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Band gap, band offsets and dielectric constant improvement by addition of yttrium into lanthanum aluminate

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ABSTRACT

We studied the effects of adding yttrium (Y) in bulk lanthanum aluminate ($LaAlO_3$ or LAO) by investigating the quaternary compound oxide, lanthanum yttrium aluminum oxide $La_{0.3}Y_{0.7}AlO_3$ (LYAO), on silicon (Si). It is found that the inclusion of Y to LAO increases the band gap by ~0.9 eV without compromising the dielectric constant. The enhancement in the band gap results in larger band offsets in LYAO and we also observe a decrease in leakage current at low voltage accumulation bias for Al/LYAO/Si as compared to Al/LAO/Si. In addition, the interface trap density of the Al/LYAO/Si structure remains comparable to that of Al/LAO/Si. Our findings show that LYAO is an attractive high dielectric constant material for use in next-generation low standby power devices.

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1. Introduction

In order to meet the stringent requirements for next-generation complementary metal–oxide–semiconductor (CMOS) devices, many complex high dielectric constant (high-k) oxides have been engineered [1–3]. These dielectrics often involve the use of rare earth materials because of their high dielectric constant values and good thermodynamic stability with silicon (Si). However, these materials often have band gap width of about 6 eV, even after the addition of aluminum (Al) that is known to increase the band gap [4]. Larger band gap oxides are necessary to provide good stability against electrical breakdown and sufficiently large band offsets are required to reduce leakage current. This is crucial for CMOS devices that require low standby power consumption. In addition, the requirement of a large band gap in excess of 6 eV will be important in the use of high-k oxides as the inter-poly dielectric in floating-gate memory architectures or as the blocking oxide in charge-trapping devices for flash memory applications [5]. This is because very low leakage (~10^{-8} A/cm^2 at 1 V) is required for typical charge retention specifications and this becomes increasingly difficult with feature size scaling for equivalent oxide thickness (EOT) values down to 0.35 nm. Although oxides with larger band gap widths, such as aluminum oxide (~8 eV) can exhibit low leakage current density (as low as 4.3×10^{-8} A/cm^2 at 1 V), they tend to possess lower dielectric constants (~8 to 11) [5,6]. A high dielectric constant is required for EOT scaling in CMOS devices and for increasing the gate coupling factor in floating-gate memory devices [7,8]. Rare earth aluminates such as lanthanum aluminate ($LaAlO_3$ or LAO), are known to have high permittivity of about 30 (crystalline), but suffer from smaller band gap widths (~6 eV). It is therefore beneficial to engineer a material with both large band gap and dielectric constant.

An interesting ab initio calculation reveals that the alloy of LAO and yttrium aluminate ($YAlO_3$ or YAO) exhibits desirable properties for use as next-generation gate dielectrics [9]. The study shows that at compositions in the range of $0.2 < x < 0.4$ for the resultant lanthanum yttrium aluminate ($La_xY_{1-x}AlO_3$) structure, one can obtain reasonably high values for both band gap and dielectric constant. The large band gap is primarily due to the presence of YAO because yttrium introduces states 0.1 eV above the delocalized *s* and *p* states of the conduction band and causing little change to the large alumina band gap [10]. However at room temperature, YAO is not stable as a rhombohedral structure, which degrades the overall dielectric constant [9]. By doping it with lanthanum, this structure can be stabilized thus enhancing its dielectric constant. This theoretical prediction thus holds great promises for achieving a large band gap oxide with reasonable dielectric constant. Our study demonstrates the improvement in band gap width upon

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addition of yttrium while future work is still needed to show its impact on dielectric constant since the films in this study are amorphous. In addition, this quaternary LYAO compound is expected to reduce remote phonon scattering effects (leading to better channel carrier mobility) and also to form a good interface with Si due to the presence of the rare earth oxides. Despite the desirable characteristics predicted by the theoretical work in Ref. [9], there has yet to be a detailed experimental verification of the predicted beneficial improvements of LYAO over LAO.

In this article, we investigated the physical and electronic properties when yttrium is added by comparing bulk $La_{0.3}Y_{0.7}AlO_3$ (or LYAO) with LAO films. In the first part, the electronic properties are investigated, namely band gap and band offsets with Si. The interfacial layer thicknesses are measured using transmission electron microscopy (TEM). In the second part, electrical measurements are carried out to determine the EOT, interface trap density, and leakage current density of Al/LYAO/Si capacitors. Lastly, the dielectric constant of the LYAO film is extracted based on the measured EOT and the actual physical oxide thickness is determined using TEM.

2. Experimental details

In this study, the deposition of the LYAO films (using a compound oxide sputtering target of 99.9% purity) is carried out in an Anelva L3325FH multi-target sputtering system with radio frequency power of 100 W under a background pressure of $6.7–10 \times 10^{-5}$ Pa. The sputtering rate is 2.8 nm/min. The n-type Si substrates are moderately doped at ~7×10^{15} cm^{-3}. The Si substrates underwent the standard RCA etch process, followed by a final dip in 10% hydrofluoric acid (HF) for 25 s. An optical ellipsometer is used to measure the thickness of the deposited oxide by fixing the refractive index at 1.84. The refractive index of LYAO is derived through estimation by the Bruggeman effective medium approximation [11]. Capacitor structures with different LYAO thicknesses of 10.5 nm, 14 nm and 19 nm, as determined from ellipsometry, are fabricated. The capacitor dot structures are formed by thermal evaporation of front Al contacts through shadow masks, and blanket Al deposition for the back contact. All capacitor structures underwent a post-metallization anneal (PMA) in forming gas ambience of 10% hydrogen and 90% nitrogen to improve the metal contact properties. The radius of the capacitor dots is measured to be about 230 μm using optical microscopy. The physical and electronic properties of the LYAO films are characterized using high-resolution transmission electron microscopy (HR-TEM) and X-ray photoelectron spectroscopy (XPS), respectively. The HR-TEM sample is prepared using a focused ion beam system (FEI Nova Nanolab 600i) and characterized using a Philips CM300 TEM system under an accelerating voltage of 300 kV. XPS is performed in a VG ESCALAB 220i-XL system equipped with a monochromatic Al Kα (1486.6 eV) source and a concentric hemispherical energy analyzer. A magnetic immersion lens is used to maximize the photoelectron signal. Quantitative XPS analysis is firstly performed using a Shirley background subtraction before performing a least-square-error fit using a mixture of Gaussian (80%) and Lorentzian (20%) line shapes. Care is taken to ensure reasonable values for all the full-width-at-half-maximum of the fitted components. The relative atomic concentrations are then calculated after taking into account the instrument transmission function together with the Scofield photoionization sensitivity factor. All core-level peaks are referenced by assigning the adventitious carbon peak to 285 eV. Electrical measurements, such

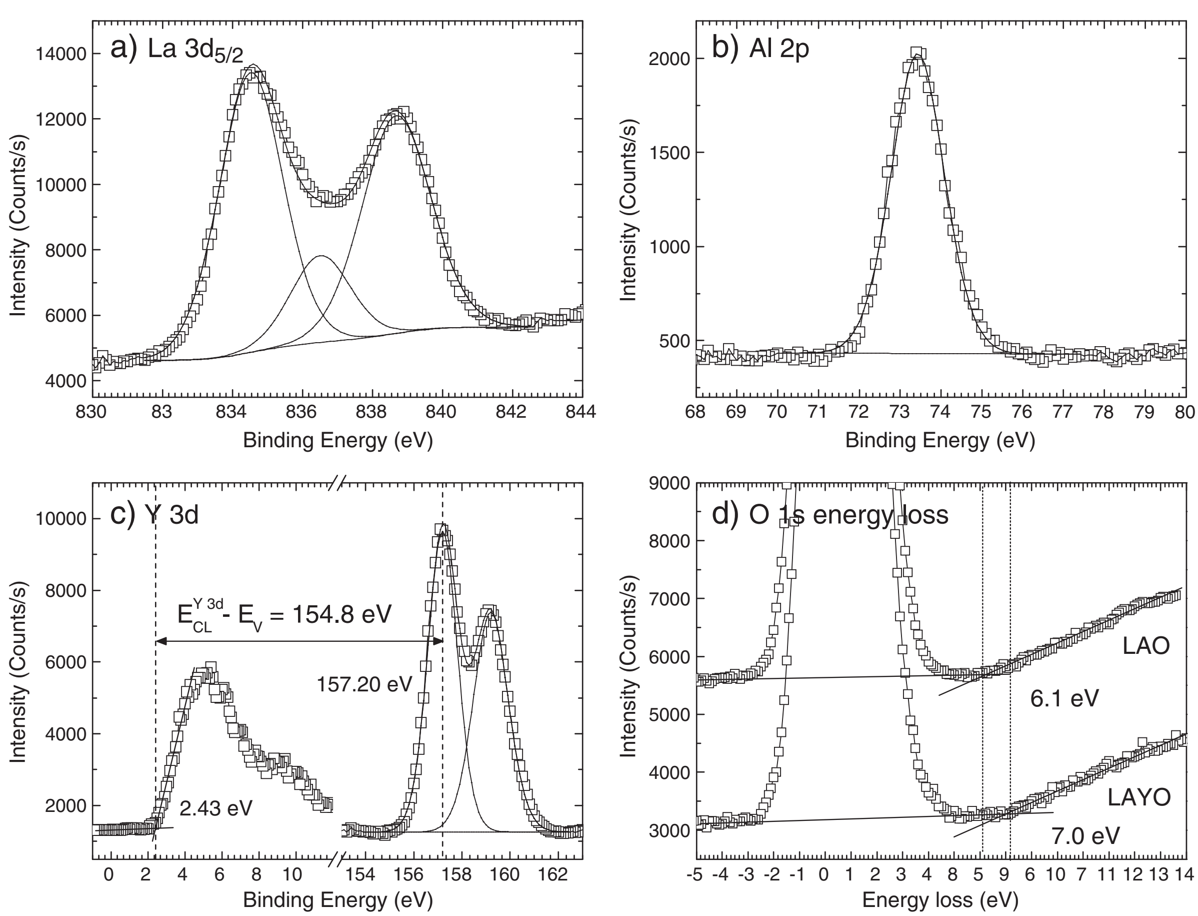

Fig. 1. XPS spectra for (a) La $3d_{5/2}$, (b) Al 2p core level peaks and (c) Y 3d core-level peak to valence band maximum separation for a 13 nm thick LYAO film on Si substrate. (d) Energy loss spectrum of O 1s for bulk LYAO and LAO films in the measurement of band gap.

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