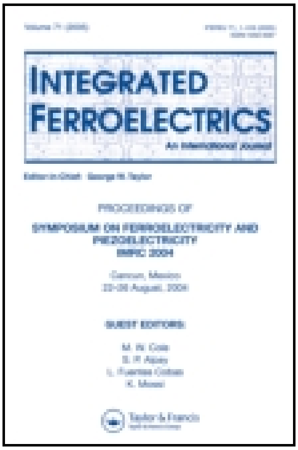

Integrated Ferroelectrics: An International Journal
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/ginf20

Current-Temperature Characteristics of Low-Temperature-Sputtered (Ba,Sr)TiO 3 Films Post Treated by Rapid Thermal Annealing

Der-Chi Shye [a] , Jyh-Shin Chen [b] , Meng-Wei Kuo [a] , Bruce C. S. Chou [b] , Chueh-Kuei Jan [a] , Mei-Fang Wu [b] , Bi-Shiou Chiou [a] & Huang-Chung Cheng [a]

[a] Department of Electronics Engineering and Institute of Electronics , National Chiao Tung University , Hsinchu, 1001 Ta Hsueh Rd., Hsinchu, 30010, Taiwan R.O.C.

[b] Precision Instrument Development Center , National Science Council , 20 R&D Road VI, Hsingchu Science-Based Industrial Park, Hsinchu, 30010, Taiwan R.O.C.

To cite this article: Der-Chi Shye , Jyh-Shin Chen , Meng-Wei Kuo , Bruce C. S. Chou , Chueh-Kuei Jan , Mei-Fang Wu , Bi-Shiou Chiou & Huang-Chung Cheng (2002) Current-Temperature Characteristics of Low-Temperature-Sputtered (Ba,Sr)TiO 3 Films Post Treated by Rapid Thermal Annealing, Integrated Ferroelectrics: An International Journal, 47:1, 217-225

To link to this article: http://dx.doi.org/10.1080/713718282

Taylor & Francis makes every effort to ensure the accuracy of all the information (the "Content") contained in the publications on our platform. However, Taylor & Francis, our agents, and our licensors make no representations or warranties whatsoever as to the accuracy, completeness, or suitability for any purpose of the Content. Any opinions and views expressed in this publication are the opinions and views of the authors, and are not the views of or endorsed by Taylor & Francis. The accuracy of the Content should not be relied upon and should be independently verified with primary sources of information. Taylor and Francis shall not be liable for any losses, actions, claims, proceedings, demands, costs, expenses, damages, and other liabilities whatsoever or howsoever caused arising directly or indirectly in connection with, in relation to or arising out of the use of the Content.

This article may be used for research, teaching, and private study purposes. Any substantial or systematic reproduction, redistribution, reselling, loan, sub-licensing, systematic supply, or distribution in any form to anyone is expressly forbidden. Terms & Conditions of access and use can be found at http://www.tandfonline.com/page/terms-and-conditions

Downloaded by [National Chiao Tung University] at 23:55 30 April 2014

Integrated Ferroelectrics, 2002, Vol. 47, pp. 217–225
Reprints available directly from the publisher
Photocopying permitted by license only

DOI: 10.1080/10584580190044687

Current-temperature Characteristics of Low-temperature-sputtered (Ba,Sr)TiO$_3$ Films Post Treated by Rapid Thermal Annealing

DER-CHI SHYE, JYH-SHIN CHEN*, MENG-WEI KUO,
BRUCE C. S. CHOU*, CHUEH-KUEI JAN, MEI-FANG WU*,
BI-SHIOU CHIOU AND HUANG-CHUNG CHENG
Department of Electronics Engineering and Institute of Electronics,
National Chiao Tung University, Hsinchu, 1001 Ta Hsueh Rd.,
Hsinchu, 30010,Taiwan R. O. C.
**Precision Instrument Development Center, National Science*
Council, 20 R&D Road VI, Hsingchu Science-Based Industrial Park,
Hsinchu 30010, Taiwan R. O. C.

(Received in final form October 15, 2002)

ABSTRACT

This work reports the current-temperature characteristics of the low-temperature-sputtered (Ba$_{0.8}$Sr$_{0.2}$)TiO$_3$ (BST) film post treated by rapid thermal annealing (RTA) in O$_2$ ambient. The top electrode was biased under negative/positive voltage to investigate the interface properties of the Pt/BST/Pt multifilm. As the results, the current density of the RTA-treated BST film was greatly reduced owing to compensation of oxygen vacancies. The RTA-treated BST thin film biased at negative voltage exhibits a negative temperature-coefficient-resistivity (NTCR) behavior, but, intriguingly, that biased at positive voltage reveals a positive temperature-coefficient-resistivity (PTCR) behavior. According to the leakage current analysis, the Schottky emission dominates the negative biased current at upper interface, but the Heywang barrier scattering confines the positive biased current.

Keywords: (Ba$_{0.8}$Sr$_{0.2}$)TiO$_3$, rapid thermal annealing (RTA), NTCR, PTCR, Schottky emission, Heywang barrier

INTRODUCTION

Recently, the high temperature-coefficient-resistivity (TCR) thermistors are widely applied as thermal detectors owing to their low cost and excellent responsivity [1]. In general, the thermistor using positive TCR (PTCR) material can prevent the effect of thermal-run-away, so the complementary circuits can be simplified [2]. Since the abnormally high PTCR of $BaTiO_3$ (BTO) ferroelectric materials was observed at Curie temperature (Tc) in 1955 [3], the conduction mechanisms and applications of these perovskite films have attracted great attention in the past decades. The substitution of Sr for Ba in BTO solid solution, forming the $(Ba,Sr)TiO_3$ (BST), can effectively adjust the Currie temperature to obtain a proper operation condition for a device.

The high TCR thin film is employed on microthermistor array using the fabrication technique of microelectromechanical-system (MEMs) as an infra-red (IR) image sensor. Low temperature process is indeed necessary for the integration of the high TCR film into the MEM system, because a high temperature process will damage the under-layer structures of MEMs and junction profile of ICs. In this study, the low-temperature-prepared BST films were post treated by rapid thermal annealing (RTA) to achieve excellent electric properties. However, the TCR phenomenon of the bulk BST is discussed in many works, but very few works have reported the TCR effect of the thin BST film [4]. The conduction current of BST was measured in the temperature ranging from room temperature to 410°K, and the conduction mechanisms were systematically investigated.

EXPERIMENTAL

A testing structure of Pt/BST/Pt/TiN/Ti multifilm resistor with intercrossing top/bottom electrodes was employed for TCR measurement, as shown in Figure 1. In general, an extra thermal expansion stress is caused by directly probing on the heated-BST film, so the intercrossing electrodes prevents the unwanted disturbance from TCR measurement. The testing area ($20\times20\ \mu m^2$) of this thin multifilm is defined by the overlap between the top and bottom

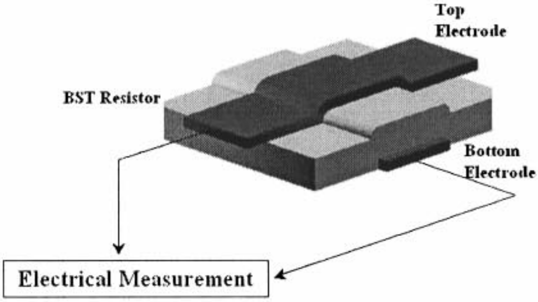

Figure 1. The testing structure of Pt/BST/Pt/TiN/Ti multifilm with intercrossing top/bottom electrodes.

electrodes, and both of top and bottom electrodes are patterned by using the lift-off technique.

The Pt/TiN/Ti multifilm with 100nm/50nm/10nm, serving as the metal/barrier/adhesion layer, was sputtered onto the SiO_2/Si substrate at room temperature. $Ba_{0.8}Sr_{0.2}TiO_3$ film (300nm) was then deposited using a RF magnetron co-sputtering system at a substrate temperature of 300°C. The BST films were subsequently post treated by RTA 400°C (5 minutes) and 500°C (1 minute).

The current-temperature characteristic was studied by using the Pt/BST/Pt

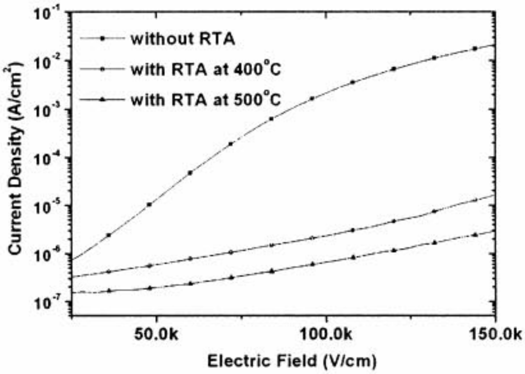

Figure 2. The conduction current of Pt/BST/Pt with various RTA temperatures measured at 320°K.

(metal-insulator-metal, MIM) structure. An automatic measurement system that combines HP 4156A and a probe station was conducted to obtain the current-voltage (I-V) characteristics of BST resistors. The current-temperature testing was carried out using a hot plate.

RESULTS AND DISCUSSION

Figure 2 presents the leakage current of BST films post treated at various RTA temperatures. In general, the conduction current of BST film is induced by oxygen vacancies according to $O_O \leftrightarrow V_O^{++} + 2e^- + 1/2O_2$, where O_O, V_O^{++} and e^- denote the oxygen ion at its normal site, oxygen vacancy and electron, respectively. However, a higher concentration of oxygen vacancies leads to n-type conductivity of the BST materials due to the generated electrons, causing larger conduction currents [5]. The conduction current of BST film with post RTA treatment in oxygen ambient is suppressed due to oxygen vacancies compensated by O_2 ambient gas.

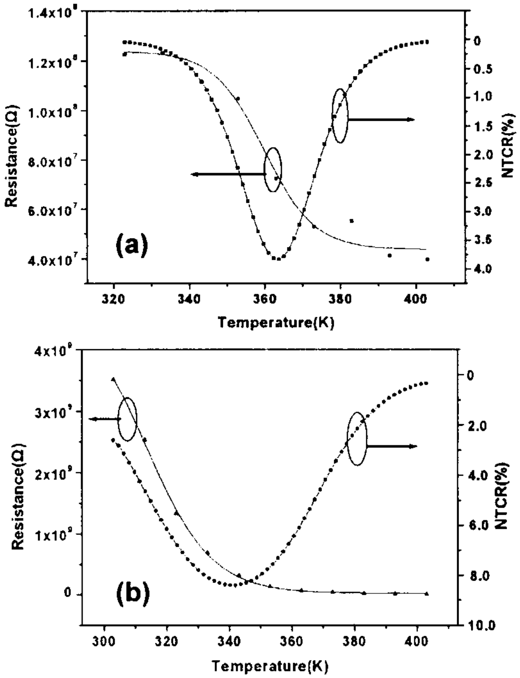

Figure 3. TCR of BST films (a) without post treatment and (b) with post RTA treatments at 500°C.under –3 volts.

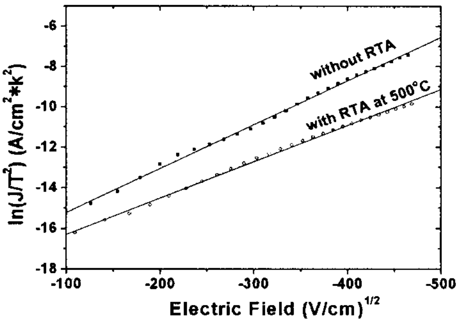

Figure 4. The log (J/T^2) versus $E^{1/2}$ plot showing Schottky emission fitting for the BST films under negative bias.

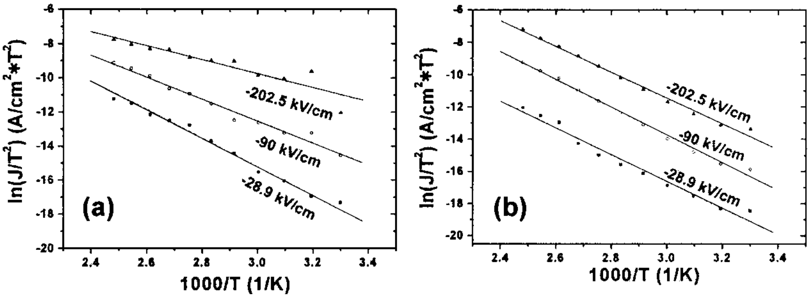

Figure 5. The log (J/T^2) versus 1/T plot showing Schottky emission fitting for the BST films treated (a) without RTA and (b) with RTA at 500°C under negative bias.

The temperature-coefficient-resistivity (TCR) is defined as $TCR = \frac{1}{Rs}\frac{dRs}{dT}\Big|_T$, where T and Rs represent the temperature and the resistance. Figure 3 indicates the TCR of BST films (a) without post treatment and (b) with post RTA treatments at 500°C. Both the films behave NTCR at a bias of -3 volts.

The conduction mechanisms of metal/BST/metal capacitors are usually interpreted as interface-limited and bulk-limited conductions [6]. In general, the Schottky emission (SE) governs the interface-limited current for the Pt/BST films. The leakage current governed by SE behavior is expressed as [7],

$$\log (J_{SE}/T^2) = -q[\varphi_B - (qE/4\pi \varepsilon_d \varepsilon_0)^{1/2}]/(kT\square \ln 10) + \log(A^*) \qquad (1)$$

where, A* is the effective Richardson's constant; φ_B is the potential barrier height at the surface; ε_d is the dynamic dielectric constant of the ferroelectric material in the infrared region; q is the unit charge; k is Boltzmann's constant; T is temperature, and E is the external electric field. Figure 4 shows that both the plots of $\log(J/T^2)$ against $E^{1/2}$ exhibit linear relations under negative bias. In addition, the plots of $\log(J/T^2)$ against 1/T exhibit linear relations, too, as shown in Fig. 5. Consequently, both the samples biased at negative voltage show the Schottky emission behavior.

The BST film without RTA treatment exhibits NTCR under positive bias, as shown in Fig. 6 (a), but the films treated with RTA at 500°C reveals PTCR in the range of current-measurement-temperature from room temperature to 330°K, as shown in Fig. 6 (b). The BST film treated without RTA exhibits SE behaviors,

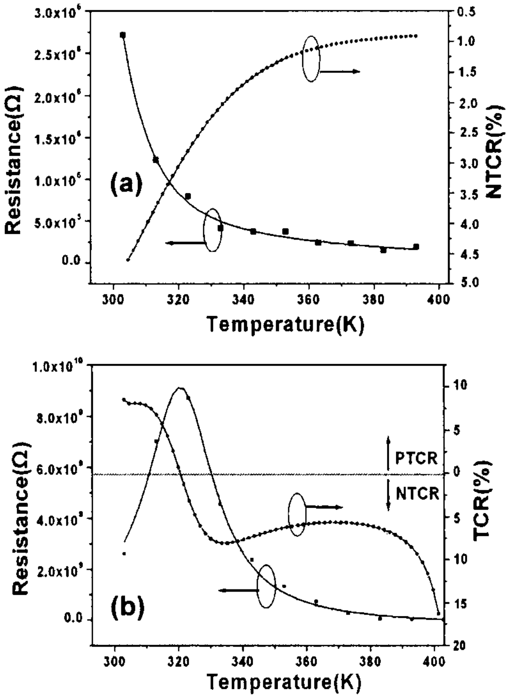

Figure 6. TCR of BST films (a) with RTA treatment and (b) with RTA treatment at 500°C under +3 Volts.

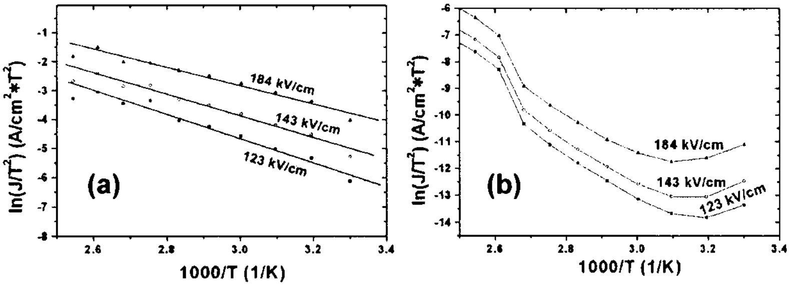

Figure 7. The log (J/T^2) versus 1/T plot showing Schottky emission fitting for the BST films treated (a)without RTA and (b) with RTA at 500°C under positive bias.

but the film treated with RTA at 500°C doesn't exhibit SE behavior at all, as shown in Fig. 7.

As mentioned above, the upper surface of BST film post treated by RTA in oxygen ambient is dominated by SE behavior under a negative bias. Hence, the negative-biased conduction current exhibits NTCR phenomenon according to the current-temperature relation of Eq. (1). On the other hand, the 500°C RTA treatment applied on BST/Pt films will behave a large thermal stress due to the temperature abruptly decreasing, and thus many interface states are induced at the interface of BST/Pt-bottom-electrode. The injected electron current, governed by the bulk-limited mechanism, is formed by the thermal-stress-induced interface states, and this bulk-limited conduction current film is confined by the Heywang barrier scattering [8]. The resistivity for the Heywang model are represented as $Rs \propto \exp(-q\phi / kT)$ and $\phi = \frac{e^2 N_d d^2}{2\varepsilon\varepsilon_0}$, where□ and d are the potential height and depletion width at grain boundary, so the resistivity increases as temperature rises. Thus, the RTA-treated BST film reveals PTCR phenomenon under positive bias.

CONCLUSIONS

The post RTA treatment in oxygen ambient can greatly suppress the conduction current for the Pt/BST/Pt films owing to the compensation of the oxygen vacancies. In this work, both of the Pt/BST/Pt multifilms reveal NTCR behavior under negative bias, but the sample treated by RTA exhibits PTCR behavior under positive bias. The results of the SE fitting indicate the Schottky barrier formed at upper interface, which results in the NTCR phenomenon for the sample biased at negative voltage. On the other hand, the thermal-stress-induced interface states cause the injected electron current, which is confined by Heywang barrier scattering. Hence, the positive biased BST film with RTA treatment reveals the PTCR phenomenon.

ACKNOWLEDGEMENTS

This work was supported in part by the Republic of China National Science Council (R.O.C. NSC) under the contract NSC-90-2215-E-009-074. Thanks are also due to the Semiconductor Research Center (SRC) in National Chiao Tung University and the National Nano Device Laboratory (NDL) of the R.O.C. NSC for their technical support.

REFERENCES

[1] Uchino Kenji: Ferroelectric devices. .New york: Marcel Dekker Inc; 2000.

[2] Wang Lichen and Zhou,Zhigang: A new smart material-multilayer PTC thermistor. Components, Packaging, and Manufacturing Technology, Part A, IEEE Transactions on. 1995; 18: 249 - 251

[3] Saburi O., J. Phys. Soc. Jpn., 1959, 14: 1159

[4] Lee KH, Hwang CS, Lee BT, Kim WD, Horii H, Kang CS, Cho HJ, Lee SI and Lee MY: Variation of Electrical Conduction Phenomena of Pt/ (Ba, Sr)TiO3/Pt Capacitors by Different Top Electrode Formation Processes. Jpn. J. Appl. Phys. 1997; 36: 5860-5865

[5] Smyth DM, Harmer MP and Peng PJ; J. Am. Ceram. Soc.; 1989; 72(12): 2276.

[6] Hwang CS, Lee BT, Kang CS, Kim JW, Lee KH, Cho HJ, Horii H, Kim WD, Lee SI, Roh YB and Lee MY; J. Appl. Phys.; 1998; 83(7): 3703.

[7] Tsai MS and Tseng TY; J. Electrochem. Soc.; 1998; 145(8): 2853.

[8] Lines ME and Glass AM: Principles and applications of ferroelectrics and related materials. Oxford, Clarendon Press;1977: 534-561