Experimental study of gradual/abrupt dynamics of HfO_2-based memristive devices

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The resistance switching dynamics of $TiN/HfO_2/Pt$ devices is analyzed in this paper. When biased with a voltage ramp of appropriate polarity, the devices experience SET transitions from high to low resistance states in an abrupt manner, which allows identifying a threshold voltage. However, we find that the stimulation with trains of identical pulses at voltages near the threshold results in a gradual SET transition whereby the resistive state visits a continuum of intermediate levels as it approaches some low resistance state limit. On the contrary, RESET transitions from low to high resistance states proceed in a gradual way under voltage ramp stimulation, while gradual resistance changes driven by trains of identical spikes cover only a limited resistance window. The results are discussed in terms of the relations among the thermo-electrochemical effects of Joule heating, ion mobility and resistance change, which provide a positive and negative closed loop processes in SET and RESET, respectively. Furthermore, the effect of the competition between opposite tendencies of filament dissolution and formation at opposite metal/HfO_2 interfaces is discussed as an additional ingredient affecting the switching dynamics.

THE MANUSCRIPT

Resistance switching phenomena have recently been the subject of renewed interest aimed at opening novel application windows, as theorized for the general class of memristive devices.[1–3] Indeed, their rich switching dynamics enable their use in various applications, e.g. sensing through spike integration[4], emulation of synaptic plasticity[5,6] and short term memory[7–9]. The interest in the dynamics of the switching operation is attested by the large number of dedicated works: for instance, the kinetics of a variety of resistance switching devices has been discussed extensively in ref. 10; transient resistance changes have been analyzed in TiO_x, WO_x and chalcogenide devices[6–8]. In recent works, the dynamics of internal state variables has been exploited to demonstrate the temporal

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coupling between pulse pairs.[11,12] A detailed experimental and theoretical study has been recently reported on the RESET process (transition from low to high resistance state) of Ta_2O_5-based devices.[13]

A distinction has been taking place in the last years between memristive devices featuring analog rather than digital behavior, i.e. gradual vs. abrupt resistance transitions, for both of which dedicated system designs have been developed, e.g. for neuromorphic computing.[14–16] In particular, oxide filamentary memristive devices, those whose resistance changes rely on the formation and dissolution of conductive filaments within the insulating layer, usually show gradual dynamics for RESET (from low to high resistance) and abrupt resistance change for SET (from high to low resistance). The gradual RESET transition allows a simple way of multilevel or analog programming that has been recently exploited for implementing synaptic functions useful in neuromorphic architectures.[17] On the other hand, gradual transitions for both SET and RESET would simplify the implementation of both synaptic facilitation and inhibition processes required for learning.[18] In particular, gradual transitions should be achieved by the repetition of identical spikes.[5,14,19,20]

In this context, with the present manuscript, we propose an experimental study of SET and RESET dynamics of TiN/HfO_2/Pt resistance switching devices, which have already been demonstrated to present good memory performances in terms of reliability, endurance, retention and scalability.[21–26] In particular, gradual and abrupt dynamics of switching driven by pulsed voltage ramps or by train of identical spikes are analyzed and discussed in relation to the thermo-electrochemical effects of Joule heating, ion mobility and resistance change, which provide a positive and negative closed loop processes in SET and RESET operations. It is worth noticing that only recently few works were dedicated to the obtainment of analog resistance transitions driven by trains of identical spikes in filamentary-type memristive systems,[5,19,20] though this operation mode is currently of great interest and deserve further understanding.

The devices under investigation are constituted by a 40 nm TiN / 5.5 nm HfO_2 / 50 nm Pt structure. Details of the fabrication process are reported elsewhere.[21,22,24,27] The electrical tests were performed either by using a custom memristor testing board[28] or the Source Measuring Units (B1511B and B1517A) of a B1500A Semiconductor Device Parameter Analyzer by Keysight. Throughout the manuscript, for pulsed measurements, the device resistance is read at low voltage (250 mV) after the delivery of each programming pulse. The acquisition software is able to evaluate when the measured resistance overcomes a specified target resistance and eventually stop the pulse train. It must be pointed out that in order to obtain the described resistance control no current limitation is imposed. Voltage is applied to the Pt electrode, while the bottom TiN electrode is always grounded.

Firstly, we discuss the forming process, of which figure 1a-b reports an example. The resistance measured after each programming pulse is reported in figure 1a. The voltage level of each pulse in the ramps is shown in figure 1b and the pulse timewidth is fixed to 100 μs. The resistance drops from about 1 MΩ down to about 1.5 kΩ with pulse number 10 (figure 1a). The forming process is carried out with voltage pulses without any external current limitation.

As an indication of the validity of the forming process, the subsequent RESET demonstrates that the forming is partially reversible, as expected.[21,27,29] Furthermore, the resistance evolution during RESET operation (from about 15 pulses on) displays the same features as those already reported for DC operation, i.e. an initial resistance decrease from 1.5 kΩ to few hundreds Ω followed by the actual resistance increase.[27] The entire resistance evolution has been explained as a complementary resistance switching (CRS) involving filament formation and dissolution at opposite metal/oxide interfaces in a previous publication, where the procedure to move from CRS to normal bipolar switching, and vice versa, is also described.[27] The competition between opposite interfaces is established because of the presence of TiO_xN_y interlayer that forms at the TiN/HfO_2 interface and acts as a source/sink of oxygen vacancies. The CRS operation obtained after DC forming with current compliance reported in ref. 27 has been reproduced also with pulsed forming and no current limitation in this work, which indicates that the latter forming procedure is as effective as the former. The short duration of the voltage stimuli together with the competition between switching at opposite interfaces cooperate in preventing the destructive breakdown during pulsed forming with no current limitation.

After initiation with the described forming process, resistance switching of TiN/HfO_2/Pt is obtained with pulsed voltage ramps as shown in figure 1c. Negative voltage ramps produce sharp resistance drops, i.e. SET operation., in every examined condition: timewidths of 10 μs and 100 μs are investigated as shown in figure 1c and various voltage steps, resulting in different ramp durations are employed (see supplementary information for the details). SET is performed without any external current limitation. On the contrary, RESET transitions are gradual as pulse voltage increases (figure 1c, positive branch). These results are in agreement with general observations reported in literature: for oxide filamentary memristive devices, SET transitions are usually abrupt (downward arrow for a representative SET transition in figure 1c) whereas RESET transitions are gradual (upward arrow for a representative RESET transition in figure 1c) as a function of the increasing applied voltage.[13,17,23,30] Indeed, SET/RESET operations drive processes of formation and disruption of a conductive filament that follow qualitatively different mechanisms. In SET operation, resistance decrease boosts thermal Joule heating and migration of the ions constituting the filament, which internally accelerates the process. On the contrary, in the

RESET operation, resistance increase progressively reduces the temperature in the filament slowing the process down.[17,30,31]

The switching dynamics has been investigated in response to trains of identical spikes, which is, for instance, the algorithm of choice to drive memristive devices as artificial synapses in a neuromorphic architecture, as cited above.

Let us begin the discussion from the SET transition. Figure 1c clearly indicates the existence of a voltage threshold for SET transition:[23] pulses with voltages definitely higher than such threshold trigger a SET transition and those definitely lower than the threshold do not affect the device resistance. Hence, the effect of pulse repetition has been investigated within voltage levels around the threshold.

Resistance evolution shown in figure 2a is the result of the delivering of pulses with voltages reported in figure 2b: SET and RESET are performed with trains of pulses of -0.8 V and 1.6 V until 1 kΩ and 10 kΩ target resistances are reached, respectively. The resistance undergoes a gradual resistance lowering in the SET transition for negative voltages. The five SET operations (from SET #1 to #5) reported in figure 2 show that the analog character of the transition is always respected, while a large variability in the dynamics of the process is evident. Being a phenomenon occurring across the threshold for switching,[23] such variability can be attributed to the fact that initial high resistance states can be characterized by different microscopic configurations even for the same resistance value. However, it is straightforward to recognize a common subdivision into three phases (see bars on top of figure 2a). In the first one (i), the resistance changes slowly and its duration in terms of number of pulses is subjected to a large variability (from about 10 to 100 pulses). In the second (ii), which takes about few tens of pulses, most of the resistance change occurs and, in the third phase (iii), the resistance change undergoes a saturation. Such a resistance evolution is characterized by changes of slopes (which is seldom found in the literature) and has been interpreted in terms of a two steps filamentary model in ref. 23 as a first gradual closing of the gap within the conductive filament and a following filament lateral growth. Interestingly, a similar change of slope was reported by Zhao et al. for the RESET process driven by trains of identical spikes and it was qualitatively explained by filament lateral thinning leading to the opening of a gap that then enlarges.[32]

It is interesting to notice that the SET process is abrupt when the voltage is swept through pulsed ramps, whereas it can be controlled in an analog way by carefully selecting the voltage of the pulse stimulus to be repeated. This is demonstrated in figure 2c where the resistance evolution driven by trains of identical pulses with different voltage values are used. The figure shows that high voltages drive an abrupt resistance change with no further changes due to the following pulses; pulses with slightly lower voltages apply a minor change in the microscopic arrangement of the oxygen vacancies within the interrupted filament. Such minor modifications are accumulated from pulse to pulse until leading to the formation of a thin conductive filament connecting the electrodes, in correspondence of the steep resistance decrease (phase ii). The saturation of the resistance value (phase iii) is finally explained as a process of filament growth.[23] Obviously, low voltage pulses do not succeed in modifying significantly the resistance state of the devices as evident in figure 2c for voltages of -0.70 V. A narrow window of about 100 mV useful for gradual resistance transition is found.

In the measurements reported in figure 2, the RESET operation has been performed with voltages high enough to raise the resistance to about 10 kΩ within few pulses. However, we found impossible to drive a gradual resistance change over the same resistance window in RESET operation by repeating the same stimulus. Indeed, figure 3 demonstrates that is always necessary to increase the pulse voltage (figure 4b voltages from 0.75 V to 0.95 V) to obtain a RESET operation even on a small resistance range (figure 4a from a low resistance state of 2.5 kΩ to 7 kΩ). The effect of the repetition of identical pulses is very limited as can be appreciated in the zoom of the first 150 pulses reported in the inset of figure 3.

In agreement with results obtained with voltage ramps, a continuous voltage increase is necessary (e.g. see RESET sweeps in figure 1c) to cover a significant resistance range with a gradual transition. On the contrary, the SET operation, which occurs in an abrupt way when driven by voltage ramps, can be made gradual by the use of trains of identical spikes that continuously lower the resistance over a significant resistance range.

The overall results of the present work are in agreement with electro-thermal considerations that are usually raised to explain filamentary resistance switching operation, where the drift and diffusion of ion species constituting the filaments are activated by high temperatures. Let us consider the SET operation first. Abrupt SET transitions in voltage ramps are usually explained in terms of a positive feedback loop process in which the voltage increase stimulates resistance lowering and the increasing Joule dissipation favors ion mobility and further resistance drop.[17,30,31] Indeed, a detailed analysis conducted on oxide memristor based on $SrTiO_3$ revealed that the switching kinetics undergoes a steep non-linear acceleration with the applied voltage because of Joule heating boosting ion mobility.[33] Thus, Joule heating is the driving force of this positive feedback.[31] Programming pulses with limited time duration and with an amplitude close to threshold voltage for SET (as found with voltage ramps) provide a quenching of the Joule heating and the breaking of the positive feedback loop, which results in a gradual resistance decrease. Furthermore, as cited above, the presence of a TiO_xN_y interface layer acting as a source/sink of oxygen vacancies establishes a competition between tendencies of filament formation and rupture at opposite interfaces.[27] For this reason, the positive feedback may be mitigated,[27,30] which allows obtaining gradual SET dynamics without need of any current limitation. It must be pointed out that results have been reproduced with time interval among pulses as large as 150 ms, which avoids any second order effect related to pulse-to-pulse proximity as those reported in ref.s 11,12.

On the contrary, in the RESET transition, the loop between temperature, ion mobility and resistance increase closes with a negative feedback because, as the process goes on, the resistance increase freezes ion motion.[17,30] For this reason, a continuous voltage increase is needed to counterbalance the temperature reduction and to lead the RESET process over a significant resistance range. On the other hand, even if very moderate, a certain effect of the repetition of the same pulse to the resistance in RESET is visible in the first pulses of the RESET process figure 3a (zoomed in in the inset, R_{max}/R_{min}~ 1.5). This result can be ascribed to the fact that the maximum resistance change that can be achieved with a certain voltage stimulus (e.g. 0.75 V) takes a time that is longer than the pulse duration that we applied (i.e. 100 μs). In this case, successive pulses play the role of gradually completing the interrupted RESET process. Indeed, in filamentary TaO_x-based devices, Marchewka et al. showed that the duration of the RESET process duration ranges from few ns to few μs as a result of the variation of the applied voltage by only few hundreds of mV.[13]

In summary, we attribute the cumulative effects of identical SET/RESET pulses to different processes. In the SET process, the limited time interval in which the voltage is applied is not sufficient to establish the positive feedback that leads to abrupt the resistance transitions found with voltage ramps and, in the meanwhile, the positive feedback can be mitigated by the opposite switching tendencies developing at opposite interfaces as explained above. The effect of repeated pulses on the RESET operation, even if limited, is found when amplitudes and timewidths are chosen in such a way that the process is slow enough not to be completed in one single pulse.

In this sense, our results appear qualitatively coherent with the usual description of SET and RESET processes in filamentary memristive devices. On the other hand, it must be cited that, in literature, results that move away from ours can be found regarding filamentary systems involving similar, as well as different materials. For instance, gradual transitions for both SET and RESET dynamics over a window of more that 3 (= R_{max} / R_{min}) have been found in filamentary devices base on a Ta_2O_{5-x}/TaO_y bilayer structure.[11] For what concerns HfO_2-based memristors, contrasting results can be found. For instance, the groups of Prof.s Nishi and Wong found a nice gradual RESET behavior over a significant resistance range driven by repeated identical pulses in a $Pt/HfO_x/TiO_x/HfO_x/TiO_x/TiN$ as well as in $Pt/HfO_2/TiN$ devices in two separate publications[17,32] and did not report about any gradual transition for SET operation. Gao et al. firstly reported preliminary observations on the pulse repetition number on the resistance change on $TiN/HfO_2/Pt$ devices,[20] as later confirmed by the present authors.[23]

On the other hand, few works on HfO_2-based memristors demonstrate gradual SET and RESET operations as function of the number of pulse repetitions.[5,19,34]

This literature survey demonstrates that fully understanding the switching dynamics in memristor is still a matter of study and with the present manuscript, we present the details of $TiN/HfO_2/Pt$ switching dynamics for SET and RESET in different voltage conditions. The results have been discussed in terms of the relations among the thermo-electrochemical effects of Joule heating, ion mobility and resistance change which provide a positive and negative closed loop processes in SET and RESET, respectively. Furthermore, the effect of a competition between opposite tendencies of filament dissolution and formation at opposite metal/HfO_2 interfaces is supposed to play a role in determining the observed dynamics. Related to this competition is the demonstration of forming and SET processes conducted without any current limitation, which constitutes a possibly interesting breakthrough for many practical applications, even though the reduction of the involved current still must be addressed.

SUPPLEMENTARY MATERIAL

See supplementary material for the details of the SET operation driven by trains of pulses with increasing amplitude.

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FIGURES

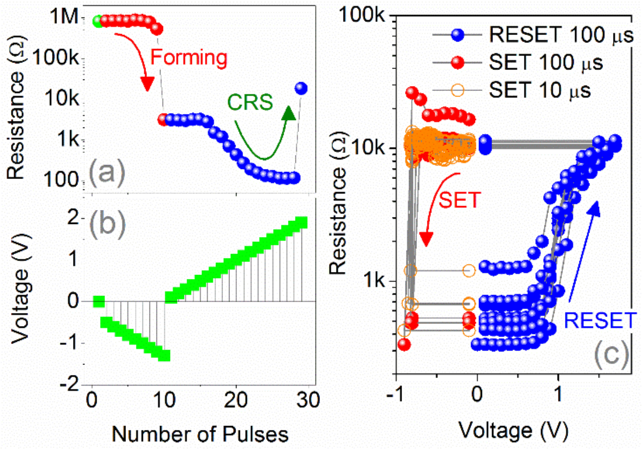

FIG. 1. (a) Resistance evolution of a representative forming and first RESET processes driven by 100 μs pulses with voltages reported in panel (b). The target resistance of 4 kΩ is set for forming process. The first reset after forming displays a typical CRS behavior. (c) Bipolar resistance switching obtained with train of pulses with increasing amplitude. The duration of the pulses is 100 μs for RESET and 100 μs and 10 μs for SET according to the symbols indicated in the legend. RESET operation in correspondence of positive pulse voltages is stopped at 10 kΩ.

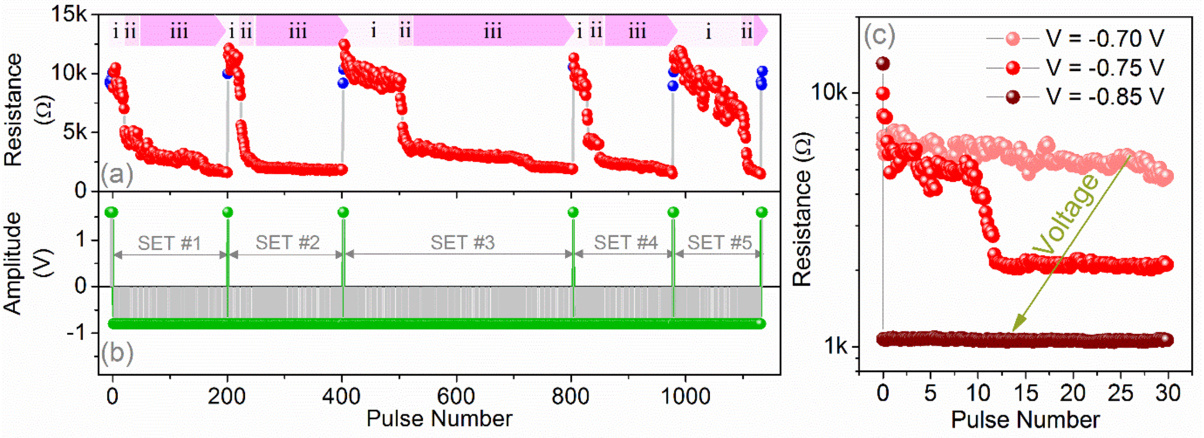

FIG. 2. Resistance switching operation under trains of identical spikes. Resistance evolution (a) shows gradual SET dynamics and sharp RESET transitions for negative and positive pulse voltages (b). Target resistances of 10 kΩ and 1 kΩ are imposed for RESET and SET, respectively. Target resistance for SET is reached in SET #3 and #4. (c) SET operation driven by trains of identical spikes for the case of -0.70 V, -0.75 V and -0.85 V. Pulses are 100 μs long.

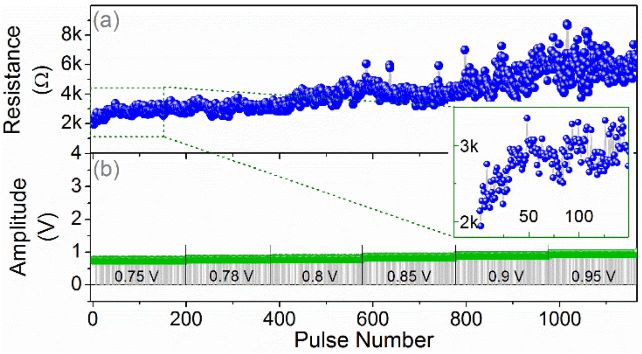

FIG. 3. RESET operation through successive six groups of identical spikes. The pulse voltage is increased from one group to the next one starting from 0.75 V to 0.95 V. Pulse duration is 100 µs. (a) Resistance values after the programming pulse and (b) voltage of the programming pulse. The inset reports a zoom around the first 150 pulses.