Transactions of the Materials Research Society of Japan 25 [1] 201-204 (2000)

Novel Current Transport Phenomena in Epitaxial Ferroelectric Heterostructures

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Ferroelectric heterostructures were grown by depositing ferroelectric films epitaxially on lattices matched perovskite oxides. Current transport through the heterostructures is investigated and novel characteristics are found. The current flowing at the ferroelectric surface in a FET structure, is controlled by ferroelectric polarization. Current flowing through the heterostructures is programmed by external voltage pulses. This is regarded as a quasi-three terminal operation of two terminal diode. An anomalous temperature dependence of current density was found, which may be regarded as a low temperature positive temperature coefficient of resistance (PTCR) effect. Current conduction at nm-scale contacts are also reported.
Keywords : ferroelectric, heterostructure, memory, field effect

1. INTORODUCTION

The application of the perovskite transition metal oxides for electronic devices are extensively pursued. Perovskite transition metal oxides possess a wide variety of electronic properties such as superconductivity, magnetism, magnetoresistance, and ferroelectricity. Moreover, despite a wide variety of electronic properties, these transition metals have similar crystallographic properties and lattice constants that facilitate an epitaxial heterostructures of these materials and an integration of the functionality. It is important to explore the new possibilities, although it may not be readily commercialized in a Si-based electronic market on except for the applications in space.

We have been explored the possibility of new devices based on unique properties of perovskite oxides and an integrated functionality, utilizing the ferroelectricity, the metal-insulator transition, and the semiconductivity. We may list the followings as merits of perovskite semiconductors: 1) epitaxy with ferroelectric and magnetic perovskites, 2) epitaxy with insulating and metallic oxides, 3) appropriate band gap, 4) tunability of band, 5) field effect using metal-insulator transitions, 6) epitaxial 3-dimensional-IC, 7) resistance to radioactive irradiation in space use.

In the following, we report a few new phenomena and devices proposed and found by us for the first time: (1) a ferroelectric FET (field effect transistor) using a metal-insulator transition of the parent phase of the high-*Tc* superconductor, (2) non-volatile pulse modulation of current in ferroelectric/perovskite semiconductor diodes, (3) an anomalous temperature dependence of current conduction in ferroelectric/perovskite semiconductor diodes at low temperature (*T*). The current transport at nm-scale contacts (4) is also presented.

2. EXPERIMENTAL

The *c*-axis oriented $Pb_{1-y}La_yZr_xTi_{1-x}O_3$ (PLZT), $Pb_{1-y}La_yTiO_3$ (PLT) and $PbZr_xTi_{1-x}O_3$ (PZT) films were epitaxially grown by the pulse laser deposition on (100) $SrTiO_3$ doped with 0.5 weight % Nb (STON) as well as on 100-nm-thick $La_{1.99}Sr_{0.01}CuO_4$ (LCO).[1,2] The ferroelectric layer is 200-300 nm thick. All the measurements were done at room temperature (RT), if not other wise stated.

3. ALL-OXIDE FERROLECTRIC FET

The spontaneous polarization (*Ps*) can induce a nonvolatile field effect modulation of conductance. Since the proposal of the ferroelectric-field-effect variable resistor or transistor in 1957, its performance has been reported by many research groups. Despite recent intensive works, *a long time retention of a transconductance modulation by a short pulse voltage* has not been demonstrated before our reports.[3-5] The difficulty originates from the degradation of properties of the ceramic ferroelectric grown *directly* on Si or GaAs. So far, even an epitaxial growth of ferroelectric directly on Si or GaAs is extremely difficult. The semiconducting perovskite oxides allow a direct epitaxy of single-crystal like ferroelectric on it. This suppresses the instability due to the depolarization field, reduces defects and traps at the interface, and has a possibility of suppressing the "half select disturb pulse threshold". We proposed and demonstrated this approach for the first time, which was followed by similar approaches[6] that yielded a lower modulation or a lower speed and a shorter retention time than ours. The following results are for a prototype FET consisting simply of a PLZT gate and a LCO channel without an enhanced-doping source/drain. The gate of the devices was effectively 40 μm and 200μm long and was defined by an ion milling etching, The FET was written/erased

The relaxation current at very low bias is also programmable (Fig.6). An advantage of this approach over FET is the reduction of the wiring for a high density integration. Furthermore, using the present diodes, synapse elements in neural circuits and disk memories can be developed.[12]

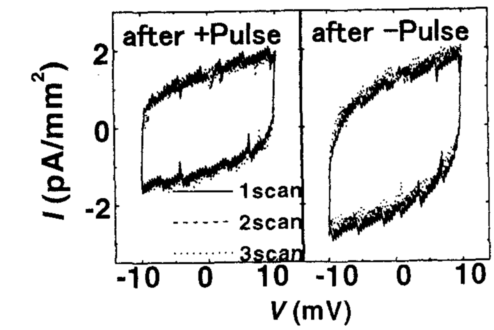

Fig.6 Low voltage IV characteristics of PLZT/STON after ±6 V 25µs pulses. Three IV curves taken consecutively are displayed and are almost identical.[11]

5. LOW-T PTCR

An anomalous T-dependence of resistance of ferroelectric, known as a PTCR effect, has been studied and is now utilized in commercial application. Tunneling current shows also an anomalous T-dependence. It was first discovered in a narrow Ge pn junction 40 years ago and has been reported in various structures. However, no tunneling current through an oxide pn junction has not been reported so far.

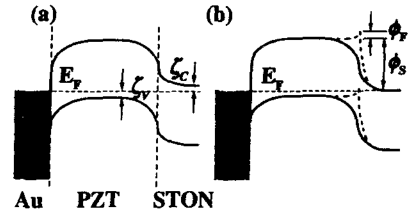

Fig.8. Band diagrams of Au/PZT/STON at RT (a) and at low-T (b).[13]

An anomalous increase of the current density with decreasing T was found in Au/PZT/STON.[13] In the T-dependence of IV curves of Au/PZT/ STON in Fig. 7(a), a diode-like current is evident at positive bias above 270 K and decreases with reducing T. On the other hand, the current grows at negative bias with reducing T. A pair of IV curves near 300 K before and after the IV measurements at low T are almost identical, indicating no irreversible change during the measurement.

On the contrary, no anomalous behavior was observed in Au/PLZT/LCO. In Fig.7(b) diode-like current was observed at negative bias at 290 K and decreased with reducing T. Diode-like current was suppressed at 85K for the bias scan amplitude of ±3 V.

One of the possible causes of the increasing conductance with decreasing T may be the T-dependence of the *avalanche ionization*. Although this model is appealing, it can not explain the shape of the IV curves.

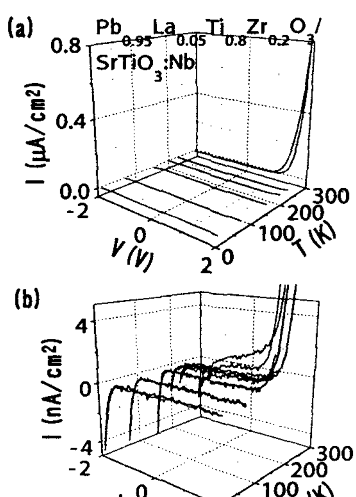

Fig.7 T dependence of IV hystereses of PZT/STON (a) and PZT/LCO (b). The arrows indicates the direction of scan.[13]

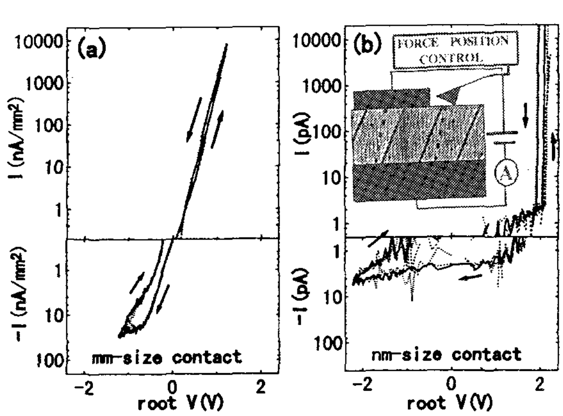

Fig.9. Experimental set-up for IV measurements using an AFM with a Pt-coated tip and metal electrodes on BTO. surface IV curves of Pt (area: 0.4mm^2)/BTO/ STON and Pt coated tip/BTO/STON.[15]

at ±7V with a pulse width of 10ms, yielding the resistance modulation of 10%(Fig.1). The write/erase speed was determined by the RC time constant, and, therefore, by the device size.

In the present device size, the minimum speed was 10 μs. The memory was fully retained for one day, and the 50% of modulation was retained over 10 months at RT.

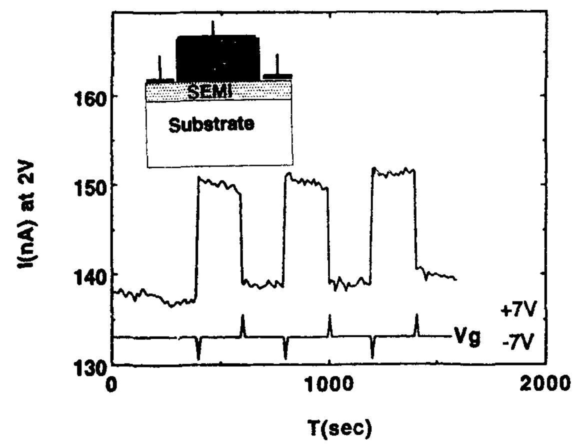

Fig.1 Pulse switching of transconductance in ferroelectric FET.[2]

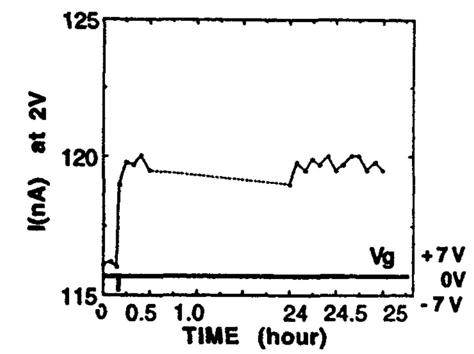

Fig.2 24 hour retention characteristics.[2]

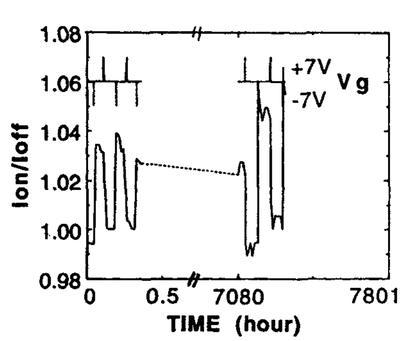

Fig.3 10 month retention characteristics of an on-state kept in ordinary atmosphere without packaging.[3]

4. PROGRAMMABLE FERROELECTRIC DIODE

The *pn* junction (*p*: hole carrier, *n*: electron carrier) formation in ferroelectric heterostructures opens a possibility of developing a new class of semiconductor heterostructures and also ferroelectrically tunable *pn* junctions.[7-11] Ferroelectric perovskites change *Ps* by the application of a short voltage pulse, while they conduct the carrier when a *dc* voltage is applied. Namely, ferroelectric behaves as an insulator in a short time scale and as a semiconductor in a long time. These properties allow a time domain control of a three-terminal device like operation of diodes (Fig.4). The conductance modulation programmed by a short voltage pulse is perfectly retained for 10 days at RT and is nondestructively read (Fig.5) for the first time.[11]

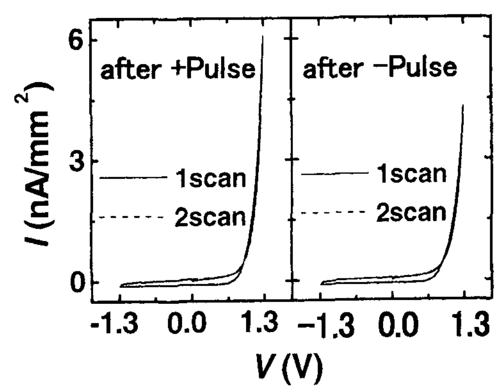

Fig.4 The current-voltage (*IV*) characteristics of PLZT/ STON (*pn* junction like).[11] The +6 V 25μs pulse increased the forward bias current, and the −6 V 25 μs pulse decreased it. For each pulse, two *IV* curves taken consecutively are displayed. Two *IV* curves measured after +6 V (left) and −6 V pulse (right) are shown but are completely overlapped, indicating that the reading process is nondestructive.

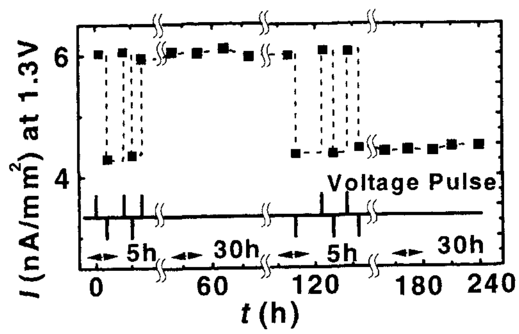

Fig.5 Long time retention of *IV* characteristics modulation in PLZT/STON (*pn* junction like) by ±6 V 25μs pulses.[11] The current density *I* at V_{dc} = +1.3V is plotted. [*I* is taken from the 3rd *IV* on each day to eliminate the effect of trap emissions.] Abscissa has a dual scale: The arrows of the length of one division show the scales.

Alternatively, the band diagram[14] of PZT/STON at RT at zero bias in Fig. 8(a) suggests a possibility of the tunneling current at reverse bias. Even for $\varepsilon= 5$ of STON and PZT, the depletion layer width is approximately 30 nm and very wide as compared with those of ordinary tunnel junctions. However, the tunneling seems the most plausible conduction mechanism. First, a replot of Fig.7(a) confirms that the Fowler-Nordheim-type tunneling fits the results satisfactorily for $V < 0$. Second, the T-dependence of the current density agrees with the tunneling model.

6. *IV* at nm-scale contacts

The current transport through on mm- and nm-size contact on epitaxial $BaTiO_3$ (BTO) films was investigated by using atomic force microscope (AFM) with a conducting tip. The breakdown voltage and the current density at both nm-size and mm-size contact were controlled by the Schottky barrier. However, in marked contrast with the mm-size contacts, the nm-size contacts conducted little current below breakdown voltage and repeatedly exhibited abrupt breakdowns having a giant current density > 10 Amm^{-2}. The breakdown field was as high as 0.45 MVcm^{-1} at the forward bias, while no breakdown occurred up to 0.5 MVcm^{-1} at the reverse bias.

7. CONCLUSION

New current transport phenomena that may be potentially useful for application is presented.

Acknowledgement

This work is support by the grants in aid No.09450156 from the Ministry of Education, Science, Sports and the Iketani Science Foundation. The author acknowledges also the contributions of Mr. M. Okano, Mr. D. Sawamura, and Mr. T. Shigesue to a part of works presented in the paper.

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(Received December 16,1999 ; Accepted January 31,2000)