

Effect of training pulse parameters on the synaptic plasticity of a ZrO₂(Y)-based memristive device

Maria N. Koryazhkina, Margarita A. Ryabova, Evgeniya V. Okulich, Alexey I. Belov, Ivan N. Antonov, Maria E. Shenina, Sergey A. Shchanikov, Alexey N. Mikhaylov, Dmitrii O. Filatov, Davide Valenti, and Bernardo Spagnolo*

Dr. M. N. Koryazhkina, M. A. Ryabova, E. V. Okulich, Dr. A. I. Belov, I. N. Antonov, Dr. M. E. Shenina, Dr. S. A. Shchanikov, Dr. A. N. Mikhaylov, Dr. D. O. Filatov
Lobachevsky State University of Nizhny Novgorod, 23 Gagarin Ave. Nizhny Novgorod 603022 Russia
E-mail: mahavenok@mail.ru

Dr. D. Valenti
Dipartimento di Fisica e Chimica “Emilio Segrè”, Group of Interdisciplinary Theoretical Physics, Università degli Studi di Palermo, Viale delle Scienze, Edificio 18, I-90128 Palermo, Italy

Prof. B. Spagnolo
Lobachevsky State University of Nizhny Novgorod, 23 Gagarin Ave. Nizhny Novgorod 603022 Russia
Dipartimento di Fisica e Chimica “Emilio Segrè”, Group of Interdisciplinary Theoretical Physics, Università degli Studi di Palermo, Viale delle Scienze, Edificio 18, I-90128 Palermo, Italy

Keywords: resistive switching, memristor, potentiation, depression, yttria-stabilized zirconia, neuromorphic system

The effect of training pulse parameters on the synaptic plasticity of a ZrO₂(Y)-based memristive device has been investigated. It is shown that the potentiation and depression significantly depend on the amplitude and shape of the training pulses. The most stable synaptic plasticity is observed when considering training pulses with rectangular shape and maximum amplitude value. On the other hand, there is a rather weak dependence of the potentiation and depression on the number of pulses in trains. The observed effect can be

explained in terms of the work required to change the resistive state. Moreover, a $\text{ZrO}_2(\text{Y})$ -based memristive device exhibits distinguishable potentiation and depression for at least 1000 cycles.

1. Introduction

Nowadays, neuromorphic systems have drawn significant interest.^[1,2] Research and development of neuromorphic devices were inspired by human brain that is able to process energy-efficiently a large amount of information ($\sim 1\text{--}100$ fJ per synaptic event).^[2,3] The promising candidate for emulating brain's synaptic functions, such as learning, forgetting and memorizing, is a memristor as a part of neuromorphic circuit. Memristive devices are based on the resistive switching effect, that is the change of conductivity under the external electric field, and recently largely investigated.^[4–7] Memristive devices have multilevel and incremental changes in conductance whereas the continuous change in the weight of a biological synapse is a fundamental process for transmitting and processing information in human brain.^[8–10] Therefore, in neuromorphic devices, the memristor is assessed as an artificial synapse, and the change in its conductivity is considered as synaptic plasticity which plays the crucial role in the brain's memory function.^[11] It has already been shown that the increase and decrease of the electrical conductance of the memristor correspond respectively to the potentiation and depression in a robust biological synapse.^[12,13] However, there are some serious problems regarding the synaptic plasticity. One of the problems is the dependence of the training accuracy of neuromorphic networks on the degree of conduction nonlinearity in potentiation and depression.^[14] At the same time, memristive devices as part of neuromorphic systems must be resistant to failure, which means a long endurance without significant degradation in performance.^[15] According to recent investigations, the problem solving method includes both optimization of training protocols to modify the resistive state of the memristor and the component materials, the latter means the correct choice of electrode materials and functional dielectric.^[16] Focusing on the former, changes in pulse amplitudes and duration, as well as the intervals between pulses, are taken into account in studying the influence of the switching protocol on synaptic plasticity.^[17–20]

This work considers the effect of training pulse parameters (shape, amplitude and number of training pulses in trains) on potentiation and depression in the $\text{ZrO}_2(\text{Y})$ -based memristive device.

We chose a memristive device based on the Ta/ $\text{ZrO}_2(\text{Y})$ /Pt stack due to their low variation in resistive switching parameters from both cycle-to-cycle and device-to-device.^[21] Furthermore,

it has previously been shown that these memristor devices demonstrate stability in the selected range of low voltages, indicating new perspectives for their use as low-power elements for memory and neuromorphic systems.^[21]

2. Experimental Methods

A memristive device based on the Ta/ZrO₂(Y)/Pt stack (see **Figure 1**) was fabricated using a Torr International 2G1-1G2-EB4-TH1 thin film deposition vacuum system. The ZrO₂(Y) film was deposited by radio-frequency magnetron sputtering of a target, which consisted of ZrO₂ (88 mol %) and Y₂O₃ (12 mol %) powders. The substrate temperature during ZrO₂(Y) deposition was 250 °C. The metal layers were deposited by direct current magnetron sputtering at a substrate temperature of 200 °C. A standard photolithography facility was used to form the cross-point topology. The active area of memristive device was 20 × 20 μm².

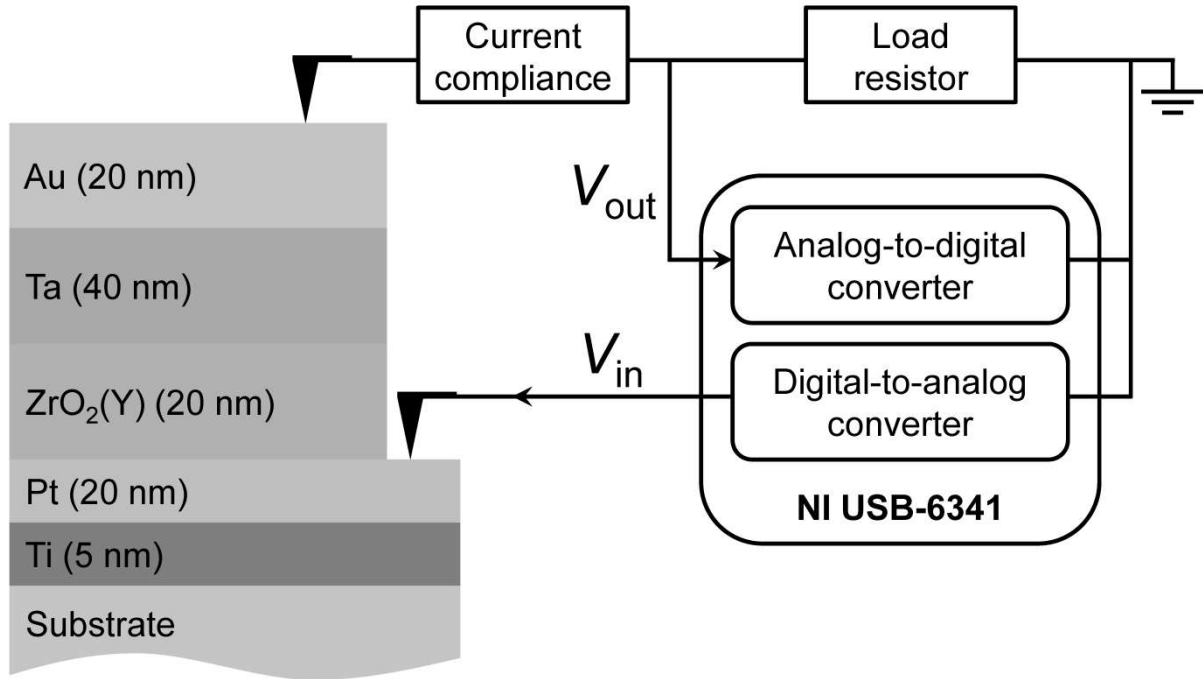


Figure 1. Schematic representation of a memristive device and an experimental setup

Investigations of the effect of training pulse parameters on the potentiation and depression were carried out using USB-6341 data acquisition system (National Instruments). A schematic representation of the experimental setup is shown in Figure 1. The electrical contacts to the contact pads of the memristive device were provided using the Everbeing EB-6 probe station. The sign of voltage across the memristive device corresponds to the potential of bottom electrode relative to the potential of top electrode (see Figure 1). Current compliance

is 300 μA and is based on a simple field-effect transistor circuit.^[22] All investigations were carried out on the same memristive device at room temperature.

To study potentiation and depression, a signal was applied to the memristive device, consisting of trains of N identical pulses (in amplitude and shape) with fixed positive amplitude A and N identical pulses with fixed negative amplitude $-A$. See the schematic representation of a cycle of the input signal in **Figure 2**, where input signals with rectangular and exponential training pulses are shown. We used training pulses of different amplitudes $|A|$ (0.9–1.2 V) and shapes (exponential, triangular, sinusoidal and rectangular) and varied the number of training pulses N in the trains ($N = 10, 20, 50$ and 100). As will be shown below, the device demonstrated several resistive states when an increasing voltage, in absolute value, was applied (see **Figure 4**). However, the detected effect turned out to be more significant in the positive voltage region: each subsequent voltage increase in the range from +0.9 to +1.2 V resulted in a significant increase, compared to the curves obtained in other cases of voltage, resistance of the device. Based on these results, the training pulse amplitude range was selected. The training pulse duration was 5.5 μs . After each training pulse, the current through device was measured by applying reading pulses with an amplitude of +0.4 V and a duration of 0.1 ms. The response of memristive device to the input signal was recorded from a load resistor (100 Ω) connected in series (Figure 1). To determine the effect of the input signal parameters on the potentiation and depression, the average (over 100 data points) value of current during the reading was calculated (see **Figure 5** for the explanation).

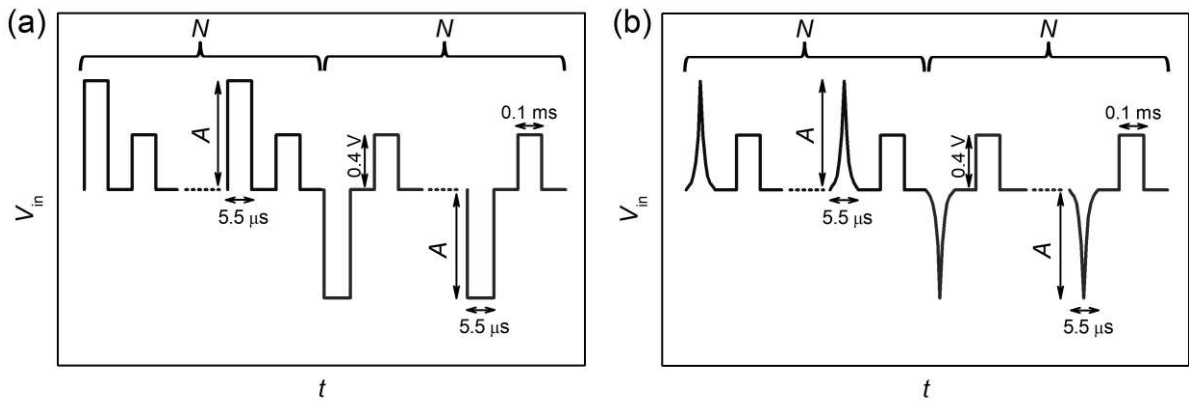


Figure 2. Schematic representation of one cycle of the input signal with (a) rectangular and (b) exponential training pulses

3. Results and Discussion

The investigated memristive device exhibits stable and predominantly gradual I - V curves of bipolar switching type (**Figure 3a**). The average threshold voltages for SET (transition from

HRS into LRS) and RESET (transition from LRS into HRS) processes are -0.7 and $+0.7$ V, respectively. These values almost coincide with those obtained earlier for a $\text{ZrO}_2(\text{Y})$ -based memristive devices.^[21] The dependence of current through the investigated device on the number of switching cycles indicates that the difference between resistive states persists over 100 switching cycles (see Figure 3b). Moreover, on average, the resistive states differ by more than an order of magnitude. The coefficient of variance, which is the ratio of standard deviation to its mean, for currents through device in LRS and HRS are 0.19 and 0.49, respectively. These values are different from those previously obtained.^[21] Note, however, that the previously obtained results are calculated from pulse measurements. In any case, a trend can be seen that the variation in the case of LRS is less pronounced than in the case of HRS.

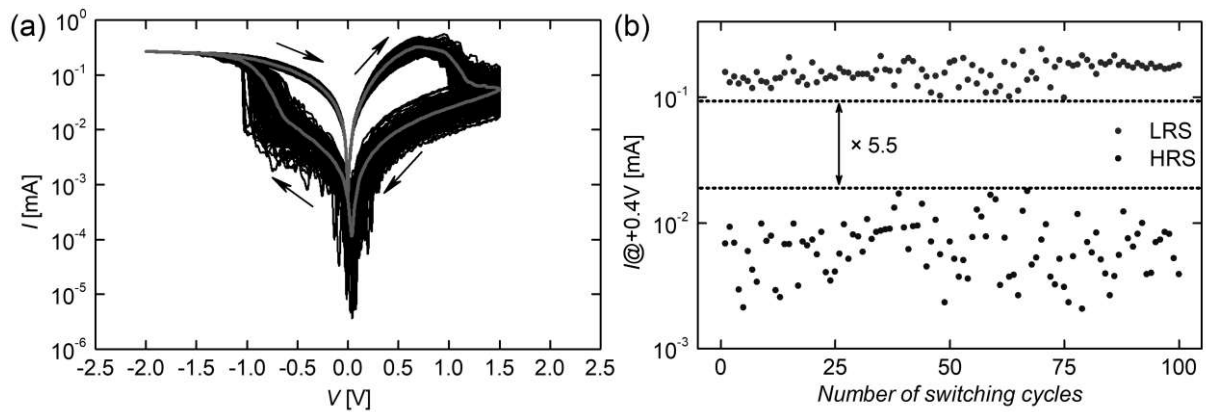


Figure 3. (a) One hundred I - V curves (black) and averaged I - V curve (pink) for the device under study; (b) dependence of the current through the device at $+0.4$ V on the number of switching cycles obtained from I - V curves

The studied device demonstrates asymmetrical synaptic behavior (see Figure 4), in particular with one voltage polarity, in which the resistive state change is gradual, and with the other voltage polarity in which this change is more abrupt.

A typical response of a memristive device to one cycle of the input signal with rectangular training pulses with $|A| = 1.1$ V and $N = 20$ is shown in Figure 5a. The response of a memristive device is a waveform of the current flowing through it. As a result of a train of pulses with a fixed positive amplitude, the resistance of memristive device gradually increases. It can be seen that the current through device fluctuates during reading, but generally decreases.

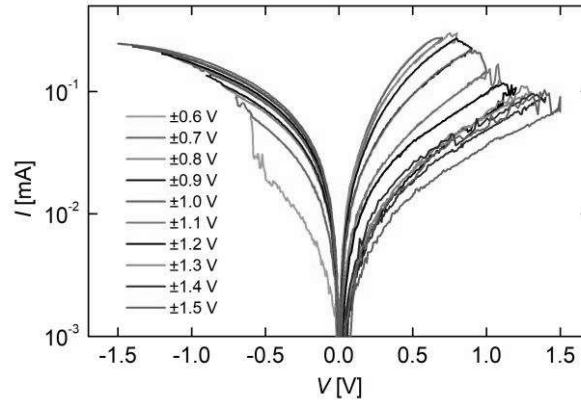


Figure 4. Asymmetric synaptic behavior of the memristive device under study

And vice versa, due to the effect of a train of pulses with a fixed negative amplitude, the resistance of memristive device gradually decreases. It can be seen that the current through device increases during reading. This behavior is similar to depression and potentiation, that is the ability of a synapse to change its strength.^[8] Note that for both polarities of the training pulse amplitude, the current when reading after the first and twentieth (last) training pulse differs approximately 1.3 times.

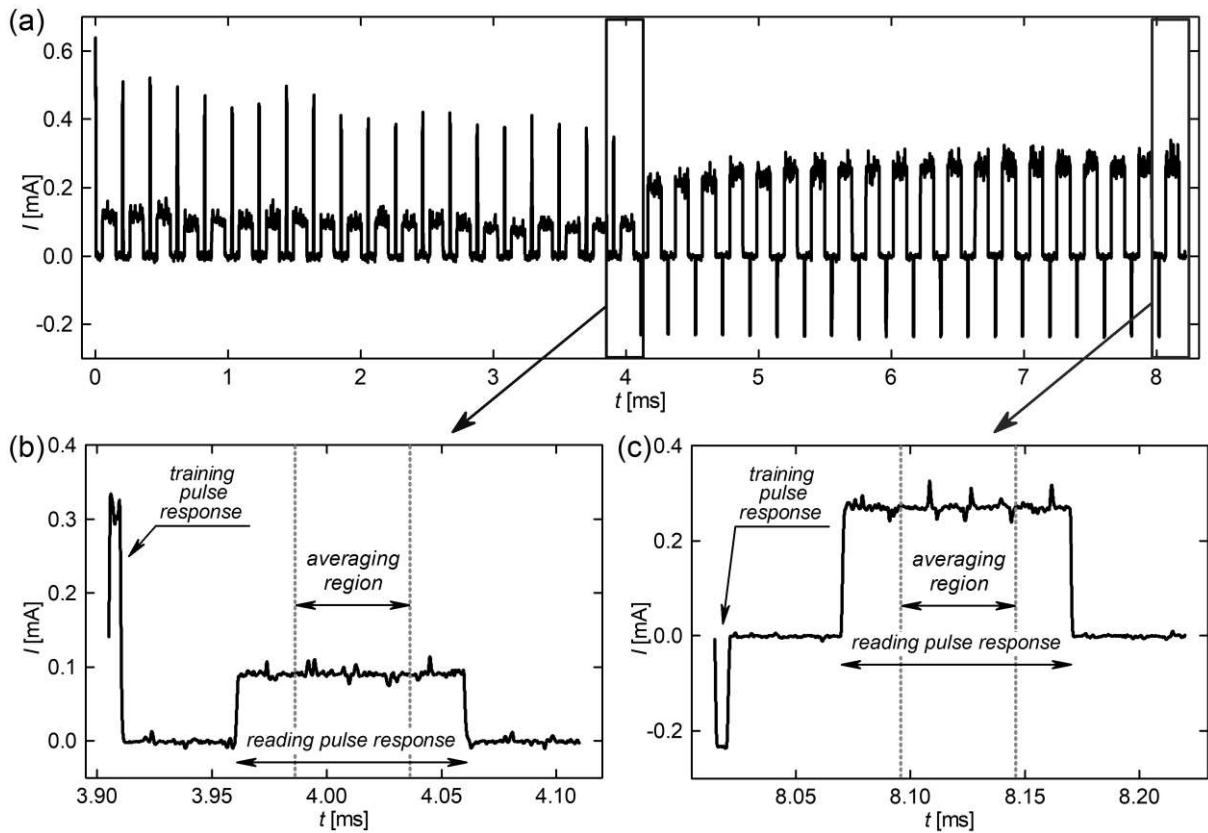


Figure 5. (a) Typical waveform of current through a memristive device obtained by applying one cycle of the input signal. Device response to the last training pulse of (b) positive or (c)

negative amplitude and reading after it. The data are given for the case of the input signal with rectangular training pulses with $|A| = 1.1$ V and $N = 20$

Figure 5 shows the response of the memristive device to the last training pulse of positive (Figure 5b) and negative (Figure 5c) fixed amplitude and the reading pulse after it. Note that, for the case under consideration (rectangular training pulses with $|A| = 1.1$ V and $N = 20$), the value of currents when reading differs by ~ 3 times. The value of current during the last reading (in sequence of positive/negative training pulses) was used as a measure of the potentiation and depression effectiveness. In other words, this value allows us to understand if the resistive state has changed a lot and if it has changed completely.

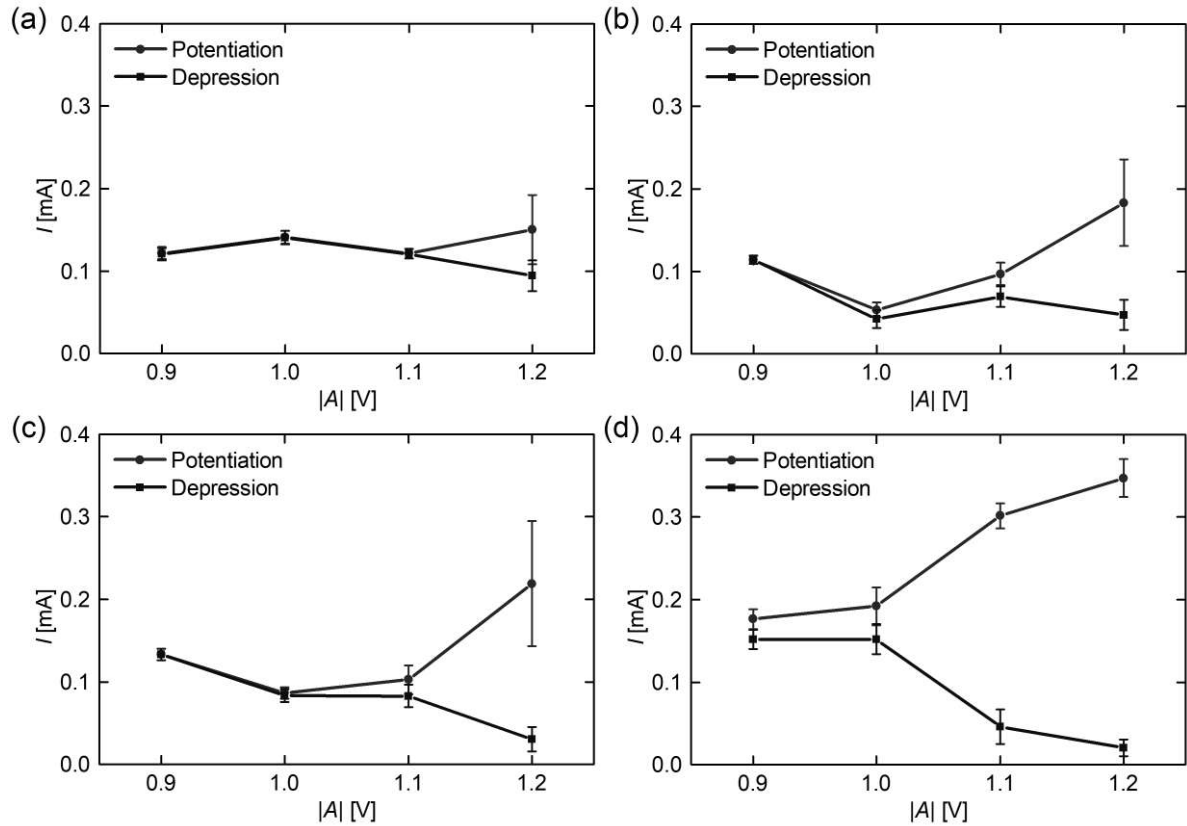


Figure 6. Dependence of the average current value on the amplitude of training pulses with different shapes: (a) exponential, (b) triangular, (c) sinusoidal, and (d) rectangular. Data are given for $N = 50$

Figure 6 and **Figure 7** show the dependence of current value during the last reading on the amplitude (Figure 6) and the number (Figure 7) of training pulses of various shapes. The average was performed over 500 potentiation and depression cycles. The error bars are the standard deviation of the mean current.

Let us first consider the effect of exponential, triangular and sinusoidal pulses with a fixed $N = 50$ (Figure 6a, 6b, and 6c) on the response of the memristive device and, accordingly, the current value during the last reading. It can be noted that, for an amplitude equal to 0.9–1.1 V (for exponential pulses) or 0.9–1.0 V (for triangular and sinusoidal pulses), the average values of the potentiation and depression currents during the last reading differ slightly or not at all. In these cases, there is little or no response to the input signal over 500 cycles of potentiation and depression. This indicates the absence of changes in the resistive state of the device under the effect of trains of positive and negative training pulses. With an increase in the amplitude of the exponential pulses to 1.2 V, the average values of the potentiation and depression currents during the last reading differ by a factor of 1.6, but their standard deviations overlap. In this case, the memristive device demonstrates a response to each cycle of the input signal during the first 282 cycles of potentiation and depression. During the remaining 218 cycles, the resistive state of the device remains almost unchanged when exposed to trains of positive and negative training pulses. With an increase in the amplitude of triangular and sinusoidal pulses to 1.1 V, the average values of potentiation and depression currents during the last reading differ by 1.4 and 1.2 times, respectively. In these cases there is an unstable response (not for each cycle of the input signal) of memristive device and significant current fluctuations during the reading from cycle to cycle of potentiation and depression are observed. An increase in the amplitude of triangular and sinusoidal pulses to 1.2 V result in a significant difference in the average values of potentiation and depression currents during the last reading and the absence of overlapping of their standard deviations: the average values differ by 3.9 and 7.1 times, respectively. In these cases, the memristive device exhibits the response to almost every cycle of the input signal (for all 500 cycles). Then, we consider the effect of rectangular pulses with a fixed $N = 50$ (Figure 6d) on the response of memristive device and, consequently, the current value during the last reading. Note that, over the entire considered range of rectangular pulse amplitudes, the overlap of standard deviations of potentiation and depression currents is absent or insignificant. For a rectangular pulse amplitude of 0.9 and 1.0 V, the mean values of potentiation and depression currents during the last reading differ by 1.2 and 1.3 times, respectively. In these cases, an unstable response of the device and significant fluctuations of currents during the reading from cycle to cycle of potentiation and depression are observed. An increase in the amplitude of rectangular pulses to 1.1 and 1.2 V leads to a significant difference in the average values of potentiation and depression currents during the last reading: the average values differ by 6.5 and 17.3 times, respectively. In these cases, the stable response of memristive device to each cycle of the

input signal over all 500 cycles and slight fluctuations of currents during reading from cycle to cycle of potentiation and depression are observed. Note that the maximum value of the ratio of the average values of potentiation and depression currents during the last reading at the same amplitude value is demonstrated by the devices under the effect of training pulses of exactly a rectangular shape. Therefore, the results obtained indicate a strong dependence of the value of the potentiation and depression currents during the last reading on the amplitude and shape of training pulses.

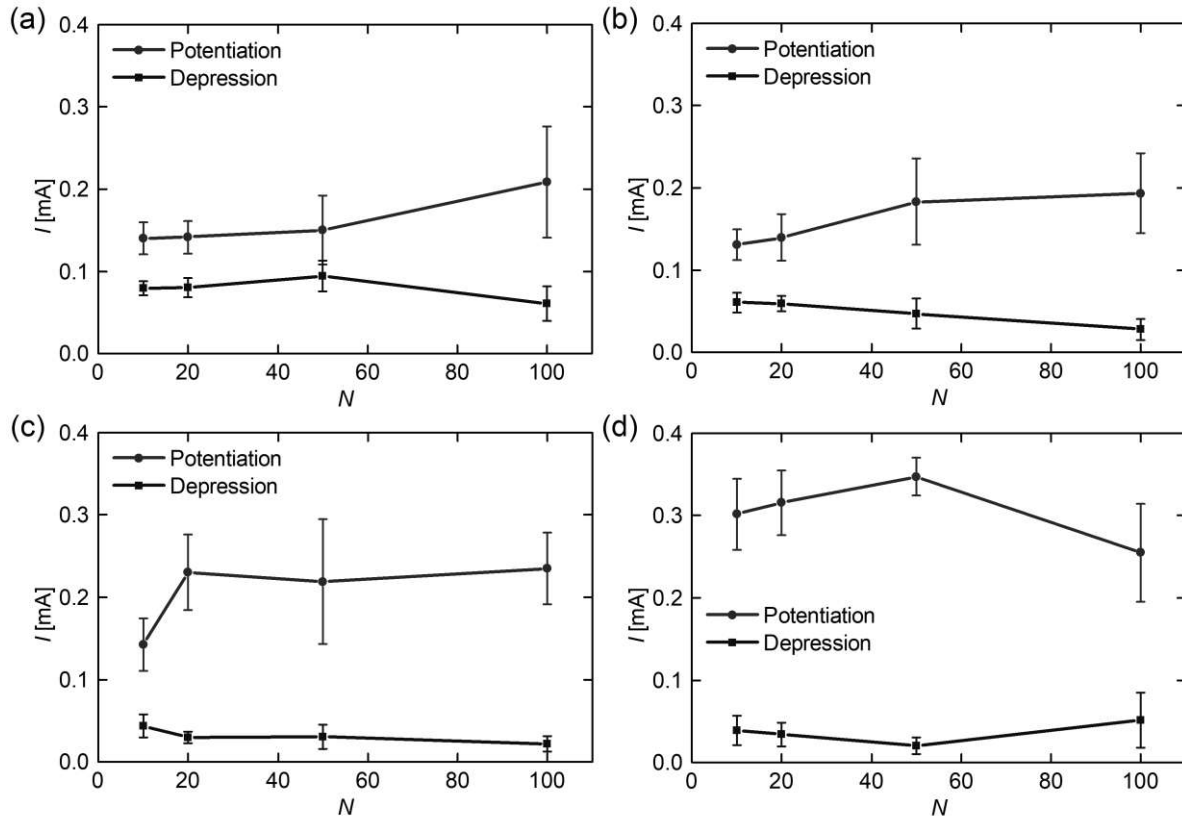


Figure 7. Dependence of the average current value on the number of training pulses with different shapes: (a) exponential, (b) triangular, (c) sinusoidal, and (d) rectangular. Data are given for $|A| = 1.2$ V

On the contrary, the number of training pulses has a less significant effect on the potentiation and depression (see Figure 7). It can be seen that, almost in the entire considered range of N , the overlap of the standard deviations of potentiation and depression currents is absent or insignificant. In any case, it can be noted that, for a fixed value of $|A| = 1.2$ V, the maximum ratio of the average values of potentiation and depression currents during the last reading is observed in the case of rectangular pulses with $N = 50$. The observed effect could be ascribed to the value of work required to change the resistive state. This work is determined by at least the amplitude, shape, duration and number of training pulses in trains.

Figure 8 demonstrates the dependence of the current through device on the number of potentiation / depression cycles. It is shown that potentiation and depression are distinguishable with at least a triple current variation even after reaching 1000 cycles. Note that the values of potentiation currents almost do not change and have a constant value. This behavior is undesirable, because this will significantly reduce the accuracy of training neuromorphic networks based on such devices. However, in this study, symmetrical amplitudes in absolute value of the training pulses were used for potentiation and depression. The effect of asymmetry will be the subject of further research.

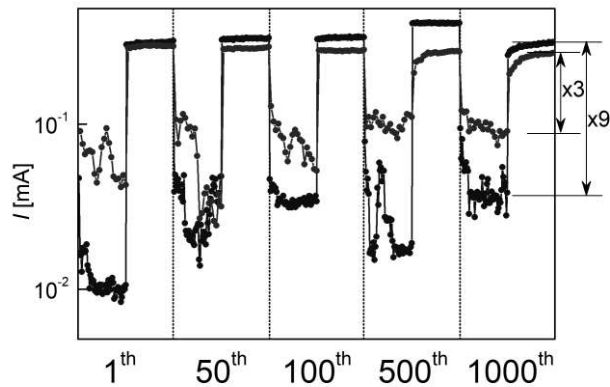


Figure 8. Dependences of the current through device on the number of potentiation / depression cycles. Data show the currents during the reading for rectangular training pulses with $|A| = 1.2$ V and $N = 50$ (blue), as well as with $|A| = 1.1$ V and $N = 20$ (red)

4. Conclusion

The effect of training pulse parameters (shape, amplitude and number of training pulses in trains) on synaptic plasticity (potentiation and depression) of a $\text{ZrO}_2(\text{Y})$ -based memristive device was studied. It is shown that potentiation and depression are significantly dependent on the shape and amplitude of training pulses and slightly on the number of pulses in the trains.

The observed effect can be explained in terms of the work required to change the resistive state. Considering the presence of a response to each cycle of the input signal and the smallest fluctuations in the current value, training pulses of rectangular shape and maximum amplitude value are found the most effective to control synaptic plasticity. The presented results indicate the promise of further potentiation and depression studies in $\text{ZrO}_2(\text{Y})$ -based memristive devices.

The study of the effect of the amplitude and number of training pulses of various shapes in a wider range of values, as well as the development of a physical model of this effect on the $\text{ZrO}_2(\text{Y})$ -based memristive devices will be the subject of further research.

Acknowledgements

The work was carried out as part of the implementation of the “Priority-2030” Academic Excellence Program of the Lobachevsky State University of Nizhny Novgorod (N-466-99_2021-2023) and state assignment for the creation of new laboratories for the electronics industry. Koryazhkina M.N. expresses her gratitude for the support of the program of scholarships under the President of the Russian Federation to young scientists and graduate students who carry out promising research and development in priority areas of modernization of the Russian economy. The studies were carried out using shared research facilities of Research and Educational Center for Physics of Solid State Nanostructures at Lobachevsky State University of Nizhny Novgorod.

Conflict of Interest

The authors declare that they have no conflict of interest.

Received: ((will be filled in by the editorial staff))

Revised: ((will be filled in by the editorial staff))

Published online: ((will be filled in by the editorial staff))

References

- [1] L. Wang, Z. Wang, J. Lin, J. Yang, L. Xie, M. Yi, W. Li, H. Ling, C. Ou, W. Huang, *Sci. Rep.* **2016**, *6*, 35273.
- [2] Y. van de Burgt, E. Lubberman, E. J. Fuller, S. T. Keene, G. C. Faria, S. Agarwal, M. J. Marinella, A. A. Talin, A. Salleo, *Nat. Mater.* **2017**, *16*, 414.
- [3] I. A. Surazhevsky, V. A. Demin, A. I. Ilyasov, A. V. Emelyanov, K. E. Nikiruy, V. V. Rylkov, S. A. Shchanikov, I. A. Bordanov, S. A. Gerasimova, D. V. Guseinov, N. V. Malekhonova, D. A. Pavlov, A. I. Belov, A. N. Mikhaylov, V. B. Kazantsev, D. Valenti, B. Spagnolo, M. V. Kovalchuk, *Chaos Solitons & Fractals* **2021**, *146*, 110890.
- [4] A. V. Yakimov, D. O. Filatov, O. N. Gorshkov, D. A. Antonov, D. A. Liskin, I. N. Antonov, A. V. Belyakov, A. V. Klyuev, A. Carollo, B. Spagnolo, *Appl. Physics Lett.* **2019**, *114*, 253506.
- [5] D. O. Filatov, D. V. Vrzheschch, O. V. Tabakov, A. S. Novikov, A. I. Belov, I. N. Antonov, V. V. Sharkov, M. N. Koryazhkina, A. N. Mikhaylov, O. N. Gorshkov, A. A. Dubkov, A. Carollo, B. Spagnolo, *J. Stat. Mech. Theory Exp.* **2019**, *2019*, 124026.

- [6] N. V. Agudov, A. V. Safonov, A. V. Krichigin, A. A. Kharcheva, A. A. Dubkov, D. Valenti, A. N. Mikhailov, A. Carollo, B. Spagnolo, *J. Stat. Mech. Theory Exp.* **2020**, 2020, 024003.
- [7] A. N. Mikhaylov, D. V. Guseinov, A. I. Belov, D. S. Korolev, V. A. Shishmakova, M. N. Koryazhkina, D. O. Filatov, O. N. Gorshkov, D. Maldonado, F. J. Alonso, J. B. Roldán, A. V. Krichigin, N. V. Agudov, A. A. Dubkov, A. Carollo, B. Spagnolo, *Chaos Solitons & Fractals* **2021**, 144, 110723.
- [8] S. Deswal, A. Kumar, A. Kumar, *AIP Adv.* **2019**, 9, 095022.
- [9] Yu. V. Ushakov, A. A. Dubkov, B. Spagnolo, *Phys. Rev. E* **2010**, 81, 041911.
- [10] Yu. V. Ushakov, A. A. Dubkov, B. Spagnolo, *Phys. Rev. Lett.* **2011**, 107, 108103.
- [11] S.-Y. Min, W.-J. Cho, *Int. J. Mol. Sci.* **2021**, 22, 773.
- [12] C. Zamarreño-Ramos, L. A. Camuñas-Mesa, J. A. Pérez-Carrasco, T. Masquelier, T. Serrano-Gotarredona, B. Linares-Barranco, *Front. Neurosci.* **2011**, 5, 26.
- [13] D. Kuzum, R. G. D. Jeyasingh, B. Lee, H.-S. P. Wong, *Nano Lett.* **2012**, 12, 2179.
- [14] H. Liu, M. Wei, Y. Chen, *Nanotechnol. Rev.* **2018**, 7, 443.
- [15] S. H. Jo, T. Chang, I. Ebong, B. B. Bhadviya, P. Mazumder, W. Lu, *Nano Lett.* **2010**, 10, 1297.
- [16] S. Li, B. Li, X. Feng, L. Chen, Y. Li, L. Huang, X. Fong, K.-W. Ang, *NPJ 2D Mater. Appl.* **2021**, 5, 1.
- [17] Y.-F. Wang, Y.-C. Lin, I.-T. Wang, T.-P. Lin, T.-H. Hou, *Sci. Rep.* **2015**, 5, 10150.
- [18] X. Yan, C. Qin, C. Lu, J. Zhao, R. Zhao, D. Ren, Z. Zhou, H. Wang, J. Wang, L. Zhang, X. Li, Y. Pei, G. Wang, Q. Zhao, K. Wang, Z. Xiao, H. Li, *ACS Appl. Mater. Interfaces* **2019**, 11, 48029.
- [19] Z. Zhou, X. Yan, J. Zhao, C. Lu, D. Ren, N. Lu, J. Wang, L. Zhang, X. Li, H. Wang, M. Zhao, *J. Mater. Chem. C* **2019**, 7, 1561.
- [20] M. Ismail, H. Abbas, C. Choi, S. Kim, *Appl. Surf. Sci.* **2020**, 529, 147107.
- [21] M. N. Koryazhkina, D. O. Filatov, V. A. Shishmakova, M. E. Shenina, A. I. Belov, I. N. Antonov, V. E. Kotomina, A. N. Mikhaylov, O. N. Gorshkov, N. V. Agudov, C. Guarcello, A. Carollo, B. Spagnolo, *Chaos Solitons Fractals* **2022**, 162, 112459.
- [22] A. Mikhaylov, A. Belov, D. Korolev, I. Antonov, V. Kotomina, A. Kotina, E. Gryaznov, A. Sharapov, M. Koryazhkina, R. Kryukov, S. Zubkov, A. Sushkov, D. Pavlov, S. Tikhov, O. Morozov, D. Tetelbaum, *Adv. Mater. Technol.* **2020**, 5, 1900607.

The table of contents

The effect of training pulse parameters on the synaptic plasticity of a $\text{ZrO}_2(\text{Y})$ -based memristive device is investigated. A strong dependence of the device potentiation and depression in the aggregate on the shape and amplitude of training pulses is found. The device demonstrates distinguishable potentiation and depression for at least 1000 cycles.

M. N. Koryazhkina*, M. A. Ryabova, E. V. Okulich, A. I. Belov, I. N. Antonov, M. E. Shenina, S. A. Shchanikov, A. N. Mikhaylov, D. O. Filatov, D. Valenti, and B. Spagnolo

Effect of training pulse parameters on the synaptic plasticity of a $\text{ZrO}_2(\text{Y})$ -based memristive device

