of GdO_x-based resistive switching memory device fabricated by post-deposition annealing, Ceramics International. 47(12) (2021) 16597–16602.

18. **Zhong T., Qin Y., Lv F., Qin H., Tian X.**, Correction to: Light-Activated Multilevel Resistive Switching Storage in $Pt/Cs_2AgBiBr_6/ITO$/Glass Devices, Nanoscale Research Letters. 17(1) (2022) 1–1.

19. **Tominov R.V., Smirnov V.A., Avilov V.I., Fedotov A.A., Klimin V.S., Chernenko N.E.**, Formation of ZnO memristor structures by scratching probe nanolithography, IOP Conference Series: Materials Science and Engineering. – IOP Publishing. (443)1 (2018) 1.

20. **Tominov R.V., Vakulov Z.E., Avilov V.I., Khakhulin D.A., Fedotov A.A., Zamburg E.G., Ageev O.A.** Synthesis and memristor effect of a forming-free ZnO nanocrystalline films, Nanomaterials. 10(5) (2020) 1007.

21. **Smirnov V.A., Tominov R.V., Avilov V.I., Alyabieva N.I., Vakulov Z.E., Zamburg E.G., Ageev O.A.**, Investigation into the memristor effect in nanocrystalline ZnO films, Semiconductors. 53(1) (2019) 72–77.

THE AUTHORS

TOMINOV Roman V.
tominov@sfedu.ru
ORCID: 0000-0002-1263-2158

SHIKHOVTSOV Ivan A.
shihovcov@sfedu.ru
ORCID: 0000-0002-0355-9665

KHAKHULIN Daniil A.
dhahulin@sfedu.ru
ORCID: 0000-0002-0058-0274

VAKULOV Zakhar E.
zvakulov@sfedu.ru
ORCID: 0000-0003-3084-4522

SMIRNOV Vladimir A.
vasmirnov@sfedu.ru
ORCID: 0000-0001-7130-2194

Received 28.10.2022. Approved after reviewing 08.11.2022. Accepted 10.11.2022.