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Electric pulse-induced resistive switching in ceramic $YBa_2Cu_3O_{7-\delta}$/Au interfaces

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ABSTRACT

We show the existence of a reversible, complementary and polarity dependant electric pulse-induced resistance (EPIR) switching effects in Au/$YBa_2Cu_3O_{7-\delta}$ ceramic superconductor interfaces. Non-volatile high and low resistance states and transition regions between them are obtained as a function of the amplitude and polarity of the pulsing voltage. Relaxation processes of the resistivity after applying the pulses, not associated with heating effects, are also observed. We also report on the temperature sensitivity of these resistance hysteresis switching loops, where both the difference between high and low resistance states and the voltage needed to produce the switching decrease with increasing temperature. Our results are consistent with a mechanism for the EPIR effect based on oxygen electromigration.

1. Introduction

Considering the fact that, in a near future, traditional memory devices based in semiconductors will reach their physical limits in the race to produce continuously smaller electronic equipments, many efforts has been devoted in the last years in order to obtain new materials and mechanisms to overcome this problem. One of the promising technologies corresponds to resistive random access memories (RRAMs) based on the electric pulse-induced resistive change effect (EPIR effect) [1]. This effect has been observed in a large variety of transition metal oxide thin films [2–10], ceramic manganites [11,12] and cuprate superconductors [13,14]. The RRAMs are constructed by placing an oxide (typically a bad conductor) sandwiched between two metal electrodes. The origin of the resistance change depends on the materials and it is nowadays an open question to be addressed. However, two mechanisms were revealed: one, unipolar, which needs a forming process, usually present in simple oxides, and the other, bipolar, which probably may be associated with oxygen electromigration, and seen on more complex oxides. We have recently shown the existence of the EPIR effect in Au/$YBa_2Cu_3O_{7-\delta}$ (YBCO)/Au interfaces. We have shown that upon applying electric pulses the superconducting state of YBCO in regions near the electrodes can be reversibly removed and restored. We also showed by performing four-wire measurements that pulsing also induces significant non-volatile changes in the bulk resistance. In this paper, in order to gain insight on the underlying mechanism of the resistive switching in the Au/YBCO interfaces, we analyze the sensitivity of the switching effect to the amplitude and the sign of the pulsing voltage and its dependence with temperature.

2. Experimental

In order to perform EPIR effect experiments we sputtered four Au electrodes (labeled 1, 2, 3 and 4) on one of the faces of an optimally doped YBCO ceramic superconductor as it is depicted in Fig. 1(a). The applied pulsing (on electrodes 1 and 2) consisted of trains of 20,000 square pulses of 10 V and 0.1 ms width at 1 kHz. After waiting 1 min to obtain a stable temperature (the pulsing generates an increase of ~1 K on a thermometer well thermally anchored to the sample), a small bias current applied through electrodes 1 and 2 is used to measure different resistances: by measuring the voltage in electrodes 1 and 3 we essentially evaluate the resistance near the interface corresponding to electrode 1 as the resistance in series coming from the bulk part of the YBCO between electrodes is negligible (as confirmed by measuring the four terminal resistance R_{4W}). The same occurs when we measure the voltage between electrodes 4 and 2; we essentially evaluate the resistance near the interface of electrode 2. We arbitrarily call R_+ the former resistance (V_{13}/I_{12}) as it follows the polarity of the applied pulses while the latter corresponds to R_- (V_{42}/I_{12}).

3. Results and discussion

The EPIR switching effect on both pulsed electrodes can be observed in Fig. 1(b). A complementary switching of the R_+ and R_- resistances to a low and a high resistance state (LRS and HRS,

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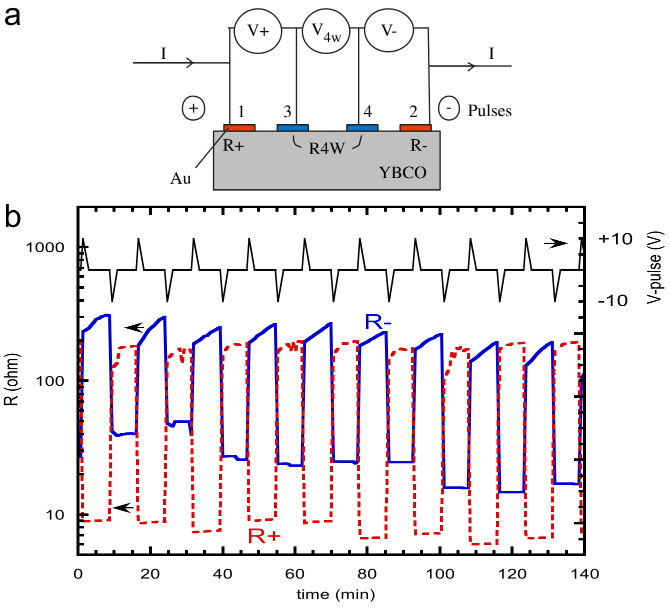

Fig. 1. (a) Diagram of the contact configuration used to study the EPIR switching on YBCO/Au interfaces and (b) resistance of the positive (R_+) and negative (R_-) electrodes as a function of time after applying successively a positive and a negative train of electric pulses (V_{-pulse}) on both electrodes. The resistance of each contact is measured using a small testing current. The switching is complementary and corresponds to a decrease of resistance after applying a positive train of pulses on a particular electrode. (A negative train of pulses on electrode "+" is a positive train of pulses on electrode "−".)

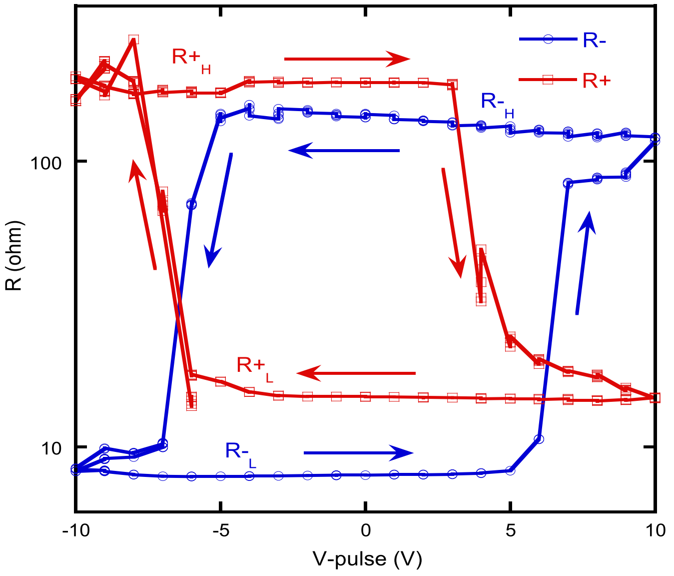

Fig. 2. Resistance hysteresis switching loops (RHSL) at room temperature for both pulsed contacts. Depending on the amplitude and on the sign of the pulsing voltage a high (H) and a low (L) resistance state can be achieved on each electrode. The switching from one state to the other always occurs firstly on the most resistive contact. The arrows indicate the sense of the evolution of the resistance when performing a pulsed-voltage loop.

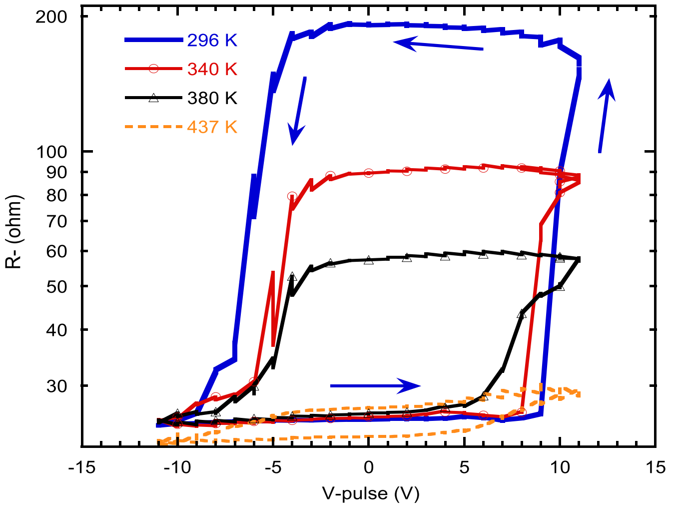

Fig. 3. RHSL of the "−" electrode for different temperatures. As temperature increases both the voltage needed to produce the switching (V_S) and the relative variation between R_H and R_L decreases. The arrows indicate the sense of the evolution of the resistance when performing a pulsed-voltage loop.

respectively) is observed upon applying successive trains of ±10 V pulses, with an overall variation of an order of magnitude between both states. Relaxation effects on the HRS of both contacts and a bad repeatability of the LRS or the HRS for each contact can be observed.

If the amplitude of the pulses (V_{-pulse}) is varied in order to perform a loop between ±10 V, the sensitivity of R_+ and R_- to the pulsed voltage can be obtained. The traced curves at room temperature, called resistance hysteresis switching loops (RHSL) are plotted in Fig. 2. The transition from the HRS to the LRS always occurs for a lower voltage than the reverse transition, indicating that the relevant parameter for the switching is the electric field (not the current).

In order to gain insight on the switching mechanism we performed RHSL of both contacts at increasing temperatures from 300 to 440 K. Fig. 3 shows the plots for R_- while in Fig. 4 we have plotted the summation of both contact's resistance. It is clear from the former figure that both the relative variation between the LRS and the HRS ($\Delta R/R$) and the voltage needed for the switching (V_S) decrease with increasing temperature (shown in Fig. 5). Fig. 4 shows a shape previously reported ("table with legs") [15] that arises from the fact already mentioned that the transitions from HRS to LRS and vice versa occurs at different pulsing voltages. It is interesting to note that depending on the value of the resistance of each contact a multilevel memory state for $R_+ + R_-$ can be performed with different high and low states depending on the value of V_{-pulse}. Another interesting feature to note in Fig. 4 is that the resistance relaxation, noticed as a sawtooth structure, is not always opposite to the resistance change induced by the pulse. This means that the pulse generates new equilibrium states that may have a higher or a lower resistance than those obtained immediately after the pulsing treatment.

A simple explanation of the temperature behavior observed in the RHSLs can be based on the temperature dependence of the high and low resistance states of the contacts, shown in particular for R_- in Fig. 6 before and after a pulsing treatment at 340 K. A semiconducting like dependence is obtained, with different activation energies.

As the resistance of the bulk well oxygenated YBCO is metallic, we can assume the existence of two oxygen depleted layers in series near the Au/YBCO interface: one with resistance R_-^L and the other with resistance ΔR_-, which gives a total resistance in series $R_-^H = R_-^L + p\,\Delta R_-$, with p a pulsing dependent factor that takes two values, 0 or 1. Thus, we are supposing that one of the layers may be short circuited ($p = 0$) or restored ($p = 1$) as a consequence of the pulses. Using this simple model, we calculate the expected temperature dependence of $\Delta R_-/R_-$, obtaining a good estimation of the measured behavior, shown as the dashed line in Fig. 5. As

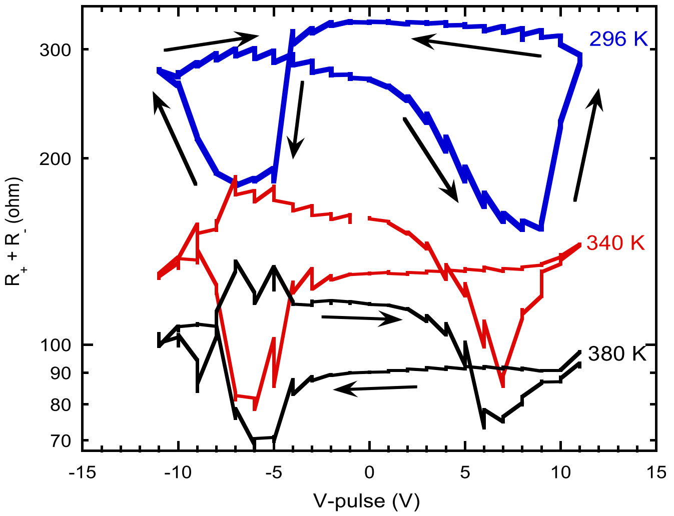

Fig. 4. RHSL of the series resistance $R_+ + R_-$ at different temperatures. Relaxation effects can be observed as a sawtooth structure. The observed minimums are due to the fact that the switching from high to low resistance states for both contacts is not simultaneous.

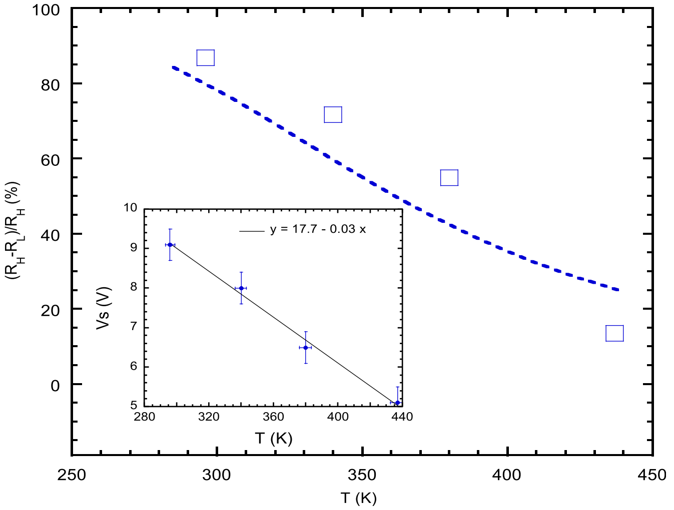

Fig. 5. Temperature dependence of the relative variation between the high and the low resistive state. The dashed curve represents the predicted behavior assuming a simple model with two interfaces (described in the text) and considering the temperature dependence of the resistance of each interface. The linear temperature dependence of the switching voltage (V_S) is shown in the inset.

YBCO, and more generally, perovskites are considered excellent oxygen-ion conductors as they possess a large number of vacancies in the oxygen sublattice (specially at grain boundaries) and a small barrier for oxygen migration [16], we may associate the resistive variations induced by the pulsing with the electromigration of oxygen. The origin of the resistive relaxation effects can then be related to oxygen diffusion. The variation of the p factor can then be due to the migration of oxygen in or out a YBCO layer which becomes metallic or semiconducting, respectively. This layer should present an extended area where the oxygen exchange with the surrounding material or with the atmosphere is favored, while the other layer, associated with R_-^L, may be located on the YBCO surface in direct contact with the sputtered electrode, where the exchange of oxygen may be prevented. Additional experiments are needed to clarify this last issue.

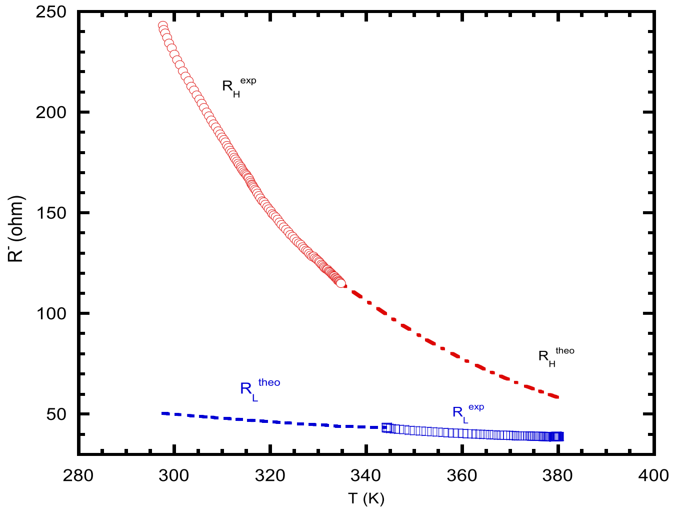

Fig. 6. Temperature dependence of the resistance of the "–" electrode in its high and low state (R_H^{exp} and R_L^{exp}, respectively) before and after a pulsing treatment at 340 K. R_H^{theo} and R_L^{theo} are their respective extrapolations assuming a semiconducting behavior.

4. Conclusions

We have shown the existence of polarity dependent resistance switching effects on Au/YBCO interfaces which are temperature dependent and present long resistive relaxation effects. Our results can be predicted by a simple model considering an oxygen-ion migration mechanism that, upon pulsing, removes or restores the metallic conductivity of a YBCO layer in a region near the electrodes.

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