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Retentivity of RRAM Devices Based on Metal / YBCO Interfaces

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ABSTRACT

The retention time of the resistive state is a key parameter that characterizes the possible utilization of the RRAM devices as a non – volatile memory device. The understanding of the mechanism of the time relaxation process of the information state may be essential to improve their performances. In this study we examine RRAM devices based on metal / YBCO interfaces in order to comprehend the physics beneath the resistive switching phenomenon.
Our experimental results show that after producing the switching of the resistance from a low to a high state, or vice versa, the resistance evolves to its previous state in a small but noticeable percentage. We have measured long relaxation effects on the resistance state of devices composed by metal (Au, Pt) / ceramic YBCO interfaces in the temperature range 77 K – 300 K. This time relaxation can be described by a stretched exponential law that is characterized by a power exponent n = 0.5, which is temperature independent, and by a relaxation time τ that increases with increasing the temperature. These characteristics point out to a non-thermally assisted diffusion process that could be associated with oxygen (or vacancy) migration and that produces the growth of a conducting (or insulating) fractal structure.

INTRODUCTION

One of the crucial points for the development of information and communication technologies is the search of new memory devices. The Si-based devices are reaching limitations in information density, endurance and power consumption [1]. This is why a new paradigm is needed to store information in a reliably way. One of the possible candidates to overcome these technological challenges is the resistive memory devices (RRAM) based on the resistive switching (RS) mechanism, which have shown excellent properties in scalability, power consumption and operation speed, making them one of the more promising candidates to replace the actual flash memories [2-3]. One of the particular properties that must fulfill a non – volatile memory device is data retention over time. Here we propose to perform a detailed characterization of the lost of data retention by studying the time relaxation process after generating a switching in the remnant resistance state in $YBa_2Cu_3O_{7-\delta}$ (YBCO) / metal (Au,Pt) interfaces in order to gain insight on the physical mechanism beneath the RS on these complex oxide devices.

EXPERIMENT

In this work, we sputtered different metals (Au,Pt) on the surface of a optimally-doped ceramic YBCO sample (T_c 90 K and J_c(77 K) 10^3 A/cm^2) in order to investigate the time relaxation of the resistive states. The YBCO sample was prepared following the same procedures described elsewhere [4]. The sputtered electrodes, depicted in figure 1, have a width of 0.5 mm and a mean separation (6 $\pm$ 2) mm. They cover the entire width of one of the faces of the YBCO slab (8 x 4 x 0.5 mm^3). Finally, silver paint was used carefully to fix copper leads without contacting directly the surface of the sample.

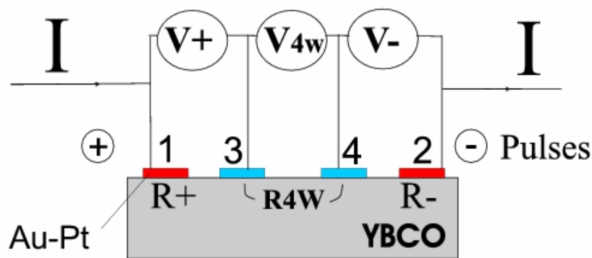

Figure 1. Contact configuration used to study the time relaxation of the remnant resistance state after producing a RS on the YBCO / metal interface. Pulses are applied on electrodes 1-2 while their remnant resistance is measured by applying a small bias current on the same electrodes and by measuring the voltage drop as indicated in the text.

Following our previous studies [5-6], at a fixed temperature (77 K < T < 320 K), we apply trains of 20,000 square pulses with an amplitude up to +5 V and 0.1 ms width at a frequency of 1 kHz to obtain a reproducible switching behavior. After that, a small bias current (~10 μA) was applied in electrodes 1 and 2 to measure different resistances using a standard DC technique; by measuring the voltage in electrodes 1 and 3 we essentially evaluate the resistance near the interface corresponding to electrode 1, as the YBCO bulk resistance between electrodes is negligible (as confirmed by measuring the four terminal resistance R_{4W}). Similarly, when we measure the voltage between electrodes 4 and 2; we essentially evaluate the resistance near the interface of electrode 2, which was always grounded. We arbitrarily call electrode 1 as R^+ (V_{13}/I_{12}) and electrode 2 as R^- (V_{42}/I_{12}.)

The initial resistance of the interfaces was in the range of 20 to 50 Ω, while the bulk YBCO resistance was about 0.1 Ω for T above T_c. Thus, as already mentioned, the bulk contribution was always small or negligible. Temperature was measured with a Pt thermometer well thermally anchored to the sample, in order to detect self heating effects. When the applied power exceeded 1 W, an increase of 2 – 4 K due to self heating was observed, followed by a 10 – 20 s decrease of the temperature to its settled value. After that transient period, temperature was controlled to remain constant within a $\pm$ 0.5 K interval during all over the relaxation measurement (up to ~ 300 minutes).

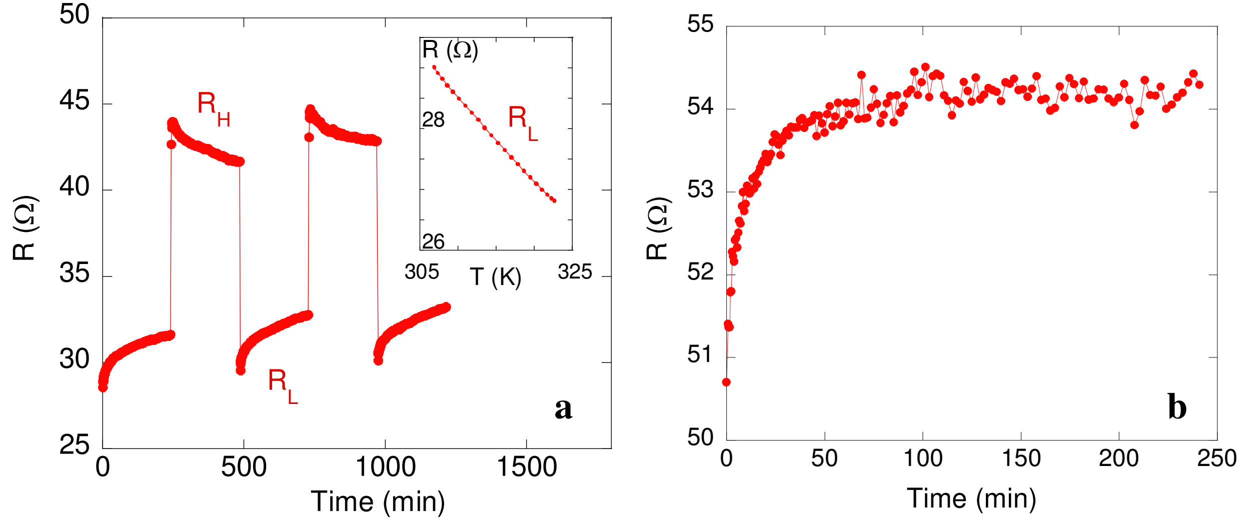

Figure 2. **a)** Time relaxation of the resistance state (R_H or R_L) after the switching process of the contact labeled R^+. The inset shows the typical semiconducting-like temperature dependence of the R_L state. **b)** Time relaxation of the remnant resistance state after producing a RS at T=123 K.

The time evolution of the remnant resistance of R^+ after applying the pulse train at a fixed temperature is shown in figure 2a. A similar behavior was obtained for R^-. It is clear that after each pulse train, the resistance relaxes towards its previous resistance state. Self-heating can be easily ruled out by considering the semiconducting-like temperature dependence of the resistance for each state, as shown in the inset of figure 2. If the main reason for the evolution of the resistance over time was related to this effect, then we should always observe a sudden reduction and a slow increase in resistance after applying the pulses as a consequence of the sudden increase of temperature and its slow reduction as the Joule heating dissipates, regardless of the state of resistance considered. This is not the case; we can consider that the observed evolution is related to a meta-stable state which evolves towards a more stable one. Relaxation effects were also reported in manganite-metal junctions [7] and interpreted as an evidence of oxygen diffusion in these complex oxide systems.

A detail of a single time relaxation of the remnant interface resistance after the RS with a saturation clearly defined is displayed in figure 2b. Each time relaxation can be described by the following equation:

$$\frac{R-R_0}{\Delta R} = X = 1 - e^{-\left(\frac{t-t_0}{\tau}\right)^n} \quad \textbf{(1)}$$

where R and R_0 are the resistances at time t and t_0 respectively, ΔR the total variation of R for $t \rightarrow \infty$, X the relative variation of resistance, τ the characteristic time and n an exponent. This equation is similar to the one established by Avrami [8] in order to describe the kinetics of a phase transition at a fixed temperature, where X represents the fraction of the volume that has transformed from one phase to another after a time t-t_0, and the exponent n is associated with the dimensionality of the growing process.

Equation **(1)** can be rewritten as follows:

$$Y = \ln\left[-\ln\left(1 - X\right)\right] = n\ln(t - t_0) - n\ln(\tau) \qquad \textbf{(2)}$$

which can be used to fit the data and determine the n and τ parameters. As can be observed in Figure3a, the linearized data is well represented by equation **(2)**.

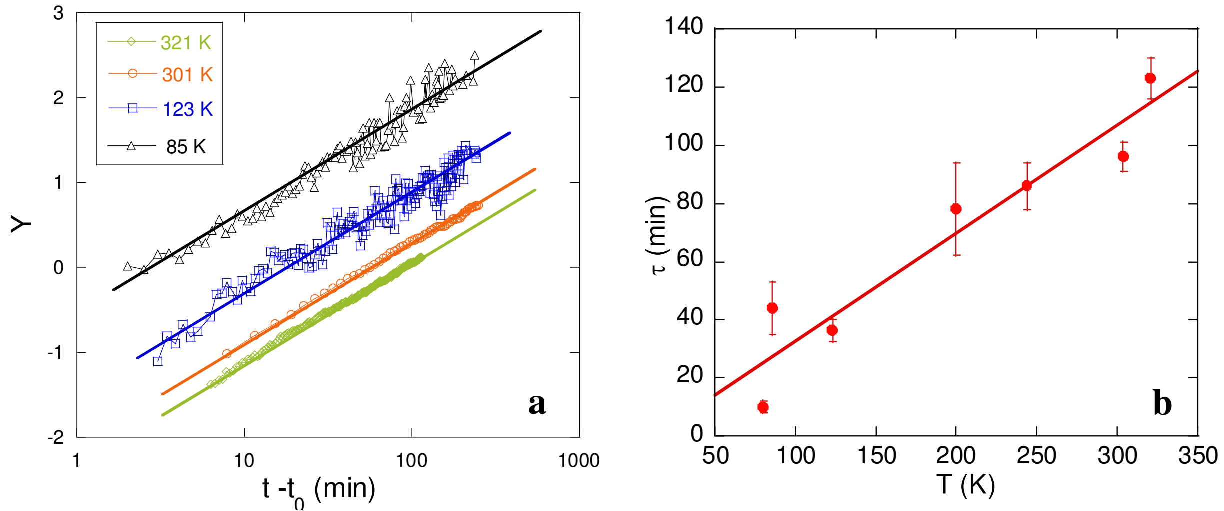

Figure 3. **a)** Linearization of the time evolution of the relative resistance variation by following equation (2), after producing a RS at different temperatures. **b)** Temperature dependence of the relaxation time τ in the range of 77 K to 320 K.

We have repeated the process for several temperatures in the 77 K to 320 K range; in all cases we obtained an exponent $n = 0.50 \pm 0.07$, independently of the resistance state, the temperature or the metal used for the electrode (see figure 3a). Note that the data cover almost two orders of magnitude without leaving the linear tendency. Additionally, we found that τ follows a linear relationship with T, as shown in figure 3b.

DISCUSSION

If we consider that the observed changes in resistance are linked to the evolution of a conductive phase growing over an insulating phase (or vice versa) due to a phase imbalance generated by the application of the electrical pulses, the similarity to the description of Avrami becomes natural. The obtained dimensionality (n), close to 0.5, points to a discontinuous 1D structure, like a dendritic structure with multiple cuts. A possible visualization of this structure is provided by the Feigenbaum fractal [9]. Within this framework, we can infer that the evolution to equilibrium of the phase unbalance generates a growing fractal structure in which a conductive phase is growing inside an insulator one (or vice versa) in a discontinuous form.

The fact that the relaxation time τ increases with increasing temperature is quite unexpected and clearly rules out that the growing of the fractal structure is solely assisted by thermal energy, as should be expected for a standard diffusion process. A non-monotonic temperature

dependence of the relaxation time, showing a local increase of τ with increasing temperature, was also observed for other systems, like in quasi 2D monolayers [10], where the hole spin relaxation time shows a non-typical temperature behavior, that was associated with carrier interactions instead of the traditional source of scattering related to electrons and phonons, which usually dominates at high temperatures. In a similar way, our results indicate that the dominating interaction that governs the time evolution of the resistance is not a phonon-assisted process.

CONCLUSIONS

We have shown the time relaxation characteristics of the non – volatile resistance state of YBCO / metal junctions as a function of temperature. The resistance evolves to an equilibrium value towards its previous state following a stretched exponential law with a temperature independent exponent $n = 0.5$ and a relaxation time τ that increases with increasing temperature. This behavior indicates that the diffusion of oxygen (or vacancies) follows a particular law, which, by analogy with the Avrami theory, may be interpreted as an indication of the dimensionality of the diffusion path. The $n = 0.5$ exponent points out to a growing phase having a fractal structure, while the temperature dependence of τ indicates that the diffusion is not dominated by thermally assisted processes. More experimental work is needed to reveal the origin of the additional dominating process.

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