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Electrical characterization of amorphous lanthanum aluminate thin films grown by molecular-beam deposition on silicon

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Amorphous LaAlO$_3$ thin films were deposited at room temperature directly on n-type and p-type Si (001) by molecular beam deposition. The dielectric properties of the stoichiometric amorphous LaAlO$_3$ thin films deposited on silicon were determined through capacitance-voltage and current-voltage measurements. The electrical measurements indicate that the amorphous LaAlO$_3$ thin films have a dielectric constant ($K$) of $K=16\pm2$. This is significantly lower than the $K=24$ of crystalline LaAlO$_3$. The equivalent oxide thickness values range between 9.8 and 15.5 Å for films deposited on n-type silicon with physical thicknesses of 45–75 Å. © 2006 American Institute of Physics. [DOI: 10.1063/1.2182019]

The scaling of SiO$_2$ in metal-oxide-semiconductor field-effect transistors (MOSFETs) is approaching its fundamental limit, which has been estimated to be 7 Å.$^{1,2}$ To transcend this limit, new gate dielectric materials must be developed to replace SiO$_2$. The 2005 edition of the International Technology Roadmap for Semiconductors anticipates the use of higher dielectric constant (high-$K$) gate dielectrics in production as early as 2007. The first high-$K$ MOSFETs to enter the market will have gate dielectrics with $K$ ranging between 10 and 20 and will later be replaced with materials having $K$ greater than 20. One of these higher $K$ candidate materials is LaAlO$_3$. LaAlO$_3$ is a promising high-$K$ gate dielectric due to its good thermal stability, high optical band gap (5.6 eV for crystalline LaAlO$_3$ and 6.2 eV for amorphous LaAlO$_3$), and high band offsets (1.8 eV for electrons and 3.2 eV for holes) for amorphous LaAlO$_3$. $^{11}$

In crystalline form, the dielectric constant of LaAlO$_3$ is 24.1±0.1 (measured at 145 GHz).$^{12}$ There is a significant discrepancy, however, in the literature for the dielectric constant of amorphous LaAlO$_3$ thin films. Part of this discrepancy could be due to the films being off composition, as $\alpha$-Al$_2$O$_3$ has a $K$ ranging from 9.4 to 11.6 as a function of direction.$^{13}$ If we restrict our consideration to only La$_{1-x}$Al$_x$O$_3$ films with $0.8\leq x\leq 1.2$, the reported values vary from $K=13$ to 27 depending on the deposition technique. La$_{0.9}$Al$_{1.1}$O$_3$ films deposited by atomic layer deposition (ALD) have a reported $K=13$, $^{14}$ sputtered LaAlO$_3$ films have a $K=14$, $^{15}$ electron-beam deposited LaAlO$_3$ films have an estimated $K=21$, $^{16,17}$ metallicorganic chemical vapor deposition deposited La$_{0.8}$Al$_{1.2}$O$_3$ films have a $K=25$, $^{18}$ and pulsed-laser deposited La$_{0.9}$Al$_{0.9}$O$_3$ films have a $K=25$. $^{19}$

In this letter, we report the dielectric properties of amorphous LaAlO$_3$ thin films deposited on silicon by molecular-beam deposition (MBD) using elemental lanthanum, aluminum, and O$_2$ sources. The LaAlO$_3$ thin films are stoichiometric and have no detectable interfacial layer between the LaAlO$_3$ and the silicon. $^{8,21}$ The dielectric properties were extracted through capacitance-voltage (C-V) and current-voltage (I-V) characterization.

Amorphous LaAlO$_3$ thin films (45, 60, and 75 Å thick) were grown by MBD in an EPI 930 MBD chamber modified for the growth of oxides. The thickness of the films was determined from x-ray reflectivity (XRR) measurements using a Bede 300 mm system. The Bede RFS software was used to fit the XRR data. The films were grown on both n-type (phosphorous doped, $0.2–1\times 10^{15}$ P/cm$^3$) and p-type (boron doped, $1–2\times 10^{19}$ B/cm$^3$) Si (001) wafers. The silicon wafers were cleaned using a 50:1 solution of de-ionized (DI) water: concentrated hydrofluoric acid (HF). The amorphous LaAlO$_3$ films were formed by codepositing lanthanum, aluminum, and molecular oxygen at a background pressure of $6\times 10^{-6}$ Torr onto unheated silicon wafers. The lanthanum and aluminum fluxes were each $2\times 10^{13}$ atoms/cm$^2$ s. This deposition process has been shown to produce an abrupt amorphous LaAlO$_3$/Si interface free of any detectable interfacial SiO$_2$. $^{21}$ The films were capped in situ with a 1000-Å-thick aluminum film that served as the top electrode. The aluminum was patterned and wet etched using the Transene-type A aluminum etch to produce 50 μm × 50 μm squares.

Several 1500-Å-thick amorphous LaAlO$_3$ films have been analyzed by Rutherford backscattering spectrometry (RBS) (1.4 MeV He$^+$, 170° scattering angle). RBS indicates that the films are stoichiometric with a ratio of La:Al = $1\pm0.05$ (not shown).

The C-V measurements were performed in a probe station using a HP 4284A. The electrical measurements were made on the as-deposited films and the results presented here are from C-V measurements made on virgin devices. The I-V measurements were made using a HP 4158A.
The thickness of the LaAlO$_3$ thin films was determined using XRR. Figure 1 shows the XRR measurement and the model for the 60±2 Å thin film. The physical thickness of the LaAlO$_3$ thin films extracted by XRR is in agreement with high-resolution transmission electron microscopy measurements on similar films.\textsuperscript{21}

Figure 2 shows the $C$-$V$ measurements of films deposited on $n$-type and $p$-type silicon, respectively. The equivalent oxide thickness (EOT) indicated in the figures was determined using the Hauser CVC modeling program.\textsuperscript{24} The EOT values range between 9.8 and 15.5 Å for films deposited on $n$-type silicon with physical thicknesses of 45–75 Å. The EOT values range between 12.2 and 20.1 Å for films deposited on $p$-type silicon with physical thicknesses of 45–75 Å.

Figure 3 shows the $I$-$V$ measurements on $n$-type and $p$-type silicon, respectively, for the same films measured in Fig. 2. The leakage current densities are significantly lower for the films deposited on the $n$-type wafers and range between $1 \times 10^{-3}$ and $6 \times 10^{-7}$ A/cm$^2$ at $V_{fb}+1$ V for films with physical thicknesses of 45 and 75 Å. The leakage current densities for the films deposited on $p$-type wafers range between 0.8 and $8.5 \times 10^{-3}$ A/cm$^2$ at $V_{fb}-1$ V for films with physical thicknesses of 45–75 Å.

Figure 4 shows the forward and reverse bias sweeps for 45 Å LaAlO$_3$ thin films deposited on $n$-type and $p$-type silicon, respectively. The $n$-type wafers showed significantly lower hysteresis than the $p$-type wafers. Several postdeposition anneals have also been performed to try to reduce the

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**FIG. 1.** X-ray reflectivity measurements and modeled data for an amorphous LaAlO$_3$ film with a physical thickness of 60±2 Å.

**FIG. 2.** (a) $C$-$V$ curves as a function of amorphous LaAlO$_3$ film thickness on $n$-type wafers. (b) $C$-$V$ measurements as a function of thickness on $p$-type wafers. The measurements were made at 190 kHz on 50 μm × 50 μm capacitors.

**FIG. 3.** (a) $I$-$V$ measurements as a function of amorphous LaAlO$_3$ film thickness on the same $n$-type wafers shown in Fig. 2(a). (b) $I$-$V$ measurements as a function of thickness on the same $p$-type wafers shown in Fig. 2(b).

**FIG. 4.** (a) Forward and reverse bias $C$-$V$ measurements on a 45-Å-thick amorphous LaAlO$_3$ thin film deposited on $n$-type silicon. The hysteresis was 66.1 mV. (b) Forward and reverse bias $C$-$V$ measurements on a 45-Å-thick amorphous LaAlO$_3$ film deposited on $p$-type silicon. The hysteresis was 272 mV.
hysteresis on p-type wafers. For example, samples annealed in situ in UHV directly after MBD at temperatures ranging between 400 and 600 °C for 30 min exhibited no improvement in the hysteresis. Subsequent ex situ annealing in forming gas (4% hydrogen and 96% nitrogen) at 350 °C for 30 min also did not improve the hysteresis of the LaAlO₃ thin films deposited on p-type silicon. The as-deposited films were also ex situ annealed in N₂ for 30 min at 750 °C, but this resulted in breaking down the films.

It was possible to eliminate the hysteresis by making multiple voltage sweeps on the same film. This indicates that there are traps in the LaAlO₃ thin films deposited on p-type silicon. It is suspected that there might be a defect level in the band structure in the LaAlO₃ near the valance band that causes the trapped states.

Figure 5 shows the EOT as a function of physical thickness for the films deposited on n-type and p-type silicon. The lack of a detectible interfacial layer results in the intersection with the plot origin. From the slope of this plot, the dielectric constant of an isotropic material. Therefore, as the molecular volume increases, the dielectric constant decreases. The increased molecular volume of our films resulting from their low density could be the reason for the reduced dielectric constant of amorphous LaAlO₃ thin films deposited by MBD compared to crystalline LaAlO₃.

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