Resistive Switching in Semimetallic SrIrO₃ Thin Films

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KEYWORDS

Resistive switching, metal-insulator transition, SrIrO₃, conductive atomic force microscopy, iridate thin films

ABSTRACT

Local electrical properties, measured by conductive atomic force microscopy, of semimetallic SrIrO₃ thin films are reported. The appearance of an Anderson-type metal-insulator transition (MIT) triggered by disorder and spatial localization due to film thickness reduction is analyzed, as well as their influence on the resistive switching behavior. For thin enough films (below ~3nm) samples are insulating with hysteretic I-V curves indicative of reversible resistive switching behavior between two states of clearly different resistance at room temperature. A

sharp transition into a low resistance state (LRS), i.e an abrupt increase of the current intensity, is detected above a well-defined threshold voltage indicative of localization of charge carriers. On the other hand, thicker samples exhibit a semimetallic character and I-V curves show progressive changes of the local resistance without a clearly defined threshold voltage, thus evidencing the absence of a MI transition with a well defined resistance jump between the different resistance states.

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Disorder and spatial localization due to thickness reduction allow generating a metal-insulator transition in semimetallic SrIrO_3 thin films. Below about 3 nm films are insulating with hysteretic I-V curves indicative of resistive switching behavior at room temperature. Current maps allow demonstrating the writing/erasing processes making evident the feasibility of the system for the implementation of Re-RAMs

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INTRODUCTION

In the search for a new generation of faster and more energy efficient electronic devices the use of reversible resistive switching (RS) phenomena has been proposed as a very appealing solution for the development of non-volatile memory devices.^{1,2} RS is the change of resistance in a reversible manner between two stable states of well-defined resistance by applying voltages pulses. This bistable behavior between high and low resistance states can be used as the core of a memory cell to implement a resistive random access memory (Re-RAM). RS behavior, typically observed in metal–insulator–metal (MIM) structures, has been found in a variety of materials, including transition metal oxides (TMOs), chalcogenides and even organic compounds, and diverse physical mechanisms, mostly based on thermochemical or electrochemical effects, have been invoked to account for the observed experimental behavior.^{3–7} In the case of TMOs one of the most common mechanisms to induce RS is based on modifications of the doping rate by valence change. The migration of oxygen vacancies, or metal cations, under electric field along defects, grain boundaries, dislocations, etc., induces a valence change of the cations nearby and, therefore, a change of the doping rate that may promote strong changes of the resistivity or even to trigger the appearance of a metal to insulator transition (MIT).^{2,3,8–10} This process of ionic migration can also occur at the interface between the metallic electrode and the active insulating oxide.⁸ Among TMOs exhibiting RS behavior Mott insulators and strongly correlated systems are of particular interest because they can exhibit different types of MIT transitions, i.e. huge changes of the electrical resistance, in response to a variety of external stimuli, such as temperature, doping rate or structural strain.¹¹ The close similarity between electronic correlations and crystal field splitting energies in TMOs provides different mechanism for tuning

their electronic properties and these mechanisms can even be widened by making use of spin-orbit coupling (SOC). In 3d TMOs SOC is not very relevant because being proportional to Z^4 , Z is the atomic number of the element under consideration, is much smaller than crystal field splitting and electronic correlations. However, in the case of 5d elements, as Ir, SOC reaches values of about 0.5 eV.^{12–14} Thus SOC becomes comparable to crystal field and electronic correlations, giving place to novel physics and exotic properties.^{15–17} In particular, the Ruddlesden-Popper series of 5d Strontium Iridates ($\text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1}$) have been investigated in the last years because of the strong interplay between these three contributions. This interplay may trigger different electronic states just by subtle structural or chemical changes. For example, the perovskite-like $n=\infty$ phase (SrIrO_3) presents a semimetallic ground state¹⁸ while on the other extreme of the series, in the $n=1$ phase (Sr_2IrO_4), a gap is opened at the Fermi level, leading the system into a Mott insulator state.¹⁹

As mentioned above, the MIT can be modified by applying different external perturbations, such as pressure²⁰ or electric field pulses as demonstrated for the $n=1$ phase²¹ and in the $n=2$ phase.²² In the case of the perovskite $n=\infty$ phase (SrIrO_3 , hereafter denoted as SIO) thin films, it was reported that a MIT can be triggered by tuning the compressive strain induced by the substrate or by reducing the film thickness.^{23–27} Different studies indicate that the MIT induced in these cases can be either of disorder driven Anderson type or unconventional Mott-Anderson type in which disorder effects and electronic correlations coexist.²⁴ However, recent results in high quality epitaxial thin films also show a concomitant divergence of the magnetic susceptibility in very thin films (4 unit cells) that clearly points to the opening of a Mott gap.^{28,29} Besides their fundamental relevance, the ability to modify at will the material resistivity by inducing a MIT is of strong interest from the application point of view. Even though the appearance of a MIT in

SIO has been reported by different groups^{23–27} and some results regarding RS effects in the $n=1$ phase,^{21,22} have been reported, reports regarding RS in the interesting semimetallic $n=\infty$ phase are very scarce.

In this work we present a systematic study of RS behavior in high quality SIO thin films prepared by RF sputtering on top of (001)-STO substrates, through local I-V curve measurements and current mapping, by using conductive atomic force microscopy (C-AFM). Epitaxial SIO films show semimetallic character and the appearance of a MIT is triggered by the reduction of film thickness. For film thickness below ~ 3 nm an Anderson-type MIT, i.e. induced by disorder, appears and resistance increases notoriously on lowering temperature. I-V characteristic curves in thin samples exhibiting the MIT show the typical hysteretic behavior associated to RS processes with two well-defined resistance states. I-V curves show directional RS depending on the polarity of the applied voltage, thus it can be classified as bipolar RS, with an abrupt increase of the current for voltages above a threshold value of around 2 V. Additionally, due to the particular features of the experimental setup, the observed RS is of interface type in which RS takes place at the interface between the metal electrode and the SIO film. Electric field pulses promote oxygen vacancies migration through the interface then, to maintain electrical neutrality, a valence change of the cations nearby takes place and therefore, a modification of the charge carriers' density and of the position of the Fermi level. A redox mechanism based on the band structure of SIO is proposed to account for the observed RS behavior. Current maps performed with voltage values above the threshold value and of different polarity allow demonstrating the writing/erasing processes making evident the feasibility of the system for the implementation of Re-RAMs. On the other hand, in the case of thicker films, initially in a semimetallic state, I-V curves present a smooth variation of resistance, without a

clear threshold voltage, suggesting the absence of two well defined resistance states separated by an energy jump. In this case, changes of resistance by applying voltage pulses are simple associated to the variation of charge carriers' density due to the migration of oxygen vacancies.

EXPERIMENTAL DETAILS

SIO films were grown by RF magnetron Sputtering on top of single crystalline (001)-SrTiO₃ (STO) substrates at 900 °C in oxygen atmosphere at 140mTorr. STO substrates were treated before deposition in order to obtain atomically flat surfaces of TiO₂ terraces. The treatment consisted in an ultrasound leaching in deionized water during 10 min followed by an annealing for 2 h at 1000 °C.³⁰ SIO samples with different thickness ranging between 2 and 26 nm were prepared. X-ray reflectometry (Siemens D-5000 diffractometer) was used to determine the thickness of SIO thick samples and to calibrate the growth rate, then the thickness of the thinnest films was determined by adjusting the deposition time.

X-Ray measurements were also used to check phase purity and epitaxial growth quality. For this purpose high resolution X-ray θ -2 θ scan of the (002) peak was performed by a using a PANalytical X'PERT PRO MRD system. Additionally, Reciprocal Space Maps (RSM) around the (-103) diffraction peak (pseudocubic notation) were also recorded by using a Bruker D8-Discover diffractometer.

The temperature dependence of the resistivity was measured by using a standard 4-probe configuration in a PPMS system from Quantum Design. Measurements were carried out in 400 μ m long x 100 μ m wide tracks patterned by U-V lithography and physical etching. Surface quality of the samples was studied by using Scanning Electron Microscopy (SEM) (QUANTA FEI 200 FEG-ESEM). An MFP3D system from Asylum was used for the acquisition of

topographic images, I-V curves and current maps. Atomic Force Microscope (AFM) topography images were recorded in tapping mode using Sb-doped Si probes (NCHV-A from Bruker). Surface roughness of the topographic images was calculated as a Root Mean Square (rms) of the height distribution of the sample surface. I-V curves and current maps were measured at room temperature by C-AFM, using doped diamond coated probes (DDESP-FM-V2 from Bruker), applying voltage to the sample while the tip was grounded. I-V curves were taken on random positions of the bare sample surfaces with a voltage sweep ranging between +5 and -5 V in ambient atmosphere. As it has been previously reported, the surface of iridates is very sensitive to air exposure and measurements of surface properties lose reliability few days after deposition of the film.^{27,31} This intrinsic surface instability is very challenging for C-AFM measurements, especially for performing measurements with long exposure times as current maps. To perform C-AFM maps a non-continuous layer of platinum nanoparticles was deposited on top of the SIO film. More details on these measurements may be found latter and in the SI.

RESULTS

High resolution Θ -2 Θ scan corresponding to a 21 nm thick SIO film deposited on (001)-STO substrate is shown in Figure 1(a). The epitaxial nature of the SIO film is clearly evidenced by the figure. The (002) reflection peak corresponding to the perovskite structure of the ($n=\infty$) phase is shown. Apart from the (00l) family, no additional peaks were found in the Θ -2 Θ scan making evident the high purity of the $n=\infty$ phase in the films. Moreover, satellite fringes can be clearly appreciated at both sides of the SIO Bragg peak. These oscillations, arising from the diffraction of the X-Ray beam at film finite planes with coherent thickness, are indicative of the high crystalline quality of the films and of the parallel orientation respect to the substrate. The peak position corresponds to an out-of-plane lattice parameter of 4.056 Å, i.e the SIO unit cell is

slightly elongated with respect to the reported bulk value ($a_0=3.943 \text{ \AA}$).³² Such value is in agreement with an elastic behavior governed by the in-plane compressive strain imposed by the smaller STO substrate ($a_{\text{STO}}=3.905 \text{ \AA}$) as usually reported in the literature.^{14,23–25} Figure 1(b) displays a reciprocal space map around (-103) reflection. It can be appreciated from the figure that both the substrate and the film peaks are aligned at the same in plane component ($Q_{\parallel}$), confirming the fully strained nature of the SIO film.

Figure 1. X-Ray Diffraction measurements of the SIO/STO thin films. (a) High - resolution Θ - 2Θ scan around (002) diffraction peaks. (b) Reciprocal space map of the (-103) peaks showing full strained state of the SIO films. For simplicity, all diffraction peaks are indexed in pseudocubic notation.

Surface morphology of SIO films, measured by AFM immediately after the deposition, corresponding to a 2nm thick and a 26 nm thick samples are depicted in Figure 2(a) and (b) respectively. Both samples show a flat topography with a surface roughness rms below 0.2 nm. In the case of extremely thin samples (rms typically below 0.12nm) the underlying terrace and steps structure of the STO substrate is replicated on the SIO surface.

Figure 2. AFM topography images of two SIO thin films on STO with thicknesses of (a) 2 nm and (b) 26 nm. Both films exhibit flat surfaces with low roughness (rms below 0.2 nm). In the case of very thin film, underlying terraced structure is still observed.

Electrical properties of SIO films are clearly thickness-dependent. Figure 3(a) depicts the temperature dependence of the sheet resistivity for a series of SIO films with thicknesses ranging between 2 nm and 26 nm. Metallic behavior is observed for film thickness down to ~ 3 nm in agreement with previous results.^{23–27,33} However, as thickness decreases a progressive upturn of the resistivity at low temperatures is detected. The temperature at which the minimum in the resistivity is found shifts up in temperature as thickness decreases and the system exhibits fully insulating behavior for the 2 nm thick sample, thus locating the MIT transition between 3 and 2 nm, in good agreement with previous reports.²⁴ Discrepancies regarding the actual thickness value at which the MIT takes place are usually attributed to differences in the microstructural quality of the samples.³⁴ For sample thickness above 10 nm the high temperature behavior of the resistance is well described by the expression $\rho(T) = \rho_0 + CT^{4/5}$ (see Figure 3(b)). The value obtained for the residual resistivity, ρ_0 , is in all the samples below the so-called Mott-Ioffe-Regel (MIR) limit³⁵ ($\hbar/e^2 \approx 26$ k Ω). In the low temperature regime, the resistivity shows a T^2 dependence down to the lowest temperature measured (~ 10 K) indicative of a Fermi liquid behavior (see inset of Figure 3(b)). This crossover from a non-Fermi liquid at high T to a Fermi

liquid behavior at low T is typically associated to strongly correlated systems,³⁶ thus we should conclude that thick SIO samples behave as a three dimensional correlated metal.

As the critical thickness corresponding to the MIT transition is approached the low temperature upturn of the resistivity shifts up to higher temperature and become more abrupt, as typically observed when the MIT transition is approached from the metallic side. In the case of the 3 nm thick sample a notable upturn of the resistivity is observed below about 60 K and the low temperature regime is well described by a characteristic $\text{Ln}T$ dependence of the sheet conductance, indicative of two dimensional weak localization (see inset of Figure 3(c)).³⁷ Once the film thickness is reduced below 3 nm the transport properties change abruptly. The resistance of the 2 nm thick film presents a typical insulating dependence with $d\rho/dT < 0$ in the whole temperature range. Clues of this transition can also be observed in the inset of Figure 3(a), where the resistivity at 300 K is almost constant for the films with thickness between 3 nm and 26 nm, but sharply increases for the 2 nm thick sample.

Figure 3. Electric properties of the SIO/STO films. (a) Temperature dependence of the sheet resistance in SIO films with thickness of 2 nm (orange), 3 nm (brown), 5 nm (green), 15 nm (blue) and 26 nm (black). Inset. Resistivity of films at 300K. A change of behavior from a semimetallic state to an insulating state when reducing thickness below 3nm is clearly observed. (b) Temperature dependence of the resistivity of 15 nm thick SIO film vs. $T^{4/5}$. Inset: Detail of the low temperature regime to make evident the crossover to a Fermi liquid behavior at low T . (c)

Temperature dependence of the resistivity of the 2 nm thick sample. The fittings using the Mott's variable range hopping (VRH) model, $\text{Ln}\sigma \propto 1/T^{1/(d+1)}$ (red line), at low T and the Arrhenius equation for thermal activation conduction, $\rho(T) = \rho_0 e^{(\Delta/k_B T)}$ at high T (blue line), are indicated. Inset: detail of the low temperature regime of the sheet conductivity of the 3 nm thick sample showing the $\text{Log } T$ dependence indicative of two dimensional weak localization.

To clarify the nature of the MIT induced by reducing the film thickness we have carefully analyzed the temperature dependence of the resistivity of the fully insulating 2 nm thick sample.³⁸ The low temperature resistance upturn is well described by using the Mott's variable range hopping (VRH) model: $\text{Ln}\sigma \propto 1/T^{1/(d+1)}$, being σ the conductivity and d the dimension.³⁹ As can be appreciated in Figure 3(c) the VRH model with $d=2$ fits experimental data almost perfectly up to about 40-45 K, while the high temperature range is properly described by using the Arrhenius equation for thermal activation conduction $\rho(T) = \rho_0 e^{(\Delta/k_B T)}$ being Δ an activation energy (a value of $\Delta \sim 6$ meV was obtained from the fit) and k_B the Boltzmann's constant (see Figure 3(c)). Therefore, from the analysis of the $R(T)$ curves we can conclude that the MIT induced in SIO films by reducing the thickness is of Anderson localization type due to disorder; in good agreement with previous reports.^{24,26}

The possibility of inducing an Anderson type MIT by reducing film thickness suggests that resistivity of SIO thin films may be sensitive to different stimuli, especially for thickness close to the critical one, therefore may be a good alternative for obtaining a strong RS response and thus, of interest for the implementation of RS devices. For this purpose, we have boarded the study of local I-V curves measured at room temperature by C-AFM of SIO thin films. To illustrate our findings, we present in Figure 4 a detail (below 100 nA) of the IV curves on samples at both extremes of the thickness series (i.e. 2 nm and 26 nm). Results at intermediate thickness (5 nm)

as well as the full IV curves may be found at the SI. In all cases hysteretic I-V curves are observed indicative of RS behavior. An anti-clockwise behavior, where positive sample voltage induces the Low Resistance State (LRS) and negative sample voltage induces the High Resistance State (HRS) is found. I-V curves were measured by applying a voltage sweep between +5 V and -5 V with no current limitation. Figure 4(a) displays the I-V curves corresponding to the 2 nm thick film. As it can be seen, the current intensity remains constant and near to zero until the threshold voltage value of about 2 V is reached at which a sudden increase of the current takes place (indicated by arrow 1 in the figure). After reaching the +5 V, the sample remains in the LRS returning to 0 V through the superior limit of the graph along the path denoted by arrow 2. In the negative voltage range, the I-V curve starts at the LRS and the current is continuously increasing for voltages up to -5 V (trajectory marked by arrow 3). Once reached this voltage, the I-V curve returns to the HRS along the path indicated by arrow 4. Figure 4(b) depicts I-V curves corresponding to the 26 nm thick sample. In this latter case resistance changes in a continuous way and no well-defined resistance states separated by a clear voltage threshold are observed. The similarity of the measured I-V curves in both samples suggests that the mechanism behind the observed variations of resistance is the same in both cases. However, the absence of a clear voltage threshold in the case of the thick sample suggests the absence of a well defined energy transition separating the LRS and HRS.

Figure 4. C-AFM I-V curves detail in fresh SIO samples. (a) 2 nm thick film. (b) 26 nm thick film. The different colors of the curves represent the different consecutive voltage cycles between +5 and -5 V. The different arrows indicate the direction of the curves and their current resistance state (HRS black and LRS red).

A deeper insight into the RS process in SIO samples may be obtained by performing current maps over extended areas in both HRS and LRS states. Nevertheless, the significant statistical gain has the drawback that measurements are more aggressive and time consuming than the local I-V curves. Performing a C-AFM map implies to move the AFM tip back and forth several times

in the same area of the sample. The continuous scanning deteriorates the electrical tip-sample contact after a few cycles. This well-known drawback in c-AFM analysis of RS phenomena is usually attributed to tip contamination⁴⁰ Experimentally we have observed that capping of the SIO film surface with a non-continuous layer of Pt nanoparticles (NPs) is useful to perform the c-AFM maps. This beneficial effect of the Pt NPs avoids degradation of the tip-sample electrical contact either by reducing the degradation of film surface either by enhancing tip life time since brushing with Pt nanoparticles reduces tip-contamination. Thus, current maps were performed in 26 nm films with their surface partially covered with platinum NPs (see further details in SI). Figure 5(a) presents a $50 \times 50 \mu\text{m}^2$ current map measured at low voltage (0.2 V) and 570 nN of normal load after the scanning of the central $35 \times 35 \mu\text{m}^2$ area at the negative bias voltage (-5 V) applied to the sample. A clear reduction of the conductivity can be observed in a central square (dark area) that corresponds to the established HRS. In contrast, the surrounded brighter area shows a higher current and corresponds to the pristine state. Figure 5(b) shows the topography image of the same zone measured simultaneously, no changes in the topography of the area switched into HRS are appreciated. Figure 5(c) shows a current map of the same zone measured after the scanning of the central $20 \times 20 \mu\text{m}^2$ area at +3 V applied to the sample. Now three different states can be observed: the pristine state at the outer parts of the current map, the HRS represented as a darker square ring, and finally, the LRS depicted by a central bright square. Similar to the I-V curves in Figure 4, HRS was induced by negative sample voltage (-5 V) and it can be reversed to LRS by scanning at positive sample voltage (+3 V). The associated topography measurement is also shown in Figure 5(d). Some minor topography changes are detected in the central area switched into LRS. They are represented by a slightly darker contrast indicating a local decrease of the film height, which, in principle, could be caused by the removal

of the Pt NPs or by a local compression of the film. Nevertheless, since accumulations of NPs were not observed along outer rims of the scanning area, and the current intensity in this central zone even increases, NPs removal should be discarded as a possible explanation of the observed changes of the film's topography. On the other hand, due to the difficulties to reset the zone into LRS, a higher normal load (1140 nN) was applied during the scanning performed at +3 V. This increment in the normal load of the AFM tip may well be responsible for the observed local film compression.

Even with the increase of the normal load in the positive voltage scan, the switching into LRS was more difficult to achieve than the switching into HRS. One possible reason for this behavior is the occurrence of undesired reactions at the sample surface for high enough positive voltages due to local anodic oxidation,^[38] decreasing the stability of the current measurements. To avoid these reactions, the sample voltage used to reset the surface into the LRS was decreased to a value of +3 V (instead of +5V). This reduction in the reset voltage is probably the main reason for the less pronounced and stable switching into LRS displayed in Figure 5(c).

Figure 5. C-AFM current maps ((a) and (c)) and topography images ((b) and (d)) of a 26 nm SIO/STO sample with Pt NPs on the top obtained simultaneously. (a) Current map showing a $35 \times 35 \mu\text{m}^2$ zone set into the HRS. (b) Topography of the same zone showing no changes in the surface topography. (c) Current map of the same zone after resetting a $20 \times 20 \mu\text{m}^2$ zone to the LRS. (d) Topography image of the same zone, where minor changes in the topography can be noticed in the switched area.

DISCUSSION

There is a large diversity of mechanisms proposed to explain RS phenomena^{3–7} In the particular case of TMOs, redox based mechanisms, driven by the motion of cations (coming from the electrodes) or defects such as oxygen vacancies, have attracted much attention.^{2,3,9} Two different categories can be distinguished: filamentary RS, consisting in the creation of filament of cations or vacancies in a TMO film sandwiched between two electrodes¹ and interface RS, in which this process of ionic migration occurs at the interface between the metallic electrode and the active

insulating oxide.⁸ Due to the in-plane geometry used in this study for the I-V measurements (the current path goes from AFM tip, through the sample surface, and finally to a metal contact placed on the sample surface) only interface RS should be considered.

In correlated metals such as SIO, the variation on the concentration of oxygen vacancies can modify the valence of the surrounding metal cations. Since the oxygen vacancies substitute an oxygen position, the surrounding cations must adapt their valence to compensate the charge variation. Therefore, new electrons would fill Ir orbitals changing the Fermi level and electric properties.¹¹ Depending on the type of majority charge carriers, this valence change will result in an increase (decrease) of the resistivity in the case of holes (electrons).² It is worth noting that the nature of the majority charge carriers in SIO thin films is not clearly established yet. As a semimetal, SIO is expected to have a conduction mechanism that involves both electrons and holes.^{19,27} Nevertheless, small differences in the charge carrier density could be critical for the RS mechanism. Recent results based on magnetotransport and thermoelectric measurements suggest that electrons and holes, have similar densities and mobilities.²⁷ However, given the small unbalance between them²⁷ and the degree of disorder present in our films (considering the appearance of Anderson localization) it seems reasonable to consider that both, electrons/holes, could be the majority charge carriers in SIO films. Therefore, two similar mechanisms are proposed to explain the observed RS behavior in SIO films.

If electrons are considered the majority charge carriers, the change in the oxygen vacancies' concentration would be driven by their internal migration within a film induced by an externally applied electric field.⁴¹ When a positive voltage is applied to the sample (negative voltage to AFM tip), oxygen vacancies would migrate towards the AFM tip thus increasing the Fermi level. The subsequent rise of the electron concentration at the tip-sample interface would decrease the

contact resistance and therefore transforming the sample surface into the LRS. On the other hand, negative sample voltage (positive voltage at the AFM tip) would act reversely, expelling the vacancies from the tip-sample interface and thus decreasing the electron concentration and conductivity of the sample surface and leading it to HRS. Alternatively, if holes are considered the majority charge carriers, oxygen vacancies could be incorporated or removed from the interface by a Redox reaction at the film surface triggered by an externally applied electric field.⁴² Contrary to the previous case, vacancies decrease the conductivity in hole-like systems since they are considered as acceptors scavengers. Positive voltage applied to sample would cause the reduction of the atmosphere's oxygen and the incorporation of oxygen anions into the oxide matrix, removing the vacancies from the interface according to the following formula (equation 1).

Then, the Fermi level would decrease, while the hole density and the conductivity would increase and transform the sample surface beneath the AFM tip into the LRS. The system can be reversed into the HRS by applying negative voltage to the sample. In this case, the following oxidation reaction takes place (equation 2).

With this reaction oxygen anions would be removed from the matrix leaving vacancies behind. These vacancies would reduce drastically the number of holes and raise up the Fermi level, reducing this way the conductivity. Although both films seem to follow the same mechanism for

the changes of resistance, the two set of I-V curves present notable differences. Figure 4(b), corresponding to the 26 nm thick sample, shows smoothly rising I-V curves for the positive voltage range from 0 V to the saturation of the current detector. On the other hand, Figure 4(a), corresponding to the 2 nm thick film, which has undergone MIT, shows an almost zero current up to a clear voltage threshold value of about 2V, when an abrupt increase of the current intensity occurs. This different behavior is closely related to the differences in the band structures of the films that are induced by the MIT. On one hand, it is reported that thick SIO films (such as the employed 26 nm thick one) behave as a semimetal with electrons and holes having similar densities.^{18,19,27} On the other hand, even though the band structure of very thin films (such as the 2 nm thick one) has not been measured so far, thickness reduction may trigger the appearance of an Anderson-type MIT transition with strong carriers' localization.^{24,26} Even more, the aperture of a Mott gap in ultrathin SIO films have been recently documented.²⁸ With this picture in mind, the behavior of thick SIO film (26 nm) reflects changes of resistance in a system with continuous energy levels having thus a smooth increase of the current. Moreover, the shift of the Fermi level would bring the system into a state with higher Density of States, finally giving higher conductivity and setting it into the LRS. On the other hand, in the case of the very thin film (2 nm), as long as Fermi energy lies inside localized states, charge carrier's mobility is seriously hampered or even suppressed and the current would be almost zero. Nevertheless, when the Fermi level crosses the mobility edge an energy transition between localized and extended states takes place for high enough positive voltage, an abrupt increase of the current would be observed and obviously the system would be switched in the LRS with a higher conductivity.

CONCLUSIONS

The local transport properties of SrIrO_3 thin films prepared by RF sputtering on top of (001)- SrTiO_3 substrates have been analyzed as a function of the film's thickness. Macroscopic measurements of the film's resistivity as a function of temperature allow demonstrating that an Anderson type MIT transition can be induced, triggered by thickness reduction, below ~ 3 nm. On the other hand, SIO films exhibit hysteretic I-V curves indicative of RS behavior irrespective to their thickness. I-V curves show directional RS depending on the polarity of the applied voltage, thus it can be classified as bipolar RS. Additionally, due to the particular experimental setup used, it is concluded that the observed RS is of interface type in which RS takes place at the interface between the metal electrode (AFM tip) and the SIO film. Electric field pulses promote oxygen vacancies migration through the interface then, to maintain electrical neutrality, a valence change of the cations nearby takes place and therefore, a modification of the charge carriers' density and of the position of the Fermi level. However, the features of the RS process are clearly different in the case of very thin samples (below 3 nm), where the appearance of the MIT, with the concomitant carriers' localization, is reflected in the existence of a well-defined voltage threshold value separating the low and high resistance states. In contrast, in thicker films with semimetallic behavior I-V curves present a continuous smooth variation of resistance, without a clear threshold voltage, making evident the absence of an energy sharp transition separating high and low resistance states. In this case, changes of resistance by applying voltage pulses are simply associated to the variation of charge carriers' density due to the migration of oxygen vacancies. In spite of the different behaviors, the same mechanisms involving oxygen vacancies are claimed to explain the observed results irrespective of the nature of the majority charge carriers, electrons or holes. Current maps performed with voltage values above the

threshold value and of different polarity allow demonstrating the writing/erasing processes making evident the feasibility of the system for the implementation of Re-RAMs.

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ABBREVIATIONS

AFM: Atomic Force Microscopy, C-AFM: Conductive-Atomic Force Microscopy, MIT: Metal Insulator Transition, RS: Resistive Switching, LRS: Low Resistance State, HRS: High Resistance State, Re-RAM: Resistive Random Access Memory, SIO: SrIrO_3 , TMO: Transition Metal Oxide, SOC: Spin-Orbit Coupling, RSM: Reciprocal Space Map, rms: root mean square, I-V: Intensity-Voltage, MIR: Mott-Ioffe-Regel, MIM: Metal-Insulator-Metal

SUPPLEMENTARY INFORMATION

(1) Current maps showing stable cycling between HRS and LRS states performed by C-AFM on a 26 nm thick SIO film.

(2) Non-conformal capping of the SIO surface with Pt nanoparticles to improve C-AFM measurements

- (3) Complete I-V curves of SIO thin films (2 nm and 26 nm).
- (4) Detail of the I-V curve showing resistive switching for SIO film at intermediate thickness (5 nm).

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