# LaAlO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub> Gate Insulator for Amorphous InGaZnO TFTs to Suppress Permittivity Scattering of Lanthanum Oxide

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### ABSTRACT

Although lanthanum oxide has a high dielectric constant, it is not suitable as a gate insulator for TFTs due to its hygroscopic property. To suppress this, we propose a fabrication of a LaAlO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub> gate insulator by aluminum doping and adding Al<sub>2</sub>O<sub>3</sub> barrier on lanthanum oxide.

#### **1** INTRODUCTION

Amorphous indium gallium zinc oxide (a-IGZO) thin-film transistors (TFTs) have been widely used because of its high field-effect mobility, good uniformity for wide area and low processing temperature [1]. In conventional thin film transistors,  $SiO_2$  or  $SiN_x$  have been used as gate insulators but they could not induce the high current and low threshold voltages due to their low dielectric constant [2]. Therefore, it would be a promising way that use lanthanum oxide which has high dielectric constant as gate insulator to induce the high performance of a-IGZO TFTs.

However, fabrication of lanthanum oxide film by solution process has limitation as a gate insulator because of permittivity scattering is occurred by its hygroscopic property. It has been reported that RE-based ternary oxide (e.g. HfLaO, TaLaO and HoTiO<sub>3</sub>) as gate dielectric can further improve the TFT performance by suppressing the moisture absorption of the RE binary oxide [4]. In addition, nearly all metal oxides except Al<sub>2</sub>O<sub>3</sub> are crystallized during deposition or thermal annealing process [5]. Therefore, Al<sub>2</sub>O<sub>3</sub> is proper material to suppress hygroscopic property of lanthanum oxide film.

In this paper, two methods are proposed to use lanthanum oxide as gate insulator film for a-IGZO TFTs. The first one is suppressing crystallization of lanthanum film by properly mixing La<sub>2</sub>O<sub>3</sub> solution with Al<sub>2</sub>O<sub>3</sub> solution to make LaAlO<sub>x</sub> film. The second one is improving surface roughness and suppressing additional moisture absorption by capping LaAlO<sub>x</sub> film by thin Al<sub>2</sub>O<sub>3</sub> film.

## 2 EXPERIMENT

## 2.1 Precursor Solution Synthesis

For the deposition of LaAlO<sub>x</sub> and Al<sub>2</sub>O<sub>3</sub> gate dielectric layers by a solution process, precursor solutions for each gate dielectric were prepared by dissolving corresponding metallic precursors in a 2-methoxyethanol (2ME) solvent. For the LaAlO<sub>x</sub> solution, lanthanum nitrate hexahydrate (La(NO<sub>3</sub>)<sub>3</sub>·6H<sub>2</sub>O) and aluminum nitrate nonahydrate (Al(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O) was dissolved in 2-ME with a total

concentration of 0.5 M with various concentration of 1:0, 5:1, 3:1 and 1:1. For the Al<sub>2</sub>O<sub>3</sub> solution, 0.5 M of aluminum nitrate nonahydrate (Al(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O) was dissolved in 2ME. To prepare the 0.1 M a-IGZO solution, indium nitrate hydrate (In(NO<sub>3</sub>)<sub>3</sub>·xH<sub>2</sub>O), gallium nitrate hydrate (GaN<sub>3</sub>O<sub>9</sub>·xH<sub>2</sub>O), and zinc acetate dehydrate (Zn(CH<sub>3</sub>COO)<sub>2</sub>·2H<sub>2</sub>O) were dissolved in 2-ME and these components were mixed to achieve a 7:1:2 molar ratio of In:Ga:Zn. Then, each solution was stirred on hotplate at 75°C for 12 hours and filtered through 0.1 µm polytetrafluoroethylene (PTFE) syringe filters, respectively, before spin-coating.

#### 2.2 Device Fabrication

For the metal-insulator-metal (MIM) devices, the LaAlO<sub>x</sub> precursor solution was spin-coated on ITO glass substrate at 3000 rpm for 30 s and pre-annealed at 120°C for 10 mins to remove the solvent. Spin-coating and pre-annealing process was repeated for 3 times for enough thickness of LaAlO<sub>x</sub> films. Subsequently, Al<sub>2</sub>O<sub>3</sub> precursor solution was spin-coated on LaAlO<sub>x</sub> film at a speed of 5000 rpm for 30 s. After done of deposition of LaAlO<sub>x</sub> and Al<sub>2</sub>O<sub>3</sub> dielectrics, post annealing was performed on a hotplate at 300°C for 1h in ambient air. Then, a 70 nm aluminum metal contact was deposited by thermal evaporation system on dielectric films for evaluate MIM structures.

The bottom-gate structure of a-IGZO TFTs with  $LaAIO_x/AI_2O_3$  gate dielectrics were fabricated using solution process as shown in fig. 1. Synthesized  $LaAIO_x$  solution was spin-coated at 3000 rpm for 30 s and preannealed at 120°C for 10 mins to remove the solvent. Spin-coating and pre-annealing process was repeated for 3 times for enough thickness of  $LaAIO_x$  films. Subsequently,  $AI_2O_3$  precursor solution was spin-coated on  $LaAIO_x$  film at a speed of 5000 rpm for 30 s.



Fig. 1 Schematic of the a-IGZO TFT with LaAIO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub> gate insulator.



Fig. 2 AFM images of surface morphology of La<sub>2</sub>O<sub>3</sub> dielectric films at annealing temperature 300°C.



After deposition of LaAlO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub> dielectrics, post annealing was performed on hotplate at 300°C for 1h in ambient air. Then, synthesized IGZO solution was spincoated at 4000 rpm for 30 s. After spin-coating, UV-O<sub>3</sub> treatment was performed for 2 hours to improve the properties of the IGZO film and annealed at 350°C for 3 hours. Then, source/drain aluminum electrode of 70 nm were deposited by thermal evaporation system using shadow mask. The channel width (W) and length (L) were 1000 µm and 100 µm, respectively.

To investigate the properties of the dielectric films, the capacitance–voltage (C–V) measurements were carried out using an Agilent HP 4284A impedance analyzer. The surface roughness of dielectric films was measured by atomic force microscope (AFM; Park Systems, XE-100, non-contact mode). Leakage current density of LaAIO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub> dielectric films and electrical characteristic of a-IGZO TFT with LaAIO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub> gate dielectric were carried out using a semiconductor parameter analyzer Agilent 4145B, respectively.

#### 3 RESULTS AND DISCUSSION

The surface morphology image of  $La_2O_3$  at annealing temperature 300°C is as shown in fig. 2. It shows high surface roughness RMS of 21.15 nm because of crystallization peaks of  $La_2O_3$  films. These serious surface





problems indicate that La2O3 film cannot be used as gate insulators due to high leakage current induced by bad surface roughness. Fig. 3 shows surface roughness values of LaAIO<sub>x</sub> and LaAIO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub> dielectric films with various La:AI concentration. The surface roughness of LaAlO<sub>x</sub> was greatly improved at La:Al = 3:1 and 1:1. Also, the surface roughness values of LaAlOx films with Al2O barrier (LaAIO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub>) shows great surface roughness under 0.33 nm regardless La:Al ratio. Fig. 4(a) shows leakage current density of LaAIO<sub>x</sub>/AI<sub>2</sub>O<sub>3</sub> dielectric films. The LaAIO<sub>x</sub>/AI<sub>2</sub>O<sub>3</sub> film with La:AI = 5:1 ratio has lowest breakdown voltage and as the ratio of Al increase, the breakdown voltage occurs at higher voltage. Fig. 4(b) shows frequency dependent capacitance characteristic of LaAIO<sub>x</sub>/AI<sub>2</sub>O<sub>3</sub> films. La:AI = 5:1 ratio shows highest frequency-dependent capacitance characteristic and as the ratio of Al increase, frequency dependent capacitance were reduced. These results indicated that aluminum doping at lanthanum film and adding Al<sub>2</sub>O<sub>3</sub> barrier on lanthanum films are effective way to improve quality of solution processed lanthanum oxide dielectric film.



Fig. 5 (a) I-V transfer curve and (b) output characteristics of a-IGZO TFT with  $LaAIO_x/AI_2O_3$  gate insulator film.

Fig. 5(a) and (b) shows I-V transfer curve and output characteristics of a-IGZO TFT with LaAlO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub> gate insulator film, respectively. It shows great transfer curve and output characteristic as a TFT with a high on-off current ratio of  $2.45 \times 10^6$  under a low operating voltage V<sub>DS</sub> = 2V.

#### 4 CONCLUSIONS

In this study, LaAIO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub> gate insulator was proposed for a-IGZO TFTs. To suppress hygroscopic property of lanthanum oxide dielectric films at a annealing temperature of 300°C, aluminum doping and adding Al<sub>2</sub>O<sub>3</sub> barrier on lanthanum dielectric film were proposed. Through the AFM images, leakage current density and frequency-dependent capacitance characteristics, it was founded that proper aluminum doping and adding Al<sub>2</sub>O<sub>3</sub> barrier on LaAIO<sub>x</sub> film can significantly improve dielectric film characteristics. As a result, a high-k LaAIO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub> gate insulator was successfully integrated into a-IGZO TFTs. The LaAIO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub> a-IGZO TFTs shows a great output characteristic with a high on-off current ratio of  $2.45 \times 10^6$ under a low operating voltage V<sub>DS</sub> = 2V.

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