Investigation of Flexible Memory Elements

T. V. Kundozerova and G. B. Stefanovich

***Abstract*—A resistive memory device can be used in a new area of electronics: flexible electronics. Nowadays devices of flexible electronics is finding increasing application in various fields such as flexible displays, radio frequency identification tags (RFID), electronic paper, solar cells and others technologies. In this article we present an overview of unipolar nonvolatile resistive switching in a flexible metal-oxide-metal thin-film memory cell and a model of switching mechanism.**

***Index Terms*—Resistive random access memory, oxides, flexible electronics.**

I. Introduction

THE effect of resistive switching is a sharp and reversible transition of materials between two states with a different resistance. Switching is observed in a large class of compounds: complex perovskite oxides, organic compounds, binary metal oxides such us NiO [1], CuO [2], ZnO [3], TiO_2 [4], Nb_2O_5 [5], Ta_2O_5 [6], ZrO_2 [6], HfO_x [7] etc. [8]-[10].

Resistive random access memory (ReRAM or RRAM) it is an electronic memory which is based on resistive switching effect. The ReRAM memory cell has a capacitor-like structure (metal–insulator–metal) in which an oxide layer is located between two metal electrodes. Under the voltage pulses ReRAM cells switch between high resistance state (HRS) and low resistance state (LRS). HRS and LRS represent a logical “1” and “0”, it is stable in time nonvolatile states.

In appearance of current–voltage characteristics, switching behavior (ReRAM operations) can be divided into two broad classes: unipolar and bipolar. Switching is called unipolar (or symmetric) when the switching procedure does not depend on the polarity of the voltage and current signal, it depends only on amplitude. Bipolar switching required an alternating polarity of the applied signal. The same material can show both bipolar and unipolar switching. Type of switching depends on material of electrodes, property of oxide layer, interface between oxide and electrode, and conditions of the electroforming process.

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The working process of ReRAM cell based on unipolar switching includes 3 stages: 1) electroforming (forming); 2) switching from low resistance state to high resistance state; 3) switching from HRS to LRS. The forming process should occur only once, at the beginning, and it is similar to electrical breakdown [11]. Numerous switches between LRS and HRS are working cycles of memory cells.

Despite the final convention on the switching mechanisms is still not achieved, it is widely accepted that nonvolatile resistance switching occurs through the formation and rupture of a nanoscale conducting filament [2], [12]. The presence of the filament in memristor cell leads to metallic-type conductivity. During the reset process, the conductive filament is disrupted and semiconducting properties are restored in the memory cell. HRS can be developed by various metal-dielectric phase configurations whereas a high reproducibility of LRS attributes to unique conducting percolation path. This model is confirmed by a series of experiments: temperature dependence of resistance, scaling behavior of resistance states (oxide thicknesses, area of electrodes), FIB-SEM and XPS investigation of structure, frequency dependence of impedance etc. [8], [13]-[16].

II. Experimental

The materials of oxide layers which used in resistance random access memory can be obtain by various method of thin film deposition: PLD (pulsed laser deposition), magnetron sputtering, sol-gel method, e-beam evaporation, thermal oxidation, anodic oxidation (anodizing).

Low temperature fabrication process is a critical condition for flexible electronics devices. Anodic oxidation is simple, low cost, high deposition efficiency process to deposit thin dielectric films. This method now is a well-knownб it has various industrial applications and allows to get various oxides such as Ta_2O_5, ZrO_2, Nb_2O_5, CuO, Al_2O_3, V_2O_5 etc.[17]. The method of anodic oxidation allows fabrication oxide based structures on different substrates (silicon, glass, polymers) at a room temperature, without any destruction of the substrate material. The memory cells obtained on flexible substrate do not differ from the same cells on solid silicon substrate. An oxide is formed on a metal surface by applying an electrical current or potential through an electrochemical cell with suitable electrolyte. Anodic oxidation is usually carried out isothermally (T = constant) under galvanostatic (j = const) or voltstatic (V = const) regime. In our work we used the following regime: constant current density of about 1 mA/cm^2, 0.1 N aqueous solution of H_3PO_4 acid. The thickness of the oxide layer can be controlled by anodization voltage V_f (the potential which drops through the oxide), according to the anodization constant [5]. The thickness of the obtained oxide film was about 100 nm.

In case of flexible memory structures the fabrication included epy following steps:

1) Polymer metallization. Thin metallic film of Nb was sputtered on kapton substrate by RF magnetron sputtering using a metallic Nb target in an Ar atmosphere.

2) Anodic oxidation of Nb metallic layer.

3) Deposition of Au top electrodes. Fig. 1 shows the photograph of the obtained ReRAM structure and its schematic diagram.

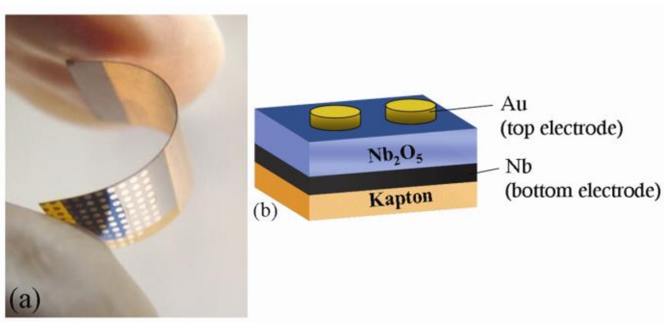


Fig. 1. (a) Photograph of flexible ReRAM and (b) schematic diagram of the flexible ReRAM device structure

The switching properties of the obtained structures were measured by Keythley 2410 SourceMeter. Positive bias voltage was applied to the top electrode, and the bottom metallic part of the field was grounded.

III. Results and Discussion

As prepared ReRAM structures do not demonstrate a resistance switching effect – the electroforming process is necessary. The electroforming was carried by the following way: on the top electrode of the structure, a linearly increasing voltage is applied (the bottom electrode is grounded). After the threshold voltage is reached, the resistance of the structure abruptly (nanoseconds range) falls by several orders of magnitude. The current which is goes through the structure during an electroforming is limited by $I_c = 5$ mA. In case of anodic oxide, a positive polarity of a top electrode during the forming processes is required. Under negative voltages, electroforming occurs at higher voltages. As a result, an energy which released in this process increases and the probability of irreversible breakdown is growing up. Note that a setting of adequate current compliance is very important during the electroforming process. Without compliance, the structure switches to irreversible low resistance state.

After the forming process, a system in low resistance state is switched to a high-resistance state by applying a threshold voltage (‘reset process’). Switching from HRS to LRS (‘set process’) is achieved by applying a threshold voltage greater than the reset voltage (Fig. 2). Note that, similarly electroforming, a set process requires a current compliance. Without current compliance the structure is switched into a permanent low-resistance state.

Current-voltage characteristic of Kapton/Nb/Nb_2O_5/Au structure is a typical characteristic of ReRAM devices that produces unipolar resistive switching between HRS and LRS with set/reset voltage ~ 0.9V/0.4V and resistance ratio $R_{HRS}/R_{LRS} > 100$ (Fig.3).

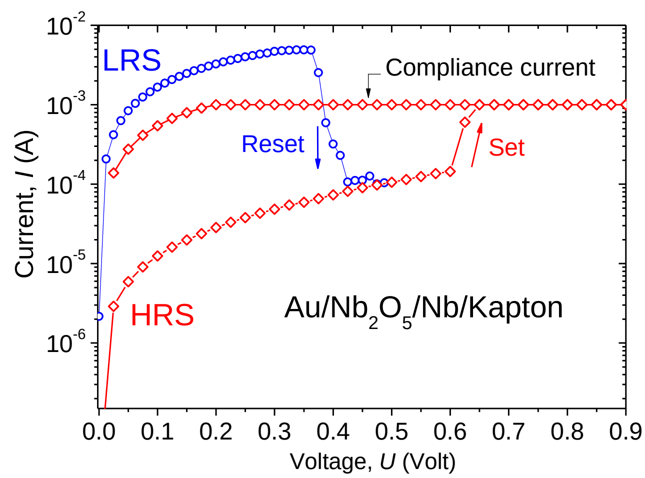


Fig.2. Resistance switching *I-V* characteristics of the flexible structure Au/Nb_2O_5(75 nm)/Nb/Kapton

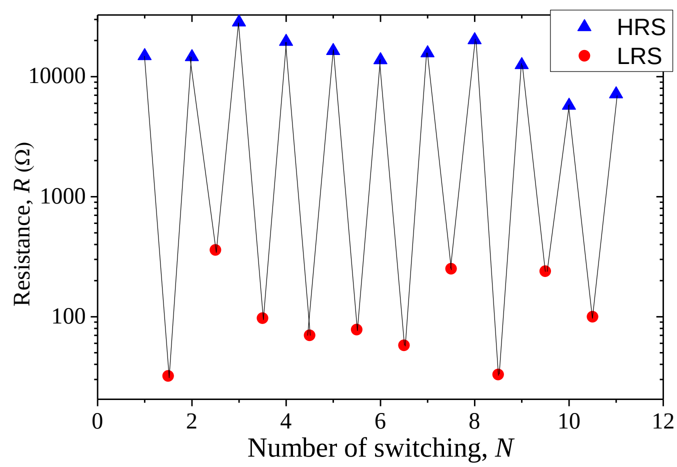


Fig. 3. Sequential switching of flexible ReRAM structure Kapton/Nb/Nb_2O_5/Au.

In order to confirm the feasibility of obtained ReRAM devices for flexible memory application, mechanical bending tests were conducted. After several flexing 1000, 5000...etc, and up to 10^5, low voltage signal (V = 0–0.1 V) was applied and the *I-V* characteristics of HRS or LRS of different structures were measured. Calculated from these characteristics resistances of LRS and HRS were not degraded after numerous bending (Fig. 4).

As was said, despite the final convention on the switching mechanisms is not achieved, it is widely accepted that nonvolatile resistance switching occurs through the formation and rupture of a nanoscale conducting filament

As a result of electroforming a constant conductive filament is generated in oxide (Fig. 5). It is confirmed by research of planar and sandwich structures [2], [9], [10]. A chemical composition of the filament is different from the material of oxide [11].

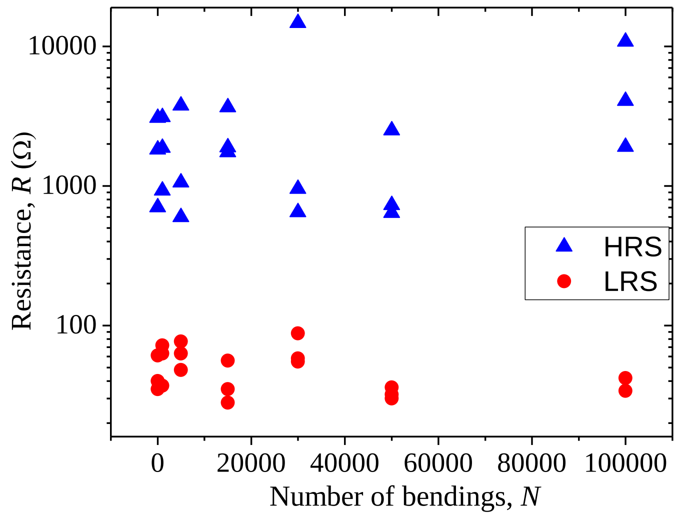


Fig. 4. The switching characteristics with continuous bending of the Kapton/Nb/Nb_2O_5/Au structure.

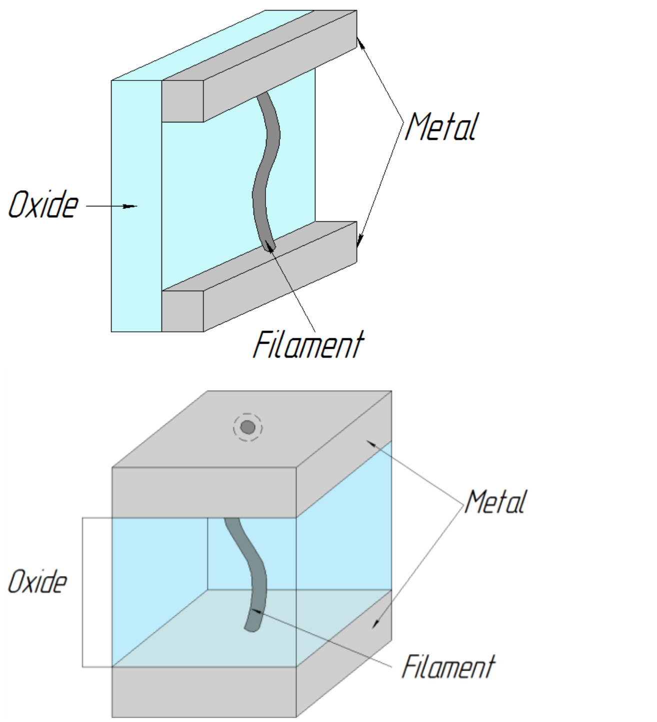
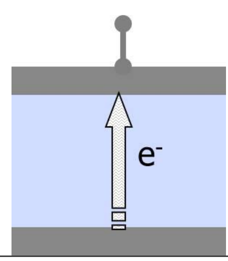
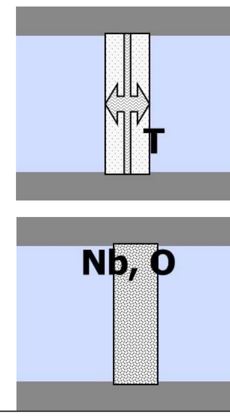
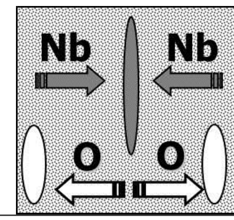
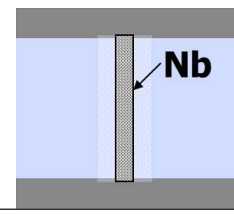


Fig. 5. Illustration of a filamentary conducting path in a planar and sandwich configuration of structure.

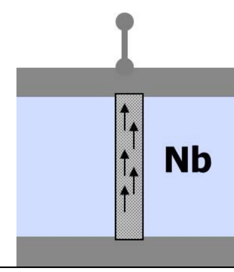
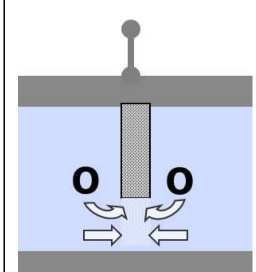
The switching process includes the following stages:

A. The forming process.

1) A dielectric breakdown of the oxide layer with a required current compliance. 2) A discharge of a capacitor. Release of energy which ReRAM as a capacitor structure stored before forming process.	
3) A sharp increasing of temperature and as a result fast local redox reactions.	
4) Under a gradient of temperature and diffusion process a Soret state is established. Segregation of metal in a center of high temperature region occurs.	
5) Due to a sharp decrease of a temperature after finishing of forming process the metallic filament is solidified.	

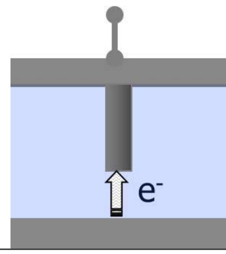
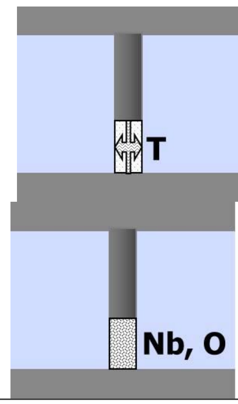
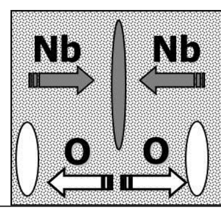
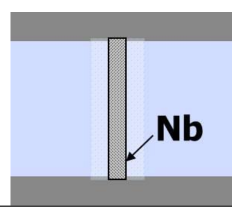
Thus the structure is switched to LRS. The total resistance of a ReRAM cell is determined by a resistance of a metallic filament.

B. The reset process (switching from LRS to HRS).

1) During the reset process a current which go through the filament becomes a source of electron wind.	
2) Electromigration of metal ions occurs by the action of the electron wind. The migration leads to the rupture of filament in an area near to cathode. A local domain with a high resistance and electric field are formed. 3) On the border of rupture a part of filaments converted to the oxide due to thermal oxidation under accelerating electric field.	

Thus a structure is switched to HRS. The total resistance of a ReRAM cell is determined by a new (reconstructed) section of oxide layer which is created by a rupture of the filament. Note that the resistance of a structure in a HRS is significantly smaller than that in an initial state before forming.

C. The set process (switching from HRS to LRS).

1) A dielectric breakdown of a reconstructed section of oxide layer with a required current compliance. 2) A discharge of a capacitor. Release of energy ReRAM as a capacitor structure stored before set process.	
3) Sharp increasing of temperature and as a result, fast local redox reaction.	
4) Under a gradient of temperature and diffusion process a Soret state is established. Segregation of metal in a center of high temperature region occurs.	
5) Due to a sharp decrease of a temperature after finishing of set process, the metallic filament is solidified.	

More detailed examination of this model has been presented in our recent work [18]. Presented model can explain numerous properties of unipolar resistive switching (scaling behavior, temperature and frequency dependences of resistance etc.).

IV. Conclusion

ReRAM elements based on anodic oxide Nb_2O_5 were obtained on a flexible polymer substrate. The performance of resistive switching was not degraded upon numerous substrate bending. Obtained devices exhibited a typical unipolar resistive switching. The switching mechanism of oxide based ReRAM structure is presented. The understanding of working mechanism and first positive results of experimental work are important and necessary steps for future development of flexible memory elements based on resistive switching effect.

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