Effect of internal noise on the relaxation time of an yttria stabilized zirconia-based memristor

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Abstract

The effects of temperature on the switching kinetics of an yttrium-stabilized zirconia-based memristor from a low-resistance state to a high-resistance state have been experimentally investigated. It was found that the memristor relaxation time depends on the temperature in a non-monotonous way, with a maximum observed at the temperature close to 55 °C. This nonmonotonic behavior is a signature of the noise-enhanced stability phenomenon observed in all physical and complex systems characterized by metastable states.

Keywords: resistive switching, memristor, yttria stabilized zirconia, metastability, noise induced stabilization, beneficiary role of noise

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1. Introduction

In recent years, the investigations of resistive switching (RS) attracted much attention [1]. The RS effect is a bistable (or multistable) switching of resistance of a thin (10-50 nm thick) dielectric film sandwiched between two conductive electrodes subjected to an external voltage [2]. The electronic devices utilizing the RS effect are called *memristors* [3]. The memristors are considered to

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be promising for applications in non-volatile computer memory of new generation [4], in novel (non-von Neumann) computer architectures [5], in neuromorphic electronic devices [6, 7], etc.

Currently, the commonly accepted understanding of the RS mechanism is based on the concept of formation of the so-called conductive filaments (CFs) between the memristor stack electrodes [8]. In metal oxide-based memristors, CFs consist of oxygen vacancies (V_O's) [9]. CFs form in the electric field between the memristor stack electrodes during the so-called forming process and shortcut the electrodes. As a result, a memristor switches into a low resistance state (LRS). The memristor can be switched back to a high resistance state (HRS) by applying a voltage pulse of appropriate polarity to the memristor electrodes which causes the rupture of the CF near one of the electrodes (so-called RESET process). The CF can be restored by applying a pulse of opposite polarity which causes the memristor to switch back to LRS (so-called SET process). The mechanism described above is usually referred to as *bipolar* RS.

At present, the wide application of memristors is limited by insufficient stability, high variability of RS parameters during the operation, lack of understanding of drift-diffusion processes and their degradation [10, 11, 12]. One of the fundamental origins of the instability of the memristor's parameters is the essentially stochastic nature of the RS process [13, 14].

Recently, new approaches to improving switching properties in various nonlinear multistable stochastic systems that use a beneficial role of noise have been extensively investigated. An archetypal model is that of an overdamped Brownian particle moving in multistable potential profiles. The beneficial or constructive role of noise usually manifests itself in a nonmonotonic dependence of the switching parameters (such as switching time, relaxation time, mean amplitude of average switching amplitude, output signal-to-noise ratio, etc.) on the noise intensity or temperature. In other words, in nonlinear systems, the effect of noise can induce new, more ordered regimes that lead to regular structures, an increasing degree of coherence, and cause new phase transitions [15, 16, 17, 18, 19, 20, 21, 22].

Very recently, noise-induced phenomena showing the constructive role of noise in the RS process, typical of nonlinear stochastic systems, have been experimentally observed in memristors using a periodic external signal superimposed to the switching voltage pulses. These are the stochastic resonance [23] and the stochastic resonant activation [24]. A positive impact of adding a white Gaussian noise source to the switching voltage pulses on the stability of memristor parameters was shown in Ref. [25].

Furthermore, the relaxation process in stochastic models of memristive devices has been investigated in Refs. [26, 27], where the coordinate of the Brownian particle is representing either location of diffusing defects or the length of conductive region (or CF). The nonlinearity in these models is caused by the internal material structure of the memristive device and by the boundary effects near the junctions with the electrodes. The theoretical analysis carried out in Ref. [26] shows that the relaxation time of the RS process can have a nonmonotonic dependence on the intensity of thermal noise. This nonmonotonic behavior with a maximum is the signature of the noise enhanced stability (NES) phenomenon, which occurs in all physical (classical and quantum), biological, chemical and ecological systems with metastable states, see the Refs. [28, 29, 30, 31] and references therein, where the NES effect was investigated in static and fluctuating metastable potential profiles. The stability of a metastable state can be enhanced by the noise and its average lifetime is a measure of this stability. This noise-enhanced metastability is a consequence of the interplay between the thermal fluctuations and nonlinearity of the complex system investigated, and it is observed by increasing the temperature.

In the present work, the dependency of the switching kinetics from LRS to HRS in memristors based on Ta/ZrO_2(Y)/Pt stacks from LRS to HRS on different values of the device temperature, which defines the intensity of internal random fluctuations, has been studied experimentally. The work is aimed at the experimental observation of the effect of increasing the relaxation time from a metastable state due to thermal noise.

The motivation for choosing ZrO_2(Y) as a functional dielectric material was the following. Enough concentration of the oxygen vacancies is needed for resistive switching in oxides. Usually, necessary vacancy concentration is achieved by deposition of non-stoichiometric oxides, by the post-deposition annealing of the oxide films in vacuum, or as a result of the electrochemical reduction/oxidation reactions at the interface between a functional oxide layer and a chemically-active metal electrode.

In ZrO_2(Y), the oxygen vacancies are the elements of the crystal structure, and the concentration of the vacancies in equilibrium is determined by the Y one (the number of oxygen vacancies is 1/2 of those of the Y atoms). Thus, one can control the equilibrium concentration of oxygen vacancy in ZrO_2(Y) by varying the Y fraction. It should be stressed here that the oxygen vacancy concentration in ZrO_2(Y) is determined by the Y one solely and almost does not depend on temperature, environment, etc. This factor along with high anion mobility makes ZrO_2(Y) a promising material for memristor applications [32]. To date, several studies have been reported on ZrO_2(Y)-based memristors demonstrating good performance [33, 34, 35, 36, 37].

2. Experiment

2.1. Sample preparation

The memristor stacks Au(20 nm)/Ta(40 nm)/ZrO_2(Y)(20 nm)/Pt(20 nm)/ Ti(5 nm)/SiO_2(700 nm)/*p*-Si(001) were deposited using the setup for thin film deposition in vacuum Torr International® R2G1-1G2-EB4-TH1. The ZrO_2(Y) films were deposited by radio-frequency magnetron sputtering of a target pressed from a mixture of ZrO_2 and stabilizing oxide Y_2O_3 (12% mol.) powders at the substrate temperature $T_g = 250$ °C. The metal layers were deposited by direct current magnetron sputtering at $T_g = 200$ °C. From these stacks, prototype cross-point memristors were fabricated by standard photolithography. The active areas were 20×20 μm^2.

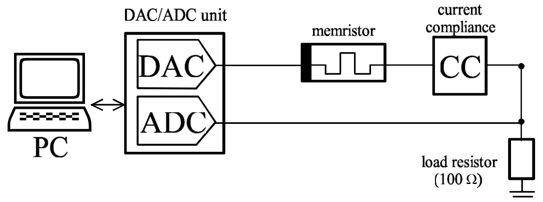


Figure 1: Schematic representation of the experimental setup.

2.2. Methods of investigation

The electrical characteristics and switching kinetics of the prototype memristors were measured using National Instruments® USB-6361 ADC/DAC module operated under LabVIEW™ control software. The electrical contacts to the contact pads of the cross-point memristors were provided using EverBeing® EB-6 probe station. The schematic representation of the experimental setup is shown in Fig. 1. To measure the temperature dependencies of the electrical parameters and the switching kinetics of the memristors, the device were placed on a sample holder with a built-in Peltier cooler/heater. The temperature was maintained using Microstat® 300 CON temperature controller with the uncertainty of ±1 °C. The temperature measurements were performed in the temperature range 20—80 °C, with the temperature step of 10 °C. In any state LRS or HRS when we apply a constant voltage V_0 which is small enough to avoid any switching, we observe equilibrium fluctuations $i(t)$ of the current $I(t) = I_0 + i(t)$. These fluctuations $i(t)$ include thermal noise i_T and flicker noise $i_f(t)$ [41]. The intensity of i_T is given by the Johnson-Nyquist formula [38, 39, 40]

$$\langle {i_T}^2 \rangle = \frac{4k_B T}{R}, \tag{1}$$

where R is resistance, k_B is the Boltzmann constant. When we apply a high driving voltage V_0, we will observe the RS process associated with the fast change in resistance R. As it was shown in [26] it is essentially a non-equilibrium transition process. During this process the fluctuations of current deviate from the equilibrium values and return to it after the RS is finished.

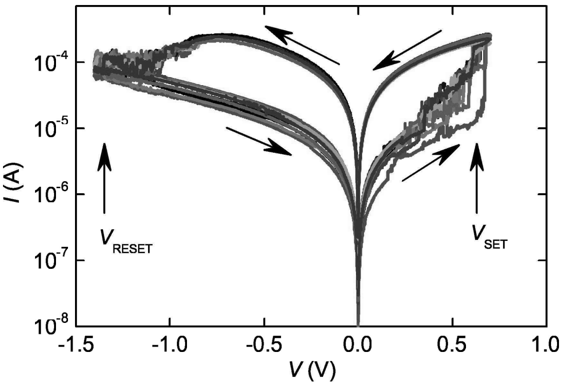


Figure 2: Typical cyclic I—V curve of a prototype memristor based on a Ta/ZrO$_2$(Y)/Pt stack.

The forming of the memristors was performed by applying the voltage in the range $V = +\ 5$—6 V at the current compliance 600 μA. All measurements were performed on different devices with the same structure and resistive switching parameters (voltage of RESET V_{RESET} and SET V_{SET} processes and current in resistive states). The investigated devices were prepared in the same technological process.

Before each measurement of the switching kinetics, the presence and parameters of resistive switching of memristors were checked by measuring several (5—6) cyclic current-voltage (I–V) curves at the current compliance 300 μA. Examples of the I–V curves are shown in Fig. 2. The voltage sweep rate was 1—5 V/s. We observed steps on the I–V curves that can be attributed to the quantization effect [42]. As the temperature increases, a less pronounced conductance quantization effect is observed due to the increase in the intensity of the thermal noise (results not shown here).

Just before each measurement of the switching kinetics, the memristor was stopped at the LRS at a voltage of -0.5 V. The switching kinetics, namely current response, of the memristors from LRS into HRS was investigated by applying a step voltage $V(t) = V_0 \cdot \theta(t)$ with $V_0 = -1.5$ V. This value was close to V_{RESET}. The current response of a memristor was recorded with a frequency $f_s = 100$ kHz from a load resistor (100 Ω), which was connected in series with a memristor. Thus, the time series of current flowing through the load resistor was equal to the time series of current flowing through the memristor. A total of 100—150 time series of current were obtained for each value of device temperature.

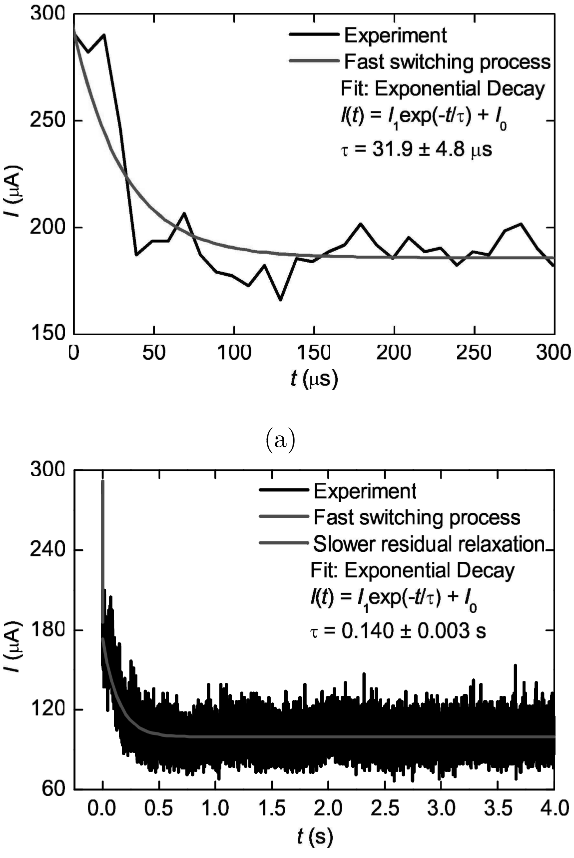
As will be shown below, the switching kinetics was characterized by at least two exponential modes: relatively fast switching from LRS to some intermediate HRS [see Fig. 3(a)] is followed by a slower residual relaxation to the stationary state with a higher value of resistance [see Fig. 3(b)]. This is in accordance with the theory [27], where the non-stationary probability distribution of memristor state variable was obtained as a sum of terms exponentially decreasing in time with different rates. The state variable defines the value of memristance, which in turn defines the value of current response investigated in the experiment. We define relaxation time as the characteristic time scale related to the slowest exponential function observed in experiment or in other words as the residual relaxation of current.

To calculate the relaxation time (τ), the resulting time series of the current was obtained as time average over a short time interval $T_a = Mt_s$, where M is the number of points in the time series of the current, measured within the interval T_a, and t_s is the time step ($t_s = 1/f_s$). The value of M was 1000. Further, the averaged current waveform was approximated by the exponential decay function

$$I(t) = I_0 + I_1 \exp\left(-\frac{t}{\tau}\right) \tag{2}$$

with fitting parameters I_0, I_1, and τ. The fitting was performed using the *nonlinear curve fit* function of Origin$^{\mathrm{TM}}$ 7.0 software.

And finally, the values of τ extracted from the resulting time series of the current measured at the same device temperatures T were ensemble averaged.



(a)

(b)

Figure 3: Relaxation kinetics of the memristor during the switching from LRS HRS (20 °C): (a) initial stage of fast switching and (b) slow relaxation regime with approximation by the exponential decay function. The current is presented in absolute values.

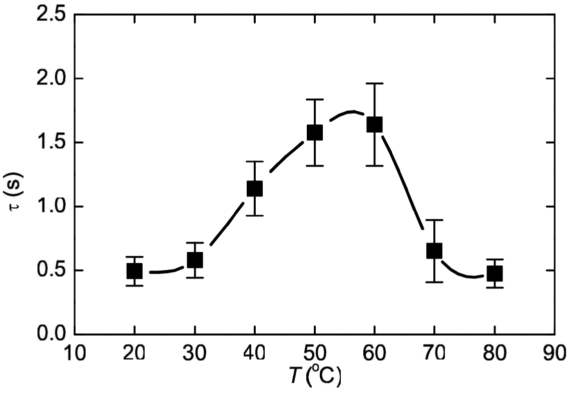


Figure 4: Temperature dependence of the relaxation time of the memristor during switching from LRS to HRS.

3. Results and Discussion

Typical relaxation curve of $I(t)$ of the prototype memristor when switching from LRS to HRS under the step voltage $V = -1.5$ V measured at room temperature is shown in Fig. 3(b). The kinetics of switching from LRS to HRS was characterized by at least two exponential modes: a fast switching from LRS, with the switching time ~ 10 μs [see Fig. 3(a)], is followed by a relatively slower relaxation of the memristor to a quasi-stationary HRS with the relaxation time $\tau \sim 0.1$ sec (see Fig. 3(b)).

The temperature dependence of the relaxation time τ is shown in Fig. 4. It is nonmonotonic and has a maximum at $T \approx 55$ °C. This $\tau(T)$ behavior obtained from the experiments is akin to that theoretically predicted in Refs. [26, 31], the NES phenomenon, where the lifetime τ of metastable state with fluctuating barrier was calculated analytically as a function of the noise intensity. To discuss how the NES phenomenon may occur in the experiment we need to consider a more realistic physical model of the memristive system. The models of memristive devices involve at least two equations: one is a first-order differential equation for a state variable, the second is an Ohmic-type relationship between voltage and current. The third equation can be added to describe the Joule heating caused by the current running through the device. Noise or stochasticity is an important inherent property of the memristor frequently observed in experiments. Therefore, the noise effects should not be ignored. To take this into account, the appropriate noise sources should be considered in the model. Therefore, the above-described equations can be written in the following general form

$$\frac{dx}{dt} = f(x, V, \xi(t)), \tag{3}$$

$$I = g(x, V, \zeta(t)), \tag{4}$$

$$\frac{dT}{dt} = h(T, x, V, I), \tag{5}$$

where x is the state variable, $\xi(t)$ and $\zeta(t)$ represent noise sources, f, g and h are some functions.

The equation for the state variable (3) can be one-dimensional. In this case the same model can describe uniform and localized resistive switching (with one or few CFs, see e.g. Refs. [26, 27, 43]), and the difference between these types of switching is neglected. Indeed, the simulations carried in Ref. [44] for oxide switching devices like Pt/TiO_2/Pt show that there is no difference between two models of switching: "localized conductive filaments" and "uniform push/pull" of the oxygen front near an interface. While the three-dimensional equation for the state variable allows investigating the appearance of localized switching [10].

The relaxation time observed experimentally earlier in Ref. [45] monotonically decreases with the temperature according to Arrhenius law, which can be written as follows

$$\tau = \frac{L^2}{l^2}\tau_0 \exp(\frac{E_a - B|V|}{k_B T}), \tag{6}$$

where E_a is the activation energy, k_B is the Boltzmann constant, l is the period of microscopic structure of the dielectric material, L is the distance between the electrodes of the memristive device, V is the bias voltage, and τ_0 and B can be treated as fitting parameters. The Arrhenius dependence (6) naturally arises in the coarse grained stochastic model of memristive device [26, 27] with an additive noise source in the differential equation (3)

$$\frac{dx}{dt} = -\frac{\partial U_{eff}(x,V)}{\partial x} + \xi(t), \tag{7}$$

where $\xi(t)$ is a white Gaussian noise with $\langle \xi(t) \rangle = 0$, $\langle \xi(t)\xi(t+\tau) \rangle = 2D_{eff}\delta(\tau)$ and the effective potential profile U_{eff} is a linear function of x

$$U_{eff}(x,V) = v_{eff}(V)x. \tag{8}$$

The values D_{eff} and v_{eff} are the effective diffusion and drift coefficients (for more details see [26]). The coarse grained stochastic model is based on the description of the thermally activated random hoping of metal ions or structural defects between identical and periodically located trapping sites of the dielectric material, represented by potential wells separated by potential barriers with height E_a. The Arrhenius dependence of the relaxation time (6) is obtained under additional assumptions of small bias voltage

$$V \ll E_a/B, \tag{9}$$

low intensity of thermal fluctuations

$$k_B T \ll E_a - BV, \tag{10}$$

and constant temperature

$$\frac{dT}{dt} = 0. \tag{11}$$

The relaxation time (6) represents the lifetime of a metastable state of the periodical potential profile. This lifetime is known also as Kramers time [26, 27]

$$\tau_K = \tau_0 \exp(\frac{E'_a}{k_B T}), \tag{12}$$

where $E_a' = E_a - B|V|$ is the activation energy of the metastable state in the presence of a bias voltage $V < 0$. If we increase T and V beyond the assumptions (9) and (10) the relaxation time can become a nonmonotonic function of temperature as it was shown in Ref. [26].

In a more general case, when the internal structure of the dielectric material is quasiperiodic or not periodic and in addition to additive thermal fluctuations there are multiplicative noise sources, the equation for the state variable x can be written in the following form

$$\frac{dx}{dt} = -\frac{\partial \Phi(x, V, \eta)}{\partial x} + \xi(t), \tag{13}$$

$$\Phi(x, V, \eta) = U(x, V) + U_\eta(x, V)\eta(t), \tag{14}$$

where $\xi(t)$ and $\eta(t)$ are the noise sources. The potential profile $\Phi(x, V, \eta)$ is a sum of two terms: the deterministic potential $U(x, V)$ and the fluctuating term $U_\eta(x, V)\eta(t)$. The deterministic potential contains different barriers and wells which are located periodically or quasiperiodically and may be not completely identical to each other. The fluctuating term can describe random changes of the internal structure (like fluctuating barriers) caused by the influence of diffusing defects on the internal energy profile of the dielectric material. The additive noise term $\xi(t)$ is the thermal noise. In this case the relaxation time of the memristive device will be conditioned by the lifetimes of the metastable states with fluctuating parameters. It was shown in Ref. [31] that due to the interplay of fluctuations and nonlinearities describing the potential profile, the lifetime of such a metastable state can be a nonmonotonic function of the intensity of the additive noise $\xi(t)$, which is defined by the temperature.

Therefore, the experimental observation of the nonmonotonic behavior of the relaxation time $\tau(T)$ shown in Fig. 4 can be interpreted as a manifestation of the NES effect, that is an enhancement of the lifetime of the metastable state due to thermal noise. Furthermore, it points to a beneficiary role of thermal noise and its possible use as a control mechanism to improve the RS stability.

4. Conclusions

In the present study, the temperature dependence of the switching kinetics from LRS to HRS in a memristor based on a Ta/ZrO$_2$(Y)/Pt stack was experimentally investigated in the temperature range of 20—80 °C. The relaxation time of the memristor resistance state depends nonmonotonically on the temperature with a maximum close to $T \approx 55$ °C, and the NES phenomenon has been observed: the experimental results show the effect of stabilization of the metastable state of the memristor by thermal noise.

A peculiarity of the relaxation process of the investigated memristor is the presence of at least two exponential modes. We observe that after a relatively fast switching from LRS to some intermediate HRS, there is a slower residual relaxation to the stationary state with a higher value of resistance. This behavior is in agreement with the one theoretically predicted in Refs. [26, 27], where the non-stationary distribution of the internal state variable was obtained as a sum of terms exponentially decreasing in time with different rates.

It is worth noting that in most memory element applications the residual relaxation is not used and the switching is considered completed after the fast stage of the switching process. This means that memristive devices are usually exploited in regimes far from the equilibrium stationary states.

This study paves the way for the use of temperature as a control parameter to improve the RS stability of memristor devices. An optimal temperature value for a particular memristor design, characterized by specific materials and thicknesses of the layers of the memristor stacks, can be experimentally determined or theoretically estimated.

Theoretical and experimental investigations on the role of internal and external noise sources on the RS process of memristors will be the subject of future studies.

Financial Support

The present work was supported by the Government of the Russian Federation under Megagrant Program, Agreement No. 074-02-2018-330 (2).

Acknowledgements

The experimental investigations were carried out using shared research facilities of Research and Educational Center for Physics of Solid State Nanostructures at Lobachevskii University of Nizhnii Novgorod.

Conflict of Interests

The authors declare no conflict of interests.

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