Temperature dependence of the current conduction mechanisms in LaAlO$_3$ thin films

Ingram Yin-Ku Chang and Joseph Ya-Min Lee$^a$
Department of Electronics Engineering and Institute of Electronics Engineering, National Tsing-Hua University, Hsinchu 30013, Taiwan

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Metal-oxide-semiconductor capacitors and transistors with LaAlO$_3$ dielectric films were fabricated and the current conduction mechanisms were studied. The LaAlO$_3$ films remained amorphous with postdeposition annealing up to 1000 °C. The leakage current density was $8.3 \times 10^{-5}$ A/cm$^2$ at $-1$ V. The low leakage current was attributed to the high barrier height of Al/LaAlO$_3$ interface. The Al/LaAlO$_3$ barrier height and the effective electronic mass calculated from Schottky emission and Fowler–Nordheim tunneling were 1.12 eV and 0.27$m_h$, respectively. The dominant conduction mechanism in the temperature range of 300 K $< T <$ 420 K was space-charge-limited current, and the trapping depth was determined to be 0.36 ± 0.1 eV. © 2008 American Institute of Physics. [DOI: 10.1063/1.3039074]

Recently, many candidates such as tantalum oxide (Ta$_2$O$_5$)$_2$, titanium dioxide (TiO$_2$)$_2$, aluminum oxide (Al$_2$O$_3$)$_3$, zirconium oxide (ZrO$_2$)$_4$, hafnium oxide (HfO$_2$)$_5$, dysprosium oxide (Dy$_2$O$_3$)$_6$, lanthanum oxide (La$_2$O$_3$)$_7$, and several of the associated aluminates have been introduced as alternative gate dielectrics. Among these high-$k$ oxides, the thermal stability ($\sim$900 °C) (Ref. 14) and the leakage current are important issues for future metal-oxide-semiconductor field-effect transistor devices. Lanthanum aluminates (LaAlO$_3$) are potential candidates to achieve high dielectric constant (13–27), large energy band gap (over 5 eV), high thermal stability (up to 2100 °C), and low leakage current density. In the literature, Shao et al. showed good thermal stability up to a high temperature of 850 °C. Yan et al. reported that the leakage current of the LaAlO$_3$ sample was $2.3 \times 10^{-4}$ A/cm$^2$ at +1 V dc bias voltage. Edge et al. observed the band offset of 1.8 ± 0.2 eV for electrons and 3.2 ± 0.2 eV for holes by x-ray photoelectron spectroscopy. However, there are relatively few reports on the temperature dependence of the conduction mechanisms with LaAlO$_3$ dielectric. In this work, the conduction mechanisms such as Schottky emission, Fowler–Nordheim (FN) tunneling, and space-charge-limited conduction (SCLC) were studied in the temperature range from 11 to 555 K.

$P$-type (100) orientation 4 in. diameter silicon wafers were used as the starting substrates. Following wafer cleaning, the LaAlO$_3$ films were deposited by rf magnetron sputtering. After LaAlO$_3$ film deposition, the postdeposition annealing (PDA) was performed at 700 °C for 30 s. Finally, aluminum electrodes were deposited using shadow mask by reactive dc magnetron sputtering. The capacitance-voltage and current-voltage measurements were performed using a high-frequency C-V/I-V meter (Megabtyek MI494) and a Keithley 236, respectively. The thickness, refractive index, and energy band gap of LaAlO$_3$ films were measured using an n&k analyzer (model 1200).

Figure 1 shows the $C-V$ curves of LaAlO$_3$ metal-oxide-semiconductor (MOS) capacitors with various thicknesses annealed at 700 °C for 30 s. The dielectric constant of capacitors with 8.5, 9.6, and 13.3 nm LaAlO$_3$ films are 13.0, 13.4, and 14.7, respectively. The leakage current density is $8.3 \times 10^{-5}$ A/cm$^2$ at $-1$ V. The inset of Fig. 1 shows the x-ray diffraction (XRD) results of LaAlO$_3$ thin films with high temperatures annealed. The LaAlO$_3$ films remain amorphous with PDA up to 1000 °C. When the annealing temperature is higher than 800 °C, the diffusion of silicon atoms into the LaAlO$_3$ film was observed in the secondary ion mass spectroscopy measurements. The application of LaAlO$_3$ is therefore limited below 800 °C. The experiments in this work were carried out at 700 °C.

Figure 2(a) shows that the Schottky emission plot of $\log(J/T^2)$ versus $E^{1/2}$ for the conduction current of LaAlO$_3$ capacitors under negative bias. The Schottky emission is given by

![Graph showing C-V curves and XRD results of LaAlO$_3$ thin films.](https://example.com/figure1.png)

**FIG. 1.** High-frequency capacitance-voltage ($C-V$) curves of LaAlO$_3$ MOS capacitors with various thicknesses annealed at 700 °C in nitrogen for 30 s. The inset shows the XRD results of LaAlO$_3$ thin films with high temperatures annealed from 700 to 1000 °C.
\[
J_S = A^* T^2 \exp \left[ -q (\varphi_B - \sqrt{qE/4ne_cE_0}) / kT \right],
\]

where \(A^*\) is the effective Richardson constant \(120(m^*/m_0) A/cm^2 K^2\), \(m^*\) is the effective electronic mass, \(q\varphi_B\) is the Schottky barrier height, \(E\) is the electric field, and \(q\) is the electronic charge. If the conduction current is governed by Schottky emission, a \(\log(J/T^2)\) versus \(E^{1/2}\) plot should show a straight line and the slope will give the dynamic dielectric constant. Because \(n_0 \approx \sqrt{e_c}\), and the measured refractive index is 1.80, the dynamic dielectric constant \(e_d\) should be close to 3.24. As can be seen in Fig. 2(a), the conduction mechanism in the electrical field from 0.4 to 1.6 MV/cm and at the temperature from 525 to 555 K is Schottky emission.

Figure 2(b) shows the FN plot of \(\ln(J/E^2)\) versus \(1/E\) in the temperature range from 11 to 260 K under negative bias. The FN tunneling is given by

\[
J_{FN} = \frac{q^2}{8\pi \hbar e} E^2 \exp \left[ -\frac{8\pi \sqrt{2m^* (q\varphi_B)^3}}{3q\hbar E} \right],
\]

where \(q\varphi_B\) is the barrier height between Al and LaAlO\(_3\). The conduction mechanism with the electric field above 0.8 MV/cm is found to be FN tunneling. In this work, the electronic effective mass \(m^*\) and the energy barrier \(q\varphi_B\) were determined from Schottky emission and FN tunneling by an iteration method. The two variables can be determined by two equations from the two conduction mechanisms. The intersection point gives both the barrier height and the effective electronic mass. The Al/LaAlO\(_3\) barrier height and the effective electronic mass thus determined are 1.12 eV and 0.27\(m_0\), respectively.

Figure 3 shows the SCLC plot using MOS capacitors with LaAlO\(_3\) film. Assuming a single trap level, the \(J-V\) relation of SCLC with traps can be written as

\[
J = \frac{9}{8} \mu e_c \theta V^2 / d^2,
\]

where \(\mu\) is the effective carrier mobility, \(e_c\) is the permittivity of free space, \(e_d\) is the dynamic dielectric constant, \(d\) is the thickness of LaAlO\(_3\) film, and \(\theta\) is the ratio of free to trapped charges. If the traps in LaAlO\(_3\) film are shallow, \(\theta\) can be expressed as

\[
\theta = N_t / N_c \exp \left[ -\left( E_C - E_t \right) / kT \right],
\]

where \(N_t\) is trap density, \(N_c\) is density of the states in the conduction band, \(k\) is Boltzmann’s constant, and \(T\) is the absolute temperature. The SCLC transition voltage \(V_{th}\) is given by

\[
V_{th} = \frac{8}{9} \times \frac{q n_0 d^2}{e_c \theta e_r}.
\]

where \(n_0\) is the concentration of the free charge carriers in thermal equilibrium. When the applied voltage exceeds \(V_{TFL}\), sufficient charges have been injected into the LaAlO\(_3\) film to fill the traps. The current thus rises rapidly. The voltage at the trap filled limit \(V_{TFL}\) is given by

\[
V_{TFL} = \frac{qN_t d^2}{2e_0 e_r}.
\]

The trap concentration \(N_t\) calculated from \(V_{TFL}\) is \(1.2 \times 10^{19}\) cm\(^{-3}\). The activation energy \(E_C - E_t\) can be calculated from the \(J-V\) relation of (II) region. Since \(N_c\) in \(\theta\) is

\[
N_c = \frac{1}{\theta} \frac{1}{N_t} \exp \left[ \frac{E_C - E_t}{kT} \right].
\]

\[
\theta = \frac{1}{N_C} \exp \left[ -\frac{E_C - E_t}{kT} \right].
\]

### Table I. A comparison of electrical properties of MOS capacitors with ZrO\(_2\), HfO\(_2\), La\(_2\)O\(_3\), Dy\(_2\)O\(_3\), and LaAlO\(_3\) (this work) extracted by the same method.

<table>
<thead>
<tr>
<th>Gate dielectrics</th>
<th>(J_{th}) (A/cm(^2))</th>
<th>(\varphi_B) (V)</th>
<th>(V_{th}) (V)</th>
<th>(V_{TFL}) (V)</th>
<th>(\mu) (cm(^2)/V s)</th>
<th>(n_0) (cm(^{-3}))</th>
<th>(N_t) (cm(^{-3}))</th>
<th>(E_C - E_t) (eV)</th>
<th>(\theta)</th>
<th>(N_c) (cm(^{-3}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>ZrO(_2) (^a)</td>
<td>(-3 \times 10(^{-4}))</td>
<td>1.06</td>
<td>(\ldots)</td>
<td>(\ldots)</td>
<td>(\ldots)</td>
<td>(\ldots)</td>
<td>(\ldots)</td>
<td>(\ldots)</td>
<td>(\ldots)</td>
<td>(\ldots)</td>
</tr>
<tr>
<td>HfO(_2) (^b)</td>
<td>(-6 \times 10(^{-5}))</td>
<td>0.98</td>
<td>0.3</td>
<td>8.15 \times 10(^{-7})</td>
<td>1.2 \times 10(^{14})</td>
<td>9.2 \times 10(^{17})</td>
<td>0.21</td>
<td>892</td>
<td>5.54 \times 10(^{18})</td>
<td></td>
</tr>
<tr>
<td>La(_2)O(_3) (^c)</td>
<td>(-5 \times 10(^{-5}))</td>
<td>0.75</td>
<td>1.3</td>
<td>1.2 \times 10(^{-8})</td>
<td>9.7 \times 10(^{17})</td>
<td>1.7 \times 10(^{19})</td>
<td>0.20</td>
<td>0.2</td>
<td>4.53 \times 10(^{21})</td>
<td></td>
</tr>
<tr>
<td>Dy(_2)O(_3) (^d)</td>
<td>(-1 \times 10(^{-5}))</td>
<td>0.75</td>
<td>1.3</td>
<td>4.0 \times 10(^{-10})</td>
<td>8.7 \times 10(^{18})</td>
<td>1.2 \times 10(^{19})</td>
<td>0.36</td>
<td>0.03</td>
<td>1.2 \times 10(^{23})</td>
<td></td>
</tr>
<tr>
<td>LaAlO(_3) (^e)</td>
<td>8.3 \times 10(^{-5})</td>
<td>1.12</td>
<td>0.27</td>
<td>0.87</td>
<td>(\ldots)</td>
<td>(\ldots)</td>
<td>(\ldots)</td>
<td>(\ldots)</td>
<td>(\ldots)</td>
<td>(\ldots)</td>
</tr>
</tbody>
</table>

\(^a\)Reference 25.
\(^b\)Reference 26.
\(^c\)Reference 13.
\(^d\)Reference 27.
\(^e\)This work.
The inset of Fig. 3 shows the activation energy obtained from the slope of plotting \( \ln(J/T) \) versus \( 1/T \). The trap depth calculated in the temperature range from 300 to 420 K is about 0.36 eV. The experimental results show that the leakage current density with \( \text{LaAlO}_3 \) dielectric is the lowest in comparison with other high-\( k \) films. The reason of the low leakage current density is attributed to the high barrier height of \( \text{Al}/\text{LaAlO}_3 \) interface.

In summary, MOS capacitors with \( \text{LaAlO}_3 \) dielectric film were fabricated and the conduction mechanisms were studied. The \( \text{Al}/\text{LaAlO}_3 \) barrier height and the effective electronic mass calculated from Schottky emission and FN tunneling were 1.12 eV and 0.27\( m_0 \), respectively. The electrical properties of MOS capacitors with \( \text{LaAlO}_3 \) dielectric were determined from SCLC conduction mechanism, and the trapping depth calculated in the temperature range from 300 to 420 K was about 0.36 ± 0.01 eV. The experimental results show that the \( \text{LaAlO}_3 \) films remain amorphous with PDA up to 1000 °C and the low leakage current density of \( \text{LaAlO}_3 \) films is attributed to the high barrier height of \( \text{Al}/\text{LaAlO}_3 \) interface.

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