Impact of Time Dependent Dielectric Breakdown and Stress-Induced Leakage Current on the Reliability of High Dielectric Constant (Ba, Sr)TiO$_3$ Thin-Film Capacitors for Gbit-Scale DRAM’s

Shintaro Yamamichi, Akiko Yamamichi, Donggun Park, Tsu-Jae King, Member, IEEE, and Chenming Hu, Fellow, IEEE

Abstract—Time dependent dielectric breakdown (TDBB) and stress-induced leakage current (SILC) are investigated for the reliability of (Ba, Sr)TiO$_3$ (BST) thin films. Both time to breakdown ($T_{BD}$) versus electric field ($E$) and $E/8$ versus $I/E$ plots show universal straight lines, independent of the film thickness, and predict lifetimes longer than 10 y at +1 V for 50 nm BST films with an SiO$_2$ equivalent thickness of 0.70 nm. SILC is observed at +1 V after electrical stress of BST films; nevertheless, 10 y reliable operation for Gbit-scale DRAM’s is predicted in spite of charge loss by SILC. Lower (Ba + Sr)/Ti ratio is found to be strongly beneficial for low leakage, low SILC, long TBD, and therefore greater long-term reliability. This suggests a worthwhile tradeoff against the dielectric constant, which peaks at a (Ba + Sr)/Ti ratio of 1.05.

Index Terms—(Ba, Sr)TiO$_3$, capacitor, DRAM, high dielectric constant, reliability, SILC, TDBB.

I. INTRODUCTION

RECENTLY, (Ba, Sr)TiO$_3$ thin films have been intensively investigated for the application to capacitor dielectrics in ULSI devices, because of their high dielectric constant. Especially, Gbit-scale DRAM’s require a very small SiO$_2$ equivalent thickness ($t_{eq}$) of less than 1 nm to provide a sufficient capacitance of 25 fF/cell in a cell size smaller than 0.1 μm$^2$. Several technologies utilizing BST thin films and Ru-based electrodes have been presented for the demonstration of 1 Gbit and 4 Gbit DRAM’s [1]–[4]. Fundamental dielectric properties of BST thin films, such as $t_{eq}$ versus voltage characteristics, are also reported in previous publications. However, before ULSI manufacturing incorporates BST thin films, it is necessary to study their long term reliability. Although there are a few papers reporting the BST lifetime under high stress voltage [5]–[8], little attention have been paid to the BST thickness and composition dependences of time dependent dielectric breakdown (TDBB) characteristics and the comparison between BST and SiO$_2$ in terms of stress-induced leakage current (SILC) and lifetime. In this paper, we describe the intrinsic TDBB characteristics for BST thin films with thickness ranging from 50 to 160 nm. The (Ba + Sr)/Ti composition ratio dependence of TDBB is also studied for 50-nm BST films to compare BST and SiO$_2$ as capacitor dielectrics. Furthermore, we report for the first time the SILC observation in BST capacitors, and predict the relationship between SILC and lifetime of BST capacitors [9].

II. EXPERIMENT

A. Sample Preparation

(Ba$_{0.7}$Sr$_{0.3}$)$_3$TiO$_3$ thin films were prepared by ion beam sputtering. The detailed sputtering conditions were described in a previous publication [10]. The substrate was a single-crystal R-plane sapphire [$c$ – Al$_2$O$_3$(10 • 2)] coated with a Pd(500 nm) bottom electrode layer deposited by dc magnetron sputtering at room temperature. Since this structure provides a very flat and stable surface for BST deposition, it is possible to investigate the intrinsic electrical properties for BST films without any influence of reaction between BST and bottom electrodes. The BST deposition temperature was 650 °C, and ion beam sputtering was carried out at 2 × 10$^{-4}$ Torr total gas pressure, with argon and oxygen partial pressures were 1.2 × 10$^{-3}$ and 8 × 10$^{-5}$ Torr, respectively. The ion beam voltage and ion beam current were kept constant at 1000 V and 40 mA during sputtering, respectively. Deposition rates estimated by measuring the film thickness with a surface profiler (DEKTAK-3030) were 0.7–0.9 nm/min. There was no thermal treatment after BST deposition. The BST film thickness was varied from 50 to 160 nm to study the thickness dependence of TDBB and SILC. The (Ba + Sr)/Ti composition ratio (A/B ratio or $x$ value) in the films, determined by inductively coupled plasma mass spectroscopy, was also varied from 0.94 to 1.10 by changing the A/B ratio of the target. For electrical measurements, Au(300 nm)/Ti(50 nm) top electrodes were deposited by dc magnetron sputtering at room temperature, and patterned into 0.1-mm$^2$ squares by photolithography and wet etching.

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B. Electrical Measurements

Capacitance and $\tan \delta$ were measured from 100 Hz to 10 MHz frequency range with a Hewlett-Packard 4194A impedance analyzer. All the films described in this paper did not show any notable frequency dispersion in the measurement frequency range. Leakage current was measured with a Hewlett-Packard 4140B pA-m/s-source. Constant dc stress voltage was applied to the BST films to characterize $T_{\text{BD}}$, and the leakage current was monitored as a function of time every second until breakdown. Breakdown is defined as leakage current increasing to 10 A/cm$^2$ or ten times higher than at the previous one second time step. The $Q_{\text{BD}}$ value is calculated by integrating the leakage current from $t = 0$ to $t = T_{\text{BD}}$. For studying SILC, after dc voltage stress, the leakage current was measured at 1 V as a function of time, and these stressing and measurement cycles were repeated many times.

III. RESULTS AND DISCUSSIONS

A. Time Dependent Dielectric Breakdown

1) Thickness Dependence of TDDB: Fig. 1 shows the leakage current versus $E$ characteristics for 50-, 130-, and 160-nm BST films. The A/B ratio is fixed at 1.05, which gives the highest dielectric constant for all the thicknesses studied. The dielectric constant is also shown in the superimposed figure as a function of film thickness. The leakage current is improved by increasing the film thickness. The 50-nm films shows a $t_{\text{eq}}$ value of 0.70 nm and leakage current smaller than $1 \times 10^{-5}$ A/cm$^2$ at 0.2 MV/cm, which is 1 V across the 50-nm film. The leakage current versus time characteristic is shown in Fig. 2 for 50-nm BST films at stress voltage from 7.8 to 10.0 V. Sudden breakdown, similar to the SiO$_2$ TDDB, was observed. This kind of breakdown is called hard breakdown, and was clearly observed for all the BST films discussed in this paper.

Log($T_{\text{BD}}$) is plotted as a function of applied voltage for 50-, 130-, and 160-nm BST films, as shown in Fig. 3. All the BST films show a longer lifetime than 10 y at +1 V. This result indicates the long term intrinsic reliability of BST capacitors for the Gbit-scale DRAM applications. These log($T_{\text{BD}}$) data are replotted versus $E$ and $1/E$, as shown in Fig. 4. Apparently, universal straight lines independent of film thickness can be obtained from both these plots. These straight lines indicate two possible extrapolations for the lifetime of BST capacitors. Table I shows the comparison of BST and SiO$_2$ lifetimes. The $E_1$ and $E_2$ represent the extrapolated electric fields for 10 y lifetime from the $E$ and $1/E$ dependences of $T_{\text{BD}}$, respectively. The 10 y breakdown field for BST capacitors estimated by $1/E$ model is longer than that estimated by $E$ model, as generally discussed in...
TABLE I

<table>
<thead>
<tr>
<th>d(nm)</th>
<th>( \beta )</th>
<th>( \beta \times d )</th>
<th>( E_1 )</th>
<th>( E_2 )</th>
<th>ref</th>
</tr>
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<tr>
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<td>30</td>
<td>58</td>
<td>1740</td>
<td>---</td>
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<tr>
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<td>0.86</td>
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<tr>
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<td>26</td>
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<td>0.24</td>
<td>0.86</td>
</tr>
<tr>
<td>BST</td>
<td>160</td>
<td>26</td>
<td>4160</td>
<td>0.24</td>
<td>0.86</td>
</tr>
</tbody>
</table>

Fig. 5. \( Q_{BD} \) as a function of \( E \). Thinner BST films show larger \( Q_{BD} \), which can be explained with universal \( T_{BD} \) versus \( E \) and larger leakage current, shown in Figs. 1 and 4.

The \( \beta \) for BST and SiO\(_2\) films. The units of \( \beta \), \( E_1 \), and \( E_2 \) are MV/cm, year, and year, respectively. The \( \beta \times d \) value is not the same for BST and SiO\(_2\). The lifetime of BST is a function of the electric field, not operating voltage.

The SiO\(_2\) case. Chen et al. and Horikawa et al. reported the linear relationships in \( \log(T_{BD}) \) versus \( 1/E \) and \( \log(T_{BD}) \) versus \( E \) for BST films, respectively, [7], [8]. However, as shown in Fig. 4, linear relationships in both \( \log(T_{BD}) \) versus \( E \) and versus \( 1/E \) can be obtained from the same data set. As for the \( 1/E \) dependence of \( T_{BD} \), which is represented as \( T_{BD} = \alpha \exp(\beta/E) \), Horikawa et al. reported that the product of exponential coefficient \( \beta \) and film thickness \( (\beta \times d_{film}) \) for 30-nm BST films was the same as that for 5-nm SiO\(_2\) films. However, when a wider BST film thickness is considered, the \( \beta \times d_{film} \) product is no longer the same as that for 5-nm SiO\(_2\) films, as shown in Table I. This simply means that the breakdown is essentially determined by the electric field, not by the applied voltage. The difference in \( \beta \) value itself might show the difference in reliability lifetime for sputtered and CVD BST films. The charge to breakdown \( (Q_{BD}) \) also depends on the electric field, as shown in Fig. 5. Additionally, it depends on BST film thickness, and thinner films show larger \( Q_{BD} \) values. This is consistent with the universal \( T_{BD} \) versus \( E \) relationship and larger leakage current in thinner BST films shown in Figs. 1 and 4.

2) \((\text{Ba} + \text{Sr})/\text{Ti} \) Ratio Dependence of TDDB: Next, the \((\text{Ba} + \text{Sr})/\text{Ti} \) dependence of TDDB is investigated especially for thin 50-nm BST films. Fig. 6 shows the fundamental dielectric properties for BST films with A/B ratio varying from 0.94 to 1.10. The leakage current decreases progressively with decreasing A/B ratio. On the other hand, the 5% A-site rich films \((x = 1.05)\) show the smallest \( t_{eq} \) value, as shown in the superimposed figure in Fig. 6. This improvement in \( t_{eq} \) values for slightly (Ba + Sr) rich films would be attributed to the crystallinity improvement, such as large grain growth, caused by compensation of the oxygen vacancies, but excess (Ba + Sr) ions degrades the leakage current characteristics [10]. For these 50 nm BST films, the larger \( T_{BD} \) are obtained for the films with smaller \( x \) values, as shown in Fig. 7. The slopes of the lines are nearly identical, and the films with smaller \( x \) values show the larger \( T_{BD} \).

Fig. 6. Leakage current versus electric field characteristics for 50-nm BST films with \( x = 0.94, 1.02, 1.05, \) and 1.10. The \( t_{eq} \) values are shown in the superimposed figure.

Fig. 7. \( T_{BD} \) versus \( E \) for 50-nm BST films with \( x = 0.94, 1.02, 1.05, \) and 1.10. The slopes of the lines are nearly identical, and the films with smaller \( x \) values show the larger \( T_{BD} \).
Instead of lifetime, the 10 y breakdown field ($E_b$) extrapolated from $T_{BD}$ versus $E$ and $T_{BD}$ versus $1/E$ plots is shown in Fig. 8, as a function of $A/B$ ratio for 50-nm BST films. The $1/E$-model shows larger breakdown field than the $E$-model, as discussed in the previous section. To compare BST and SiO$_2$ as capacitor dielectrics, the SiO$_2$ equivalent 10 y breakdown field ($E_{eq}$) is defined by the following equation, and also plotted in Fig. 8:

$$E_{eq} = E_b \times \frac{d_{th}}{t_{eq}}.$$  

From these plots, BST materials are found to be much more attractive than SiO$_2$ as capacitor dielectrics, because the $E_{eq}$ values from 60 to 110 MV/cm is obtained for BST films with $x$ smaller than 1.05 from $T_{BD}$ to $E$ model, and these values are 6 to 12 times larger than 9 MV/cm for 2.5 nm SiO$_2$ films [12].

### B. Observation of Stress-Induced Leakage Current

Fig. 9 shows the transient current characteristics for 50-nm BST films with $x = 0.94$ at +1 V. Two successive measurements show almost the same transient leakage decay characteristics, suggesting that charge trapping and quick detrapping are observed in BST films. The sample is kept to be shorted during the interval time for 30 s. However, after applying high dc stress voltage, the transient decaying current can no longer be observed but a much higher level of leakage current appears. This current is called SILC in the SiO$_2$ case. Fig. 10(a) and (b) shows the leakage current versus time characteristics for 50- and 130-nm BST films with $x = 1.05$ after applying the stress of 1.4 MV/cm, respectively. SILC is clearly observed, and increases with increasing stress current charge, which is calculated by integrating the stress current over the stress time. Similar degradation is seen in 50- and 130-nm BST films with $x = 0.94$ after applying a stress of 2.0 MV/cm, as shown in Fig. 11(a) and (b), respectively. The SILC is smaller than that for BST with $x = 1.05$ even at higher stress field. So, small $x$ is correlated with small leakage current, less SILC, and longer TBD. It is interesting that thinner BST films show smaller SILC degradation, as shown in Figs. 10 and 11. This result is probably related to the smaller number of traps in thinner films.

In DRAM applications, SILC can have an impact on capacitor lifetime. First, the charge loss ($Q_{loss}$) is calculated by integrating the leakage current from 1 ns up to 10 s, extrapolating the leakage versus time ($t$) characteristics toward 1 ns. As the transient current shows the $1/t$ dependence in Fig. 9, we assume that the leakage current for stressed films does not exceed this initial transient current in shorter time range than 1 s. Ten seconds are large enough as a refreshing time for Gbit-scale DRAM’s. Then, the $\Delta Q_{loss}$ is defined as the difference in $Q_{loss}$ before and after stressing. For 1 Gbit DRAM’s, $\Delta Q_{loss}$ should be limited to less than $5 \times 10^{-6}$ C/cm$^2$, assuming a capacitor area of 0.1 $\mu$m$^2$ and a minimum acceptable capacitance loss of 5 fF/cell. On the other hand, the total stress charge, $Q_{stress}$, is estimated to be 3 C/cm$^2$ in 10 y, assuming a leakage current of $1 \times 10^{-6}$ A/cm$^2$ at +1 V. Here, it is conservatively assumed that 3 C/cm$^2$ of stress even at low voltage of +1 V leads to the same SILC as 3 C/cm$^2$ at high stress voltage as shown in Figs. 10 and 11. Fig. 12 shows $\Delta Q_{loss}$ as a function of $Q_{stress}$ for BST films with two different thicknesses and A/B ratios. $\Delta Q_{loss}$ increase at certain $Q_{stress}$ for BST films with $x = 1.05$. On the contrary, it hardly increases for BST films with $x = 0.94$ even at a large $Q_{stress}$ of 100 C/cm$^2$. Additionally, thinner BST films show larger endurance for stressing. Consequently, the capacitor lifetime can be much longer than 10 y for 1 Gbit DRAM’s with regard to charge loss caused by SILC.

### IV. CONCLUSIONS

TDDB and SILC characteristics were investigated for the reliability of BST thin films. Both $T_{BD}$ versus $E$ and $T_{BD}$ versus $1/E$ plots showed universal straight lines, independent of the film thickness, and projected a lifetime longer than 10 y at +1 V for 50 nm BST films with $t_{eq}$ of 0.70 nm. The breakdown lifetime was correlated with the leakage current, but there was no monotonic relationship between dielectric constant values and lifetime. The SiO$_2$ equivalent breakdown field from 60 to 110 MV/cm were extrapolated for A/B ratio of 1.05 to 0.94 for 10 y lifetime using $1/E$ plot. Twenty to 75 MV/cm were extrapolated using $E$ plot. These values
Fig. 10. Current versus time characteristics at +1 V for (a) 130-nm and (b) 50-nm BST films with $x = 1.05$ after stress charge injection. Large SILC is observed at low voltage of +1 V.

Fig. 11. Current versus time characteristics at +1 V for (a) 130-nm and (b) 50-nm BST films with $x = 0.94$ after stress charge injection. SILC is smaller than that shown in Fig. 10.

Fig. 12. $\Delta Q_{\text{loss}}$ versus $Q_{\text{stress}}$ plot. The thinner BST shows the smaller $\Delta Q_{\text{loss}}$, and larger endurance for stressing. The 10 y operation is successfully guaranteed for 1 Gbit DRAM's in spite of charge loss caused by SILC.

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