Investigation of resistive switching by non-stationary signals in ZrO₂(Y) films by atomic force microscopy

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Abstract. Resistive switching in the yttria stabilized zirconia films on the conductive substrates has been studied using Conductive Atomic Force Microscopy. Switching was performed by triangle voltage pulses with superimposed high-frequency sinusoidal signal applied between the probe and the sample. The performance of the resistive switching was characterized quantitatively by the ratio of the electric current flowing through the contact of the probe to the dielectric film surface (together constituting a nanometre-sized virtual memristor) in the low resistance state and in the high resistance one (ON/OFF ratio). The increase in this ratio when applying the sinusoidal signal as compared to the switching by the triangle pulses has been observed. Also, the long-time scale stability of the mean values of the probe current in both states was found to improve when applying the sinusoidal signal. The effect of the high frequency sinusoidal signal on the performance of the virtual memristor was attributed to the resonant activation of the oxygen ion migration via the oxygen vacancies by the alternating external electric field.

1. Introduction

In recent years, the investigations of resistive switching (RS) have attracted much attention [1]. The effect of RS consists in the bistable (or multistable) switching of the resistance of a thin (from several nanometres to several tens of nanometres in thickness) dielectric films sandwiched between two conductive electrodes under the external voltage applied between the electrodes [2]. The electronic devices based on the RS effect are called memristors [3]. At present, the understanding of the RS mechanism in the metal oxides is based on the concept of forming the conductive filaments composed of the oxygen vacancies between the conductive electrodes in the electric field applied between the electrodes [4]. The switching of a memristor from the low resistance state (LRS) to the high resistance state (HRS) is achieved by the rapture of the filament by a voltage pulse of an appropriate polarity applied between the electrodes (the RESET process). The filament can be restored again by another voltage pulse of the opposite polarity that results in the switching from the HRS back to LRS (the SET process). The processes described above constitute the physical mechanism of so-called bipolar RS.

The memristors are considered to be promising for various applications, for instance, in the next generation of non-volatile computer memory (Resistive Random Access Memory, ReRAM) [5], in the
neuromorphic computers [6], etc. However, at present the application of the memristors is limited by an insufficient stability of the memristor parameters, which is a fundamental property of the RS [7]. The origin of the RS instability is the stochastic nature of the rapture and restoring of the filaments in the functional dielectric layer. A very limited (countable) number of oxygen vacancies are involved into the RS [8]. So far, memristor appears to be a typical mesoscopic system, where the fluctuations of any physical parameter may be much larger than its averaged value.

Traditional approaches to the improvement of the RS stability include the appropriate choice of the functional insulator and electrode materials, the engineering of the grain boundaries in the functional insulator, the application of electric field concentrators inside the insulator, etc. [9–11]. The alternative approaches include the circuit solutions, the application of adaptive switching protocols, etc. [12].

In the present study, the local RS in the yttria stabilized zirconia (YSZ) films was investigated by Conductive Atomic Force Microscopy (CAFM) [13] using the triangular switching voltage pulses with superimposed sinusoidal high-frequency (HF) signal.

2. Experiment

The YSZ films of ≈4 nm in thickness (the molar fraction of the stabilizing oxide Y₂O₃ in the film material was ≈0.12) were deposited by HF magnetron sputtering at 300 °C using Torr International® 2G1-1G2-EB4-TH1 vacuum setup. The Si(001) substrates with pre-deposited SiO₂ film (≈500 nm thick), Ti adhesion layer, and TiN conductive one (25 nm in thickness each) were used.

The RS in the YSZ films was investigated using Omicron® UHV AFM/STM LF1 in ultra-high vacuum (UHV) at room temperature in the contact mode. The base residual gas pressure inside the AFM/STM chamber was ~10^{-10} Torr. The NT-MDT® NSG-11 DCP™ diamond-like film coated AFM probes were used. The schematic representation of the experimental setup is shown in Figure 1. The bias voltage V_g applied between the CAFM probe and the TiN conductive sublayer was supplied by the digital-to-analogue converter (DAC) of NT-MDT® Solver Pro™ AFM controller used as an external computer-controller programmable voltage source.

The endurance diagrams for the probe current values I_t in the LRS and HRS I_ON and I_OFF, respectively during multiple cyclic write/erase operations were recorded in the experiments. The switching/measurement protocol is shown in Figure 2. After the triangular switching voltage pulses with an amplitudes V_SET = 5 V and V_RESET = −6 V and the pulse durations T_SET, T_RESET = 1 ÷ 5 s, the values of I_ON and I_OFF, respectively, were recorded at V_g set to the read voltage V_READ = 3 V several times N_READ = 10 ÷ 20 and averaged. The HF sinusoidal signal with the amplitude A = 0.2 V and frequency f = 6.5 kHz (matching the frequency of the O₂⁻ ion jumps to the nearest neighbouring oxygen vacancies in YSZ at 300 K [14]) was superimposed onto the triangle switching pulses.

![Figure 1](image-url)  
*Figure 1. Schematic representation of the experimental setup for studying the local RS in thin YSZ film on a conductive substrate by CAFM. I_t – probe current signal; DAC – digital-to-analogue converter.*
3. Results and discussion

A pronounced hysteresis typical for the bipolar RS was observed in the cyclic current-voltage ($I$–$V$) curves of the CAFM probe contact to the YSZ film (together constituting a nanometre-sized virtual memristor) (Fig. 3). The decrease of the area within the hysteresis loops with number of switching cycles $N$ was the manifestation of the virtual memristor degradation during recording of the cyclic $I$–$V$ curves. Accordingly, the $I_{ON}$ values approached $I_{OFF}$ with increasing $N$ (Fig. 4). The adding of the HF sinusoidal signal to the triangular switching pulses resulted in an increase of the local RS durability: the decrease of $I_{ON}$ began at larger values of $N$ than in the case of switching by the triangle pulses without the HF sinusoidal signal. The observed effect of enhancing the RS durability was attributed to the resonant activation of the $\text{O}_2^-$ ions migration via the oxygen vacancies under the external alternating electric field, which stimulates the elementary jumps of the $\text{O}_2^-$ ions and, this way, favours the formation and rupture of the filaments under the slowly varying external electric field during the SET and RESET processes, respectively. The effect of resonant activation belongs to a wide class of phenomena inherent to the stochastic multistable systems (along with stochastic resonance, noise-induced stabilization, etc.) and was manifested in many fields of physics and chemistry [15].

The characteristic frequency of the $\text{O}_2^-$ ion jumps onto the nearest neighbouring oxygen vacancy $f_i$ can be estimated from the ion jump activation energy $E_a$ using the following equation [16]:

$$ f_i \sim f_0 \exp \left( \frac{E_a}{kT} \right), $$

where $f_0 \sim 10^{13}$ Hz is typical lattice vibration frequency, $k$ is Boltzmann constant, and $T$ is the temperature. Assuming $E_a = 0.55$ eV for YSZ [14], one gets $f_i \approx 6.5$ kHz at 300 K. Based on this estimate, the frequency of the HF sinusoidal signal used in the experiment was selected.

Figure 2. The protocol (the dependence of $V_g$ on the time $t$) for measuring the endurance diagrams to study the effect of the superposition of the HF sinusoidal signal onto the triangle switching pulses on the RS parameters.

Figure 3. Cyclic $I$–$V$ curves of the CAFM probe contact to the YSZ film surface (virtual memristor). $N$ – the number of switching cycles.
Figure 4. Endurance diagrams for the AFM probe contact to the YSZ film (virtual memristor): ▲,△ – $I_{ON}$, ■,□ – $I_{OFF}$, ■,▲ – with HF sinusoidal signal ($A = 0.2$ V, $f = 6.5$ kHz) on the triangle switching pulses; □,△ – without the sinusoidal signal.

4. Conclusion
The present study demonstrates a beneficiary impact of additional high frequency sinusoidal signal on the triangle switching voltage pulses on the stability of the resistive switching in the contact of a CAFM probe to the YSZ film on a conductive substrate. The observed effect was attributed to the resonant activation of the oxygen ion migration via the oxygen vacancies in YSZ in external alternating electric field oscillating with corresponding frequency. The obtained results open the prospects for the development of the innovative switching protocols applicable to the next-generation RRAM devices to improve the durability of these ones.

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