



# Effects of aluminum doping on lanthanum oxide gate dielectric films

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## ARTICLE INFO

### Article history:

Received 5 October 2010

Received in revised form

18 May 2011

Accepted 22 June 2011

### Keywords:

Interface properties

Lanthanum oxide

Aluminum doping

## ABSTRACT

This work reports a novel method for improving the electrical properties of lanthanum gate oxide ( $\text{La}_2\text{O}_3$ ) by using aluminum doping and rapid thermal annealing (RTA) techniques. In the bulk of the Al-doped  $\text{La}_2\text{O}_3$  film together with 600 °C RTA, we found that the aluminum atoms were incorporated into the oxide network and the film was transformed into lanthanum aluminate complex oxide. At the interface, a thin  $\text{Al}_2\text{O}_3$  layer was formed. This interfacial  $\text{Al}_2\text{O}_3$  layer suppressed the out-diffusion of substrate Si, the formation of interfacial silicate layer and silicide bonds. These effects resulted in a significant reduction on the bulk and interface trap densities and hence the gate leakage current.

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## 1. Introduction

To maintain a proper control over the channel current with a small gate bias, high-dielectric constant (high- $k$ ) gate dielectric must be used for nanoscale CMOS devices [1–4]. Hafnium-based materials have already been used in the state-of-the-art CMOS circuit fabrication [5–7]. However, the hafnium-based dielectric will no longer be suitable for sub-nanometer EOT (equivalent oxide thickness) thick gate dielectric required in the coming technology nodes. Rare-earth (RE) lanthanum oxide ( $\text{La}_2\text{O}_3$ ), having a  $k$  value of about 27, has been considered as one of the promising candidates for this kind of application. However, there are still many issues, such as hygroscopic nature, poor interface properties, and poor thermal instability, etc., need to be resolved before this kind of materials can be brought into the actual CMOS process [8–10]. A possible solution is to use certain alloy forms or complex oxides where the desirable electrical properties can be tailored by compromising the material properties of different oxides [10–15]. The incorporation of other atoms into an elemental  $\text{La}_2\text{O}_3$  may have several sequences. It may modify the modular volume of the material and results in a different optical dielectric constant and optical band gap to the elemental ones [3,10–12]. Some impurities such as N and Si can disrupt the oxide network and suppress the formation of crystalline phases [4]. Addition of a more electro-negative transition or rare-earth metal such as Ti and Sc to the  $\text{La}_2\text{O}_3$  can increase the metal-oxygen ionicity [4,12]. It changes the

conduction band structure and produces a higher  $k$  value. Meanwhile it was reported that doping of trace amount of foreign atoms may not change the electronic structure of host dielectric but still have significant impacts on some of the electrical and material properties [16,17]. For example, it was found recently that doping of nitrogen atoms can improve some of the electrical and material properties of  $\text{La}_2\text{O}_3$  [17].  $\text{LaAlO}_3$  was found to have excellent properties for gate oxide applications [19,20]. It is stable at high-temperature processing. However, the interface  $\text{LaAlO}_3/\text{Si}$  is also unstable. It was found that the  $\text{SiO}_2$  and  $\text{LaAl}$  silicates can be formed at the interface when the film was processed at 800 °C or above [20]. Meanwhile, the bulk type of  $\text{LaAlO}_3$  film is not a good option as the  $k$  value (close to the hafnium oxide) is not large enough for sub-nanometer EOT requirement [18]. This work attempts to improve the material and electrical properties of  $\text{La}_2\text{O}_3$  by incorporation of trace amount aluminum atoms into the film. The bonding structures of the Al atoms together with other atoms in the bulk and at the interface are studied in details. The electrical properties of the Al-doped films were characterized by using current–voltage ( $I$ – $V$ ) and capacitance–voltage ( $C$ – $V$ ) measurements.

## 2. Experimental

Lanthanum oxide ( $\text{La}_2\text{O}_3$ ) films of about 10 nm thick were deposited on  $\langle 100 \rangle$  n-type silicon substrates using electron beam evaporation technique [8]. A trace amount of aluminum atoms were then introduced into the as-deposited films by using plasma immersion ion implantation (PIII) [16,17]. The energy and the dose of the implantation were 2 keV and  $2.5 \times 10^{15}/\text{cm}^2$ , respectively.

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The energy was so chosen that Al ions would not be implanted into the silicon substrate. The samples were then rapid thermal annealed (RTA) in nitrogen ambient at a temperature ranging from 400 to 600 °C for 2–5 min. For electrical measurement, an aluminum layer was thermally evaporated and lithographically patterned. The diameter of the gate electrode was 100 or 300  $\mu\text{m}$ . Post-metal annealing (PMA) was conducted in nitrogen ambient at 300 °C for 25 min. The bonding structures of the dielectric films were investigated using Physical Electronics PHI 5600 X-ray photoelectron spectroscopy (XPS). Monochromatic  $\text{AlK}\alpha$  with excitation energy of 1486.6 eV was used as the X-ray source. The current–voltage (I–V) and capacitance–voltage (C–V) characteristics of the samples were measured using Keithley 236 source-measurement units and Keithley 590 C–V measurement system, respectively. All the electrical measurements were conducted in a dark and electromagnetic shielded vacuum chamber.

### 3. Results and discussion

Fig. 1 depicts the La 3d XPS spectra at different depths for pure and Al-implanted  $\text{La}_2\text{O}_3$  films. For pure  $\text{La}_2\text{O}_3$  sample, the bulk La 3d<sub>2/3</sub> has a double peak structure with main peak energy of 851.4 eV and satellite energy of 855.2 eV. This double peak structure is due to the unpaired *f* electrons which enable a shake-up process for charge transferring from O (2*p*) to La (4*f*) [17]. In this sample, the doublet shifted slightly to higher energy side and the peak were broadened because of the present of silicate phases. At the  $\text{La}_2\text{O}_3/\text{Si}$

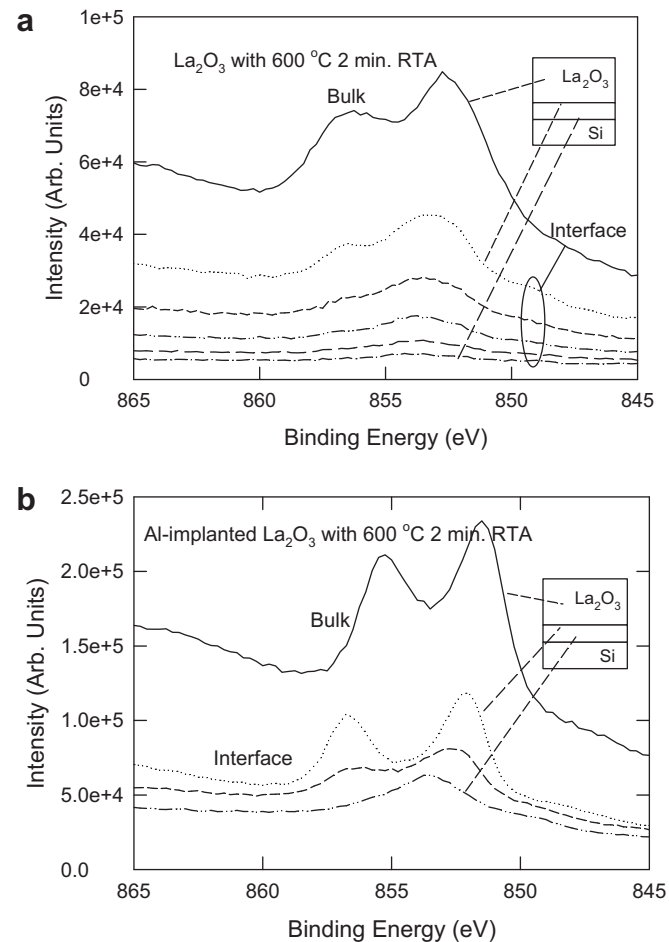


Fig. 1. Typical XPS spectra of La 3d at bulk and at the dielectric/Si interface for: (a)  $\text{La}_2\text{O}_3$ , and (b)  $\text{La}_2\text{O}_3$  with Al doping.

interface, about 10 nm from the surface, the La 3d<sub>2/3</sub> peak shifted to the even higher energy side because of more La–O–Si bonding. Different to the La–O–La bonding, the electron cloud on O moved closer to the Si side in the La–O–Si bonding because the Si atom has larger electronegativity than La atom. This effect resulted in the higher La 3d<sub>2/3</sub> binding energy in silicates. Since the electronegativity of Al atom is only slightly smaller than the Si ones, for the same reason, La ion in La–O–Al bonding should also have a larger binding energy. As shown in Fig. 1(b), the energies for the La 3d<sub>2/3</sub> doublet in the Al-implanted sample are slightly larger and their values are 852.7 eV and 856.2 eV, respectively. At the interface, the intensity of the La 3d in Al-implanted sample reduced greatly and the peak energy of La 3d increased gradually from 851.4 eV in the bulk to about 853.5 at the interface.

Fig. 2 shows the O 2*p* XPS at bulk and at interface for  $\text{La}_2\text{O}_3$  samples with and without Al implantation. In sample without Al implantation, the bulk O 2*p* peak appears as a double peak feature with a main peak at around 528.5 eV and a satellite peak at about 530.5 eV. The 528.5 eV peak can be attributed to the La–O bonding and the 530.5 eV peak should be due to the La–O–Si bonding [16,17]. At the interface, the O 2*p* peak spreads from 529 eV to 533 eV. This peak should be raised from the random mixing of La–O (528.5 eV) and Si–O (533 eV) bonds [16,17]. That is, both interface and bulk have significant amount of silicate phases. In sample with

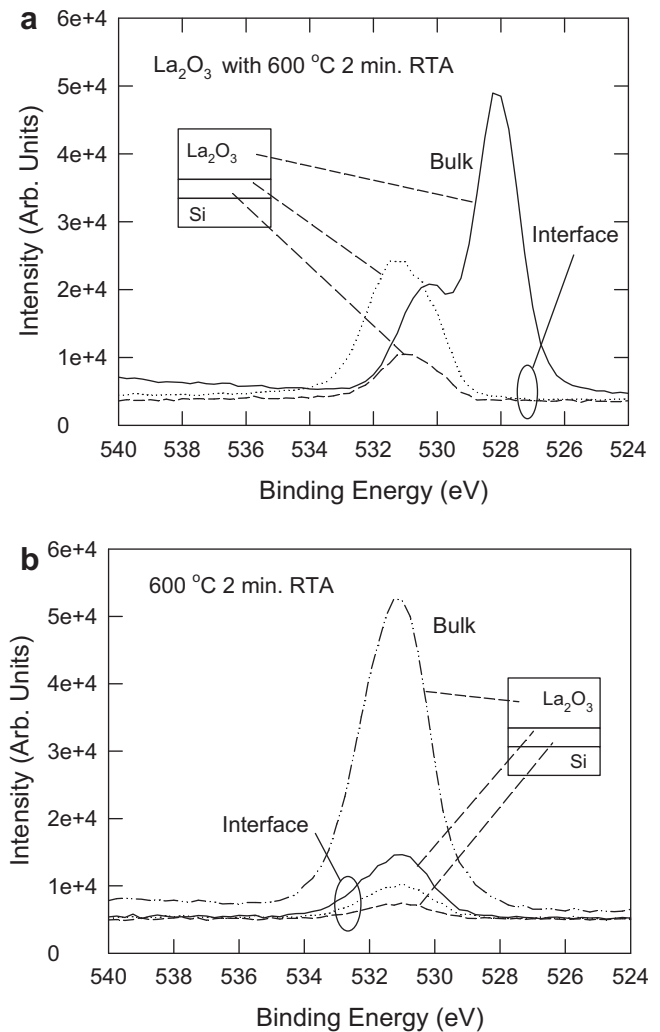


Fig. 2. O 2*p* photoelectron spectra at the dielectric/silicon interface for: (a)  $\text{La}_2\text{O}_3$ , and (b)  $\text{La}_2\text{O}_3$  with Al doping.

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