Resonant Activation of Resistive Switching in ZrO$_2$(Y) Based Memristors

V. N. Baranova$^a$,* D. O. Filatov$^a$,** D. A. Antonov$^a$***, I. N. Antonov$^a$****, and O. N. Gorshkov$^a$*****

$^a$Lobachevskii State University of Nizhni Novgorod, Nizhni Novgorod, 603950 Russia

*e-mail: vera.baranova00@mail.ru
**e-mail: dmitry_filatov@inbox.ru
***e-mail: antonov_dm@inbox.ru
****e-mail: ivant@nifti.unn.ru
*****e-mail: gorshkov@nifti.unn.ru

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Abstract—We report on a comparative study of resistive switching in the memristors based on ZrO$_2$(Y) films and on ZrO$_2$(Y)/Ta$_2$O$_5$ bilayer stacks by triangle voltage pulses with superimposed high-frequency sinusoidal signal. The dependencies of the current difference in the low resistance state and in the high resistance one on the sinusoidal signal frequency for the ZrO$_2$(Y)-based memristor and for the ZrO$_2$(Y)/Ta$_2$O$_5$-stack based one manifested one and two maxima, respectively attributed to the resonant activation of the migration of the oxygen ions via the oxygen vacancies by the alternating external electric field in ZrO$_2$(Y) and in Ta$_2$O$_5$.

Keywords: resistive switching, memristor, stability, resonant activation, yttria stabilized zirconia

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1. INTRODUCTION

Recently, resistive switching (RS) attracted much attention [1]. The effect consists in bistable (multistable) switching of the resistance of a dielectric film between two conductive electrodes under a voltage applied to the ones. Respective devices are called memristors [2]. The RS mechanism in metal oxides consists in forming of conductive filaments (composed of oxygen vacancies, VO$_\text{S}$) in an electric field between the electrodes [3]. The switching from a low resistance state (LRS) to a high resistance state (HRS) (RESET process) is achieved by the rapture of the filament by a voltage pulse of certain polarity. The filament is restored by another voltage pulse of the opposite polarity that results in the switching from the HRS to LRS (SET process).

The memristors are promising for applications in the non-volatile computer memory [4], the neuromorphic computing [5], etc. However, their application is limited by instability of the RS parameters, which is an intrinsic property of the memristors originating from the stochastic nature of the RS since a limited number of VO$_\text{S}$ near the filament tip participate in the switching process [6]. Several approaches to improving the RS stability were proposed, e.g. a choice of suitable dielectric and electrode materials, the multilayered dielectrics, etc. [7]. Another approach is based on noise-enhanced stability effect [8], which is inherent to the multistable stochastic systems [9]. Earlier, we found the high-frequency (HF) sinusoidal signal added to the switching pulses to improve the RS performance of ZrO$_2$(Y)-based memristors [10, 11]. The effect was attributed to the resonant activation of the O$^{2-}$ ion motion via the VO$_\text{S}$ in the alternating electric field, which promotes the rapture and restoring of the filaments. Resonant activation of the transitions between the metastable states is another phenomenon manifesting the beneficiary role of noise [12].

In the present work, we studied effect of the frequency of the HF sinusoidal signal added to the switching pulses on the RS parameters of the memristors based on ZrO$_2$(Y) films and on ZrO$_2$(Y)/Ta$_2$O$_5$ stacks. The Ta$_2$O$_5$ sublayers were found to improve the RS because of self-forming of the Ta-enriched nanoinclusions in the Ta$_2$O$_5$ layers, which play the role of the electric field concentrators promoting the filament growth [13]. The goal of the present study was to confirm resonant activation of the O$^{2-}$ ion motion to be the origin of the RS improvement.
2. EXPERIMENT

The functional dielectric films were deposited by HF magnetron sputtering at the substrate temperature $T_s = 300^\circ$C onto the industrial Si(001) substrates with pre-deposited SiO$_2$ film ($\sim$500 nm thick), Ti adhesion layer, and TiN conductive one (each $\sim$25 nm in thickness) using Torr International® 2G1-1G2-EB4-TH1 setup. The thickness of the ZrO$_2$(Y) ($\sim$12 mol % of Y$_2$O$_3$) films was $\sim$40 nm, the one of the Ta$_2$O$_5$ sublayers was $\sim$10 nm. The top Au(40 nm)/Zr(7 nm) contacts were deposited by direct current magnetron sputtering at $T_s = 200^\circ$C. The cross-point memristors with the active area of 20 $\times$ 20 $\mu$m$^2$ were fabricated by standard optical lithography. The RS was studied using a setup shown schematically in Fig. 1.

The voltage $V(t)$ ($t$ is the time) applied to the memristor was generated by a digital-to-analog converter (DAC) of NT-MDT® Solver Pro™ atomic force microscope (AFM) controller used as a programmable voltage source. Current compliance during the SET process was provided by a field-effect transistor. RS parameters were measured by multiple write/erase cycling according to protocol shown qualitatively in Fig. 2.

The switching of the memristor was performed by triangular switching pulses with the amplitudes $V_{SET} = 3.5$ to 4.5 V and $V_{RESET} = -3.5$ to $-2.5$ V and the durations $T_{SET}, T_{RESET} = 1$ s. Here $V_{SET}$ and $V_{RESET}$ are the switching voltages from HRS to LRS and back from LRS to HRS, respectively. The values of the current through the memristor in the LRS and HRS $I_{ON}$ and $I_{OFF}$, respectively were recorded at the read voltage $V_{READ} = 2$ V and averaged over $N_{READ} = 20$ samplings.

The HF sinusoidal signal pulses generated by the built-in oscillator of NT-MDT® SolverPro™ AFM controller with amplitude $A = 0.2$ V, duration $T_m = 0.1$ s, and frequency $f$ varied from 100 Hz to 25 kHz were superimposed onto the tops of the triangle switching pulses. The measured values of $I_{ON}$ and $I_{OFF}$ were averaged over 100 write/erase switching cycles.

3. RESULTS AND DISCUSSION

The frequency dependence of the difference $I_{ON} - I_{OFF}$ for the ZrO$_2$(Y)-based memristor (Fig. 3) manifested a maximum at $f \sim 2.5$ kHz that corresponds to the frequency of the O$_2^-$ ion jumps to the nearest neighboring VO$_2$ in ZrO$_2$(Y) at 300 K ranging from 0.4 to 8 kHz [14]. This observation confirmed the resonant activation of the O$_2^-$ ion migration to be the origin of the improvement of the RS performance when adding the HF signal to the switching pulses observed earlier [10, 11].

The frequency dependence of the difference $I_{ON} - I_{OFF}$ for the memristor based on the ZrO$_2$(Y)/Ta$_2$O$_5$ bilayered stack (Fig. 3) manifested two maxima: one at higher frequency $f \approx 6.5$ kHz and another one at lower frequency $f \approx 400$ Hz. The maximum at higher frequency can be attributed to the resonant activation of the O$_2^-$ ion migration in the ZrO$_2$(Y) layer whereas the low-frequency maximum can be ascribed to the resonant activation effect in the Ta$_2$O$_5$ sublayer. One can estimate the activation energy for the thermally activated jumps of the O$_2^-$ ions onto the adjacent V$_{O9}$ in the Ta$_2$O$_5$ sublayer $E_a$ from the corresponding maximum frequency $f_i$ according to the formula

$$f_i \sim f_0 \exp\left(-\frac{E_a}{kT}\right), \quad (1)$$
where $f_0 \sim 10^{13}$ Hz is typical lattice vibration frequency, $k$ is Boltzmann constant, and $T$ is the temperature. For $f_i = 1$ kHz one obtains $E_a \approx 0.6$ eV at $T = 300$ K. This value is approximately two times smaller than the activation energy of the oxygen diffusion in polycrystalline Ta$_2$O$_5$ at elevated temperatures ($\sim 1.2$ eV at $T > 700$ K) [16]. On the other hand, this value is $\sim 3$ times greater than the one extracted from the current–voltage curves for the Ta$_2$O$_5$ based memristors ($\sim 0.2$ eV) [17]. This discrepancy can be attributed to the formation of the non-stoichiometric Ta-enriched nanoinclusions at the Ta$_2$O$_5$/TiN interface [13]. Also, one should take into account the possibility of the formation of other oxide phases (e. g. TaO, etc.) in the Ta$_2$O$_5$ sublayer. It is worth noting that the estimate of $E_a$ for ZrO$_2$(Y) according to (1) gives $E_a \sim 0.57$ eV for $f_i = 2.5$ kHz and $E_a \sim 0.55$ eV for $f_i = 6.5$ kHz. These values coincide with the values of $E_a$ for ZrO$_2$(Y) extracted from the ion migration polarization measurements at moderate temperatures (0.53 to 0.55 eV within the range of $T = 300–500$ K) [14].

4. CONCLUSIONS

The results of present study confirm the resonant activation of the O$^{2-}$ ion migration via the $V_{O6}$ in the alternating electric field to be the origin of the beneficiary impact of adding the HF sinusoidal signal to the triangle switching voltage pulses on the performance and stability of the RS in the memristor based on the ZrO$_2$(Y) films and on the ZrO$_2$(Y)/Ta$_2$O$_5$ stacks. These results manifest the fundamental properties of memristor as a stochastic multistable nonlinear system. From the practical point of view, the results of the present study indicate the prospects for the development of the innovative switching protocols applicable to the next-generation non-volatile memory devices to improve the performance and durability of these ones.

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CONFICT OF INTEREST

The authors declare that they have no conflict of interest.

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