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Amorphous lanthanum lutetium oxide thin films as an alternative high-κ gate dielectric

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Lanthanum lutetium oxide thin films were grown on (100) Si by pulsed laser deposition. Rutherford backscattering spectrometry, atomic force microscopy, x-ray diffraction, and x-ray reflectometry were employed to investigate the samples. The results indicate the growth of stoichiometric and smooth LaLuO₃ films that remain amorphous up to 1000 °C. Internal photoemission and photoconductivity measurements show a band gap width of 5.2±0.1 eV and symmetrical conduction and valence band offsets of 2.1 eV. Capacitance and leakage current measurements reveal C-V curves with a small hysteresis, a dielectric constant of ≈32, and low leakage current density levels. © 2006 American Institute of Physics. [DOI: 10.1063/1.2393156]

The study of ultrathin gate dielectrics has recently gained great attention due to the technological need to replace SiO₂ films in metal-oxide-semiconductor field-effect transistors (MOSFETs).¹ The scaling has led to MOSFETs with ultrashort physical gate lengths (~50 nm) and insulating SiO₂-based films with thickness less than 1 nm. At such a thickness, these films suffer from excessively high leakage of charge carriers and poor reliability with respect to dielectric breakdown. Therefore, to overcome these limitations new gate dielectric materials with a higher dielectric constant must be developed to replace the SiO₂. According to the International Technology Roadmap for Semiconductors,² the implementation of high-κ gate dielectrics with a dielectric constant κ between 10 and 20 will be required by 2008, which will later be replaced by materials having a κ larger than 20, in order to meet both low leakage current density and performance requirements.

Ternary rare earth oxides (e.g., DyScO₃ and GdScO₃) are emerging as promising candidates for high-κ applications. As shown by Schom and Haeni,³ single crystals of these oxides show κ values of 20–35 which were also observed for amorphous LaScO₃, GdScO₃, and DyScO₃ films deposited on silicon (κ=22–23).⁴,⁵ In addition, these materials fulfill the requirements for large optical band gaps (5.6 eV) and band offsets (2–2.5 eV),⁴ while their amorphous phase is stable up to 1000 °C (for GdScO₃ and DyScO₃).⁴,⁵ Lanthanum lutetium oxide (LaLuO₃), as a member of this class of ternary oxides, is predicted to have similar properties.³,⁴,⁷ Experimental data related to high-κ gate applications of amorphous LaLuO₃ films are, however, still not available. In this letter, we present the results of a systematic study on the microstructural and electrical properties of amorphous LaLuO₃ thin films, deposited on silicon substrates by means of pulsed laser deposition (PLD).

LaLuO₃ films were deposited by PLD using a stoichiometric ceramic target. The target was made by milling a stoichiometric mixture of Lu₂O₃ (Alfa Aesar, 99.99%) and La₂O₃ (Alfa Aesar, 99.999%) powders with a molar ratio of 1:1. The ground powder was dried and then fired at 1300 °C in air for 12 h. After regrinding, the powder was pressed with a uniaxial press (3 tons). The pellets were then sintered at 1500 °C in air for 10 h. To increase the density of the target material it was subsequently sintered at 1600 °C in air for 12 h.

RCA cleaned p- and n-type (100) Si wafers cut in 1 × 1 cm² pieces were used as substrates. Here it is important to remark that after RCA cleaning a 1–1.5 nm thick SiO₂ layer is expected on the Si surface. The deposition process took place at a temperature of 450 °C in a 2 × 10⁻³ mbar O₂ ambient. Layers with thicknesses varying from 6 to 60 nm were grown, as determined by x-ray reflectometry (XRR). After deposition, the films were investigated with respect to their composition, surface morphology, and thermal stability by means of Rutherford backscattering spectrometry (RBS), atomic force microscopy (AFM), and x-ray diffraction (XRD). A combination of internal photoemission (IPE) and photoconductivity (PC) measurements was employed to determine the band gap width of the oxide and the conduction and valence band (CB, VB) offsets at the Si/high-κ interface.⁶

For the electrical characterization of the films, capacitor stacks were prepared. 70 nm thick Pt top contacts with an area of 245 × 245 μm² were deposited by electron beam evaporation through a shadow mask. The Ohmic backside contact was made by deposition of 120 nm Al followed by a forming gas annealing (90% N₂+10% H₂) at 450 °C for 10 min. This treatment improved the backside Al contact and the interface between the high-κ oxide and the silicon. The capacitor stacks were investigated using an impedance ana-
lyzer (HP 4192A) for capacitance-voltage (C-V) curves and a semiconductor parameter analyzer (HP 4155B) for current-voltage (I-V) measurements.

The RBS measurements (not shown) reveal a stoichiometry close to the nominal composition of LaLuO₃, presenting a La:Lu ratio of about 1:1.1. From the correlation between the RBS (atomic coverage per unit area) and the XRR data (physical thickness), a density of the films of ≈88% of the single crystalline density is deduced.

Additionally, the LaLuO₃ films exhibit a smooth surface morphology, as determined by AFM over a 2 × 2 μm² scan area (not shown). Films with thicknesses up to 12 nm show a root mean square surface roughness <0.2 nm and a peak-to-valley roughness of about 1.5 nm. The roughness of even thicker layers (from 20 to 60 nm) does not exceed 0.4 and 2.5 nm, respectively.

In order to study the thermal stability of the LaLuO₃, rapid thermal annealing in ultrapure N₂ (99.999%) was performed at atmospheric pressure and temperatures between 700 and 1200 °C for 10 s. Figure 1 shows the XRD patterns for 11 nm thick films as a function of temperature. The LaLuO₃ remains amorphous up to 1000 °C. At 1100 °C crystallization is observed as indicated by the arrows. The peaks seen in the spectrum do not fit to the available data for crystalline La₂O₃, Lu₂O₃, LaLuO₃, or related silicates and therefore more investigation is needed to identify the crystalline phases present after annealing at 1100 °C. The crystallization temperature is comparable to that observed for rare earth scandate (GdScO₃ and DyScO₃) films, also grown by the PLD technique, however, significantly higher than that observed for HfO₂ films.

IPE and PC experiments were performed using MOS capacitors fabricated by evaporation of semitransparent (15 nm thick) Au electrodes of 0.5 mm² area using an experimental arrangement described earlier. The quantum yield of IPE or PC was defined as the photocurrent, normalized to the incident photon flux. The square root of this quantity is not shown in Fig. 2. By extrapolating the yield to zero, the PC threshold of $E_{g}(ox) = 5.2 ± 0.1$ eV is found. There is also a 0.7 eV wide “tail” of subthreshold PC consistent with earlier observations for rare earth scandate insulators. The IPE of electrons from the VB of Si into the oxide CB is observed under positive metal bias. The threshold $\Phi_w$ was determined using the $Y^{1/3.-h\nu}$ plot as illustrated in the inset in Fig. 2. Extrapolation to zero yield results in the threshold value of $\Phi_w = 3.2 ± 0.1$ eV. Assignment of the spectral threshold observed in the curve measured with the metal biased negatively at $\Phi = 3.2 ± 0.1$ eV is not straightforward because the photocurrent might arise both from hole IPE from the CB of Si into the oxide VB and from electron IPE from the Au electrode. Nevertheless, the hole IPE interpretation seems to be the feasible one because the oxide band gap width calculated using the measured IPE barrier values as $E_s(ox) = \Phi_w + \Phi_h - E_g(Si) = 5.3$ eV (see inset in Fig. 2) coincides, within the measurement accuracy of ±0.1 eV, with the earlier indicated PC result $E_s(ox) = 5.2$ eV. Should this interpretation be correct, both the CB and VB offsets at the (100)Si/LaLuO₃ interface are equal to 2.1 eV, i.e., both are sufficiently high to prevent the injection of holes and electrons into the oxide.
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section between the CET axis and the linear fit at 2.6 nm indicates the existence of a lower-\(\kappa\) interfacial layer between the LaLuO\(_3\) film and the Si substrate. A detailed investigation on the formation of such a layer and its reduction in thickness is currently in progress.

In summary, we have demonstrated that amorphous lanthanum lutetium oxide is a promising alternative high-\(\kappa\) gate dielectric candidate. Thin films deposited on Si substrates present a high thermal stability up to 1000 °C, which fulfills the requirement for complementary MOS processing. Additionally, a large band gap width (5.2±0.1 eV) and symmetrical band offsets (2.1 eV) were observed for this material. The electrical characterization reveals good \(C-V\) behavior, a low leakage current density, and a \(\kappa\) value of about 32, which is larger than those previously determined for other alternative high-\(\kappa\) amorphous oxides (\(\kappa=22–23\)).

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