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A model for non-volatile electronic memory devices with strongly correlated materials

M.J. Rozenberg[a,b,*], I.H. Inoue[c], M.J. Sánchez[d]

[a]Laboratoire de Physique des Solides, CNRS UMR 8502, Université Paris-Sud, Orsay 91405 cedex, France
[b]Departamento de Física Juan José Giambiagi, FCEN, Universidad de Buenos Aires, Ciudad Universitaria Pabellón I, (1428) Buenos Aires, Argentina
[c]Correlated Electron Research Center (CERC), National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba 305-8562, Japan
[d]Centro Atómico Bariloche, (8400) San Carlos de Bariloche, Argentina

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Abstract

The behavior of a model for non-volatile electronic memory devices is discussed. The resistance switching mechanism that gives place to the memory effect is due to an effective doping driven transition in small domains at the interface.

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To take advantage of strong correlation effects for the design of electronic devices has long been a goal of solid state physicists. Significant progress has been achieved in recent years in fabrication techniques such as pulsed laser deposition, which is opening the way for the fabrication and investigation of metal–insulator–metal (MIM) structures that display different types of resistance switching. These devices are prototypes for two-terminal non-volatile random access memories and the resistance switching is obtained through the application of a short voltage or current pulse. The insulator of these MIM structure is usually a transition metal oxide [1–5] in which the electron correlations might be expected to play an important role. The switching effects are normally correlated with observed hysteresis in the *I–V* characteristics. These systems are collectively called resistance random access memories (RRAM) and the large resistance variations with applied voltage, i.e., external electric field is termed colossal electroresistance effect (CER).

The goal of the present work is to investigate the predictions for the switching and hysteresis properties of a recently proposed model [6,7] that introduces strong correlation effects due to carrier injection as the driving mechanism for CER. We shall first demonstrate how the model captures the qualitative aspects of resistance switching and then correlate the switching to the predictions for the *I–V* characteristics and the microscopical state of the system. We shall argue that an effective doping driven transition at the level of small nanoscopic regions near the electrodes is responsible for the switching and hysteresis effects. We also discuss what are the implications of such a scenario, in particular for non-volatility and switching speeds.

The model (Fig. 1) assumes the existence of an insulating (and inert) medium with a non-percolating structure of metallic domains that might correspond to defects, grains, phase separated regions, and so on [6]. Smaller domains are closer to the electrodes and are called “top” and “bottom”. A large domain occupies the bulk of the system that is called “central”. Carriers tunnel between domains under the action of an external electric field (or applied voltage). The probability of charge transfer depends on phenomenological parameters such as the tunneling rates, the number of states

* Corresponding author. Laboratoire de Physique des Solides, CNRS UMR 8502, Université Paris-Sud, Orsay 91405 cedex, France.
E-mail address: rozenberg@lps.u-psud.fr (M.J. Rozenberg).

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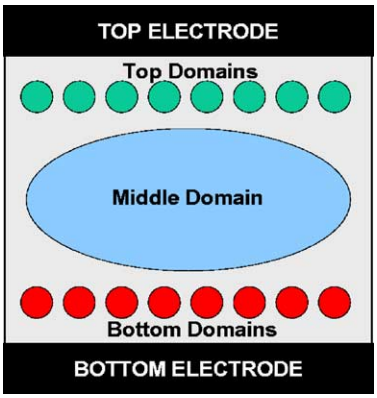

Fig. 1. Schematic view of the model with top and bottom electrodes (black), insulating medium (grey), smaller top and bottom domains (green and red) and large middle domain (blue). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

in the domains and their occupation (see Ref. [6]). The model is mathematically defined by the following system of rate equations,

$$\frac{dn_i^b}{dt} = \Gamma_i^{eb} N_e n^e \left(1 - n_i^b\right) f_{eb}(V) - \Gamma_i^{bc} n_i^b N_c (1 - n^c) f_{bc}(V) \quad (1)$$

$$\frac{dn^c}{dt} = \sum_i \left[\Gamma_i^{bc} N_b n_i^b (1 - n^c) f_{bc}(V) - \Gamma_i^{ct} n^c N_t \left(1 - n_i^t\right) f_{ct}(V)\right] \quad (2)$$

$$\frac{dn_i^t}{dt} = \Gamma_i^{ct} N_c n^c \left(1 - n_i^t\right) f_{ct}(V) - \Gamma_i^{te} n_i^t N_e (1 - n^e) f_{te}(V) \quad (3)$$

where $\Gamma_i^{\alpha\beta}$ denote the tunneling rates between the electrodes and domains, and α, β=e, t, c, b denote "electrode", "top", "central" and "bottom" and the subindex i labels the domains. N_α is the total number of states in electrode or domain α, and n^α is the occupation. $f_{\alpha\beta}(V)$ describes the dependence of the transfer probabilities between α and β on the given external voltage protocol $V(t)$. The actual value of these functions are material dependent [8] and their determination is beyond the scope of the present work.

The domain structure assumed in this model is motivated from a rather universal aspect of strongly correlated perovskites: the spatial inhomogeneity (or phase separation) that occurs at the nanoscale. This has recently emerged as one of the most spectacular properties of TMOs and possibly responsible for much of the interesting physics that they display [9,10]. Clearly, its origin is in the variety of competing electronic states that the strong correlation in perovskite 3d-electron systems bring about [11]. This feature also leads to another important and ubiquitous characteristic of TMOs that of usually having a reach phase diagram, where different phases can be access through pressure, temperature, doping, etc. In fact, the possibility of a doping driven transition within the electronic state of the domains was proposed in a recent paper [7] as the mechanism responsible for resistance switching. Following that work we allow for a doping driven metal–insulator transition in the present work, so when the level of occupation of a domain attains a given threshold we shall assume that the tunelling function $f(V)$ in Eqs. (1)–(3) is renormalized by a factor that takes into account the decrease of states at the Fermi energy when a (Mott) gap opens in the density of states of the domains. Concretely, we take for the tunelling function $f_{mit}(V)=e^{-\Delta/T} f(V)$ if the occupation of the corresponding domain is within 10% of half-filling, or else $f_{mit}(V)=f(V)$. Δ is the size of the Mott gap and T is the temperature. There are two important points to make: i) it is crucial to emphasize that the phase control operates *only* at the level of the small domains through carrier injection. Evidently, the amount of charge that one can inject to the MIM structures is not enough to macroscopically dope them, however, the small domains are structures of typical size of tens of nanometers [12] and thus even small amounts of charge would be sufficient to effectively modify their doping (or occupation) state. ii) While we are here considering the particular case of a metal–insulator transition at the level of the domains, in general, any transition driven by doping would produce qualitatively similar results for the resistance switching properties. The key point being that as one tunes from one phase to another, the conductance properties due to the variations in the number of states at the Fermi energy, are expected to be different in each phase.

In this work we set $\Gamma_i^{eb}=\Gamma_i^{te}\equiv\Gamma^{ext}=0.3\times10^{-15}$, $\Gamma^{tc}=\Gamma^{cb}\equiv\Gamma^{int}=1.0\times10^{-13}$, the number of top and bottom domains to 40 and the number of states that they hold to 10^6; the sole central domain holds 10^9 states. The actual number of domains is not a crucial quantity of the model and has been rather arbitrarily set. The actual realistic number might hopefully be obtained by imaging techniques such as those of Ref. [13]. On the other hand, the number of states in the domains are such that produce carrier densities of similar magnitude as those experimentally reported [3].

We now turn to a discussion of the results. The basic switching behavior of the model is demonstrated in Fig. 2. The results correspond to a single Monte Carlo simulation run of 6000 time steps or units of time (uot). The system starts in an initial state in which all small domains are 20% filled and the center domain is 10%. These small occupations are consistent with systems such as lightly Cr-doped $SrTiO_3$ [2,14]. The top panel of Fig. 2 shows the voltage protocol. An external direct-current (dc) voltage bias V_{read} is continuously applied between the top and bottom electrodes and produces a carrier current. At given time intervals of 1000 uot [15], voltage pulses of short duration (10 uot) are applied. The mid panel shows the current at the top electrode, and we see that positive voltage pulses cause sharp positive current spikes while the negative pulses cause negative spikes.

The bottom panel also shows the current but in a different vertical scale that reveals more detailed behavior between the pulses in the presence of a small dc bias V_{read}.

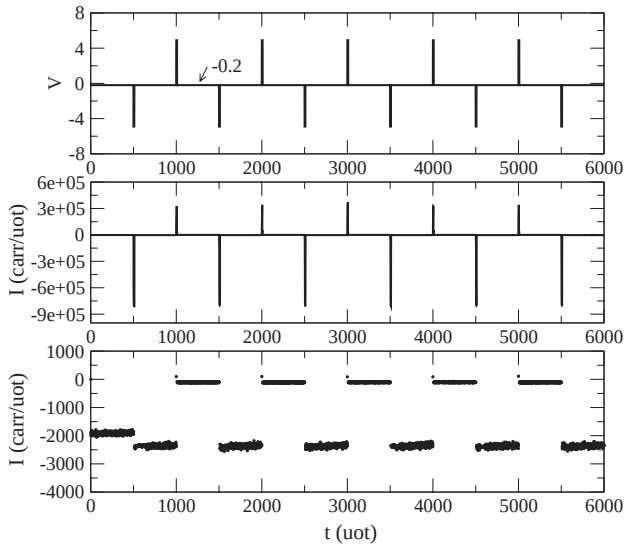

Fig. 2. Model simulation results for N_t=N_b=10^6, N_c=10^{10} and Γ^{int}=2×10^{-8}, Γ^{ext}=6×10^{-14}. The top panel shows the applied voltage protocol. The mid bottom (enlarged scale) panels show the current at the top electrode.

The most important point to note is that voltage pulses switch the system between two conductance (or resistance) states corresponding to high and low current. It is interesting to note that the system starts in an arbitrary initial state (assumed for simplicity with all domains half-filled) thus having an arbitrary R. The action of a write pulse does not properly switch the system to "1", and only after an erase pulse is applied the system gets properly initialized and begins switching between well defined I-states. This behavior is qualitatively observed in a variety of real RRAM systems [2,14,16].

The switching properties are directly related to the existence of hysteretical behavior. In fact, for the same values of the model parameters, we run simulations for the behavior of the system under a voltage ramp to obtain the I–V characteristics and we observed the hysteresis loop shown in Fig. 3. The voltage protocol is $-V_{max}\rightarrow0\rightarrow V_{max}\rightarrow0\rightarrow-V_{max}$ and the duration of the whole cycle is 3000 uot. Our results are in strikingly good qualitative agreement with those of Ref. [14].

In order to gain more insight on the mechanism for the resistance switching we can observe the occupation of the domains along the hysteresis cycle. The results are shown in Fig. 4. There are several points worth making: The sharp switching seen in the hysteresis correlated with the occupation of the top domains crossing the set threshold for the Mott metal insulator transition and opening of a correlation gap *within* the top domains electronic density of states. Only the occupation of the top domains and not of the bottom domains cross the threshold, thus the current switching is controlled by a single electrode interface. In fact, a model with small domains only on one of the two interfaces would also have a parameter regime with similar switching and hysteresis characteristics. In other words, the

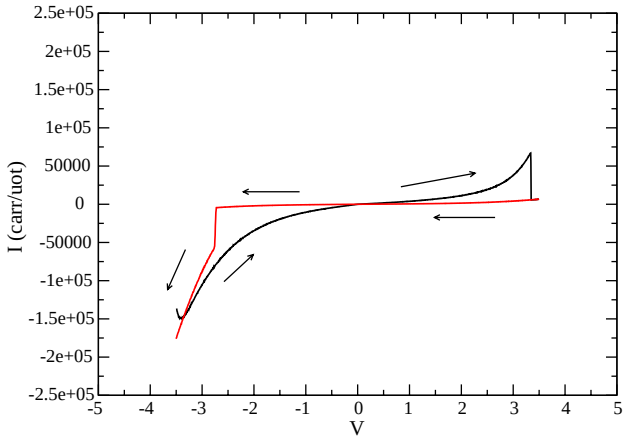

Fig. 3. Hysteresis loop for the current–voltage I–V characteristics. The voltage protocol is $-V_{max}\rightarrow0\rightarrow V_{max}\rightarrow0\rightarrow-V_{max}$.

presence of small domains at both interfaces is not strictly required. The occupation of the central domain maintains a rather constant level of occupation, thus it effectively acts as a buffer that decouples the behaviors of the two interfaces. Increasing the parameter N_c that sets the number of available states in the center domain has the effect of reducing the variation of its occupation level while does not modify the switching or hysteresis characteristics.

In conclusion, we have seen that the switching mechanisms is related hysteresis in the I–V characteristics, and that the hysteresis is itself related to a conjectured metal–insulator transition at the level of small domains. This transition is driven by the effective doping at the level of small domains due to the systems heterogeneities near the interfaces. This hetherogeneity might be of either electronic or structural origin. For instance, in lightly doped STO the Fermi energy is about 100 K and is very similar to the frequency of an intense phonon mode. It is worth noting that

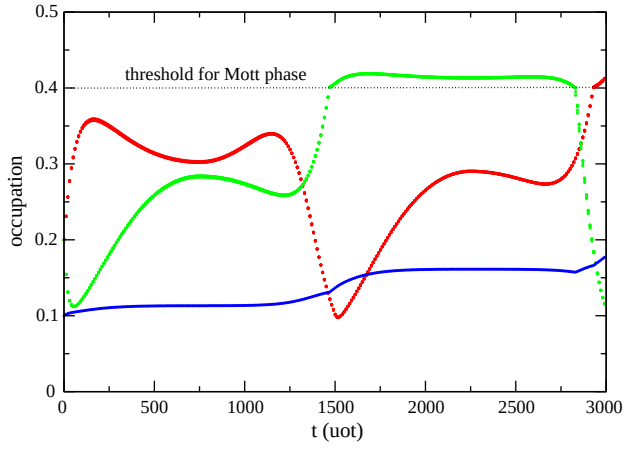

Fig. 4. Occupation of the top (green), center (blue) and bottom (red) domains as a function of the time. The applied voltage protocol is identical to that of Fig. 3. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

this may be the reason why over 1 day of "forming" is required in the STO system to start exhibiting good switching and hysteresis properties. On the other hand, inhomogeneities in manganite systems are likely to be of a purely electronic origin, as phase separation and very complex phase diagrams are a hallmark of perovskite manganites. Thus in the present scenario, the key point is that strong electric fields can inject charge into insulating systems, and that this charges go to inhomogeneous regions of the systems, that we call domains. The domains that receive charge are thus subject to an "effective doping" that may drive them across a boundary between two distinct electronic phases. These phases, whatever their particular nature, would in general have very different conductance properties, and thus lead to a switching mechanism as we described here for the particular case of a Mott-like metal insulator transition.

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