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Investigation of the electroforming process in resistively switching TiO_2 nanocrosspoint junctions

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We report on the electroforming in resistively switching nanocrosspoint devices made of a reactively sputtered TiO_2 thin film between Pt and Ti/Pt electrodes, respectively. As most resistance switching materials, TiO_2 needs to be electroformed before it can be switched. This paper presents and compares current and voltage controlled electroforming with regard to the polarity. We show that a current-driven electroforming with negative polarities leads into the switchable high resistive state without need for a current compliance. These devices show an improved stability and reliability in bipolar resistive switching performance. © *2010 American Institute of Physics*. [doi:10.1063/1.3367752]

The resistance switching effect holds the promise for the realization of an alternative type of fast and nonvolatile memory.[1,2] Resistance switching materials can adopt at least two different resistance states, interpreted as logic states with a high resistive state (HRS) representing logical "0" and a low resistive state (LRS) representing a "1." The logic states can be toggled by exceeding a threshold voltage or current value. Thus, a resistive memory cell is inherently a two terminal device. Resistance switching can be distinguished between unipolar and bipolar and was observed in several material systems, where different physical processes are invoked for the switching effect.[3–5] However, the early description of the memristor as a discrete device by Chua and the recent transfer of its definition to already known resistively switching materials, especially TiO_2 thin films by Strukov *et al.* draw significant attention to this material system.[6–8] The particular physical mechanism is not fully understood yet, but the resistance state depends on the distribution of oxygen and oxygen vacancies, respectively.[9,10] The latter act as n-type dopants that influence the conductivity of the device, in particularly, with regard to an accumulation along the cathode, here Pt.[11] However, the possibly thermally assisted electroforming process, which is required after the film deposition to initiate resistive switching turns out to be the key for the establishment of resistive switching random access memory.[3,12] Here, the electroforming process for bipolar resistive switching in a $Pt/TiO_2/Ti/Pt$ system is investigated in detail. Four different methods in form of voltage controlled or current controlled signals with both polarities are described and compared.

The examined devices are nanocrosspoint junctions on an oxidized silicon substrate with metallization line widths between 500 and 100 nm yielding cell areas between 0.25 and 0.01 μm^2 as shown in Fig. 1(a). Each junction consists of a 25 nm thin Pt bottom electrode, a 30 nm thin reactively sputtered TiO_2 film and a 30 nm thin evaporated Ti/Pt top electrode [see sketch in Fig. 1(b)]. More details about the film deposition and the lithography process are given in Refs. 3 and 13

The characterization is conducted under ambient conditions by electrical measurements in two- and four-point configuration with an Agilent B1500A semiconductor analyzer. In general, the application of a constant voltage or current for a certain time is an appropriate method to electroform a resistively switching material.[3,9] However, our experiments showed that the time-dependent electroforming characteristic is difficult to predict and to reproduce. A low voltage or current can extend the electroforming time substantially or completely prevent the transfer of the sample into a switchable state. An excessive signal might destroy the junction or transfer it into the unipolar switching mechanism. Therefore, a sweeping signal is applied to the top electrode (Pt/Ti) which offers a limited time frame and the required amplitude for the electroforming. The use of a low slew rate of several millivolts or hundreds of nanoampere per second protects the junction from overcharge.

The virgin I(V)-characteristics, which are presented in Fig. 1(b), show an asymmetric and nonlinear rectifying behavior, especially for the positive polarity. This is the result of two different interfaces between the Pt or Ti electrodes and the TiO_2, depicted in the inset of Fig. 1(b). The thin intermediate Ti layer between the Pt top electrode and TiO_2 has a lower work function. In contrast to $Pt/TiO_2/Pt$ which

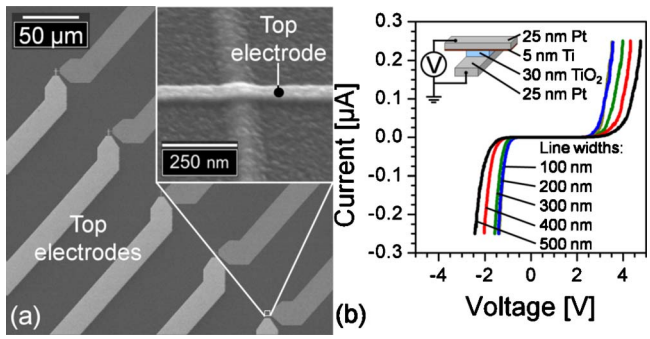

FIG. 1. (Color online) (a) Set of examined single crosspoint junctions. The inset shows the details of a 100×100 nm^2 large crosspoint cell. (b) I(v) characteristics of unformed crosspoint junctions.

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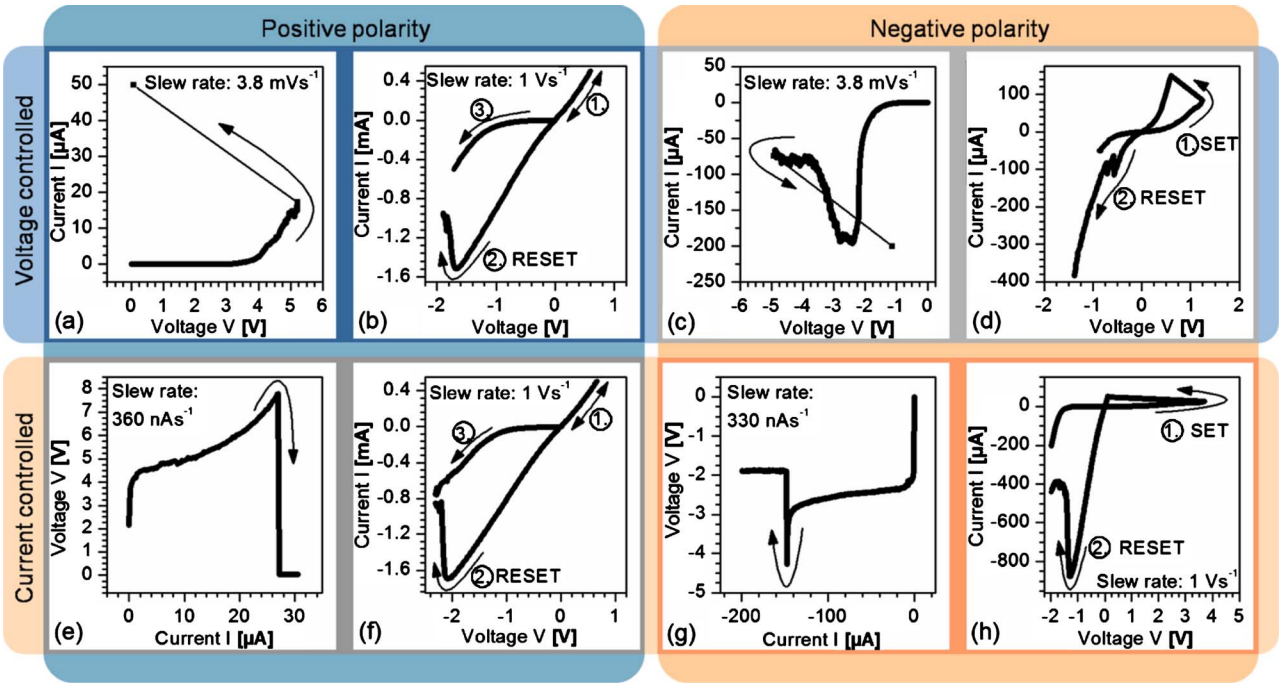

FIG. 2. (Color online) Electroforming procedures with a (a) positive voltage sweep and (b) subsequent operation, starting with a reset. (c) Forming with a negative voltage sweep resulting in a (d) HRS and a set step. (e) Positive current sweep into a (f) LRS and a reset operation and (g) negative current sweep with a (h) subsequent set sweep.

can be formed into both directions;[9] the given asymmetric system with a Pt and a Ti electrode creates a preferred switching progression. Both interfaces are additionally affected by the deposition methods and the involved energies. The application of energy for reactive sputtering is about one order of magnitude higher than for evaporation.

Thereby the initial current scales well with the junction size. A diode-like I(V) curve emerged also for the positive and negative electroforming. For the positive polarity, the current increases up to around 20 μA for comparably high voltages around 5 V before it electroforms abruptly and directly into the LRS [Fig. 2(a)] proven by the subsequent state determination shown in Fig. 2(b). As the junction is already in the LRS (R_{ON}=1.5 kΩ), it can only be switched off into the HRS by applying a negative polarity. However, for an electroforming with a negative sweep the voltage decreases to −2 V, while the current reaches around −200 μA, before it starts to oscillate and increase up to −75 μA at −5 V and finally electroforms abruptly into the HRS with R_{OFF} =120 kΩ [Fig. 2(c)]. The first switching is presented in Fig. 2(d), which starts with a positive sweep setting the device into the LRS. A subsequent triangular signal with negative polarity resets it to the HRS.

The application of a voltage sweep in combination with a decreasing resistance leads to a strongly increasing current attended by high power dissipation. Typically a thermal break or a transfer into the unipolar switching mode is observed. To prevent this effect, an appropriate current limit is necessary which enables a soft breakdown and a transfer of the junction into the bipolar switching mode. Due to deviations within the electroforming characteristic an appropriate limit is difficult to find. Although less critical, this is also an issue in case of negative polarity.

However, to bypass this aspect, a current controlled electroforming is tested. Here, the voltage increases proportional to the current depending on the resistance. As a result, at the time where the breakdown occurs, the resistance drops abruptly as well as the corresponding voltage and the power dissipation in the junction. As presented in Fig. 2(e) by the characteristic for a positive current sweep, no voltage or current limit is needed. The voltage increases up to nearly 8 V for a current of 27 μA, which is comparable to the voltage controlled forming [Fig. 2(a)]. In this case the resulting state is also the LRS as indicated in Fig. 2(f). Nevertheless, the junctions tend to transfer into a very low LRS which often cannot be switched off. The negative current sweep forms the cell abruptly into reliably functional HRS at a current of −150 μA (R_{OFF}=83 MΩ) [Figs. 2(g) and 2(h)]. As a result, the current controlled electroforming transfers the junctions into the switchable mode and avoids the use of an appropriate current limit. Both polarities prove their suitability for electroforming, but the positive sweep generates a very low LRS which is often rather qualified for unipolar switching with the need of high switching currents.

Based on these findings, negative current controlled electroforming is most reliable and used to form junctions of different sizes. In face of the deviations which motivate the forming by sweeps, a linear dependency between forming current and electrode width could be observed instead of a dependency with respect to the junction area (Fig. 3). An obvious explanation is the geometric inhomogeneity along the step where the functional layer covers the lower electrode. In this area the electric field is typically enhanced and also the layer texture might be affected. Once the junctions are electroformed, no obvious scaling of the switching parameters depending on a geometrical size could be observed as depicted in Fig. 4. Apart from some variations in the I(V)- or rather R(V)-characteristics concerning the resistance R_{OFF} and the threshold voltage V_{th}^{+} of the HRS the key parameters for the LRS were nearly constant. This observation suggests an electrode-independent size of the conducting path for the

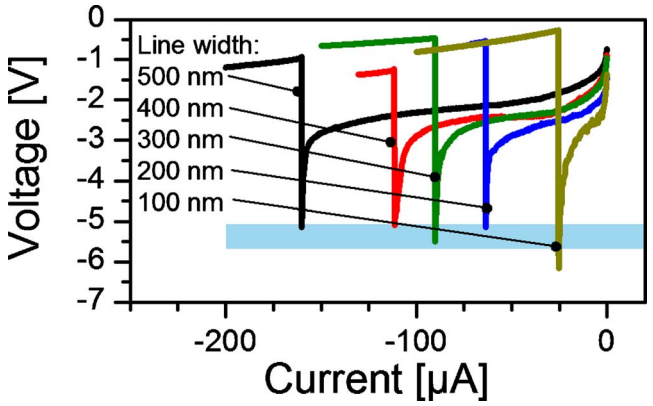

FIG. 3. (Color online) V(I) characteristics of current driven negative electroforming procedures for junctions with different metallization lines widths.

LRS as a result of a formation along the weakest point in the junction setup along the fraction of a step at a bottom electrode.

In summary, the application of a voltage or current sweep for the electroforming of resistively switching nanocrosspoint junctions is investigated for positive and negative polarities. The polarity dependence of the forming, resulting in a LRS or HRS, is in full agreement with the drift direction of the oxygen/oxygen vacancies toward anode or cathode, respectively. So, the results support the model of a semiconducting filament by oxygen vacancies which in addition alter the barrier at the Pt/TiO_{2-x} interface. Current controlled electroforming circumvents the use of a current limitation by an active power source or resistance. Investigations concerning the polarity of the forming sweep emphasize the electroforming with negative polarities to the top electrode. The resulting procedure transferred the initial high ohmic device (several giga-ohm) reliably into a HRS (range of mega-ohm), which then can be switched between LRS and HRS. The forming and switching characteristics suggest a localized, electrode size independent conduction path for the LRS.

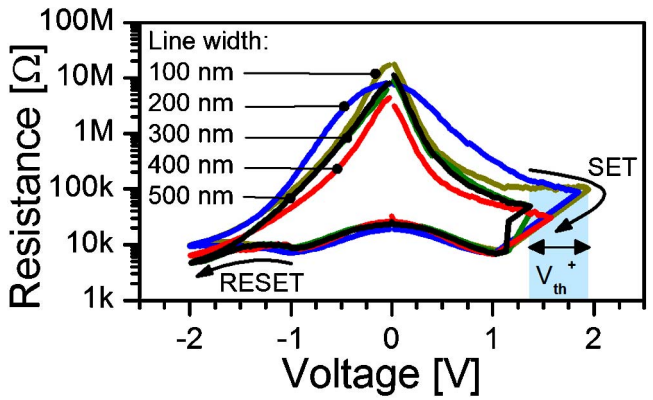

FIG. 4. (Color online) R(V) switching characteristics of junctions with sizes between 0.25 and 0.01 μm^2.

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