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Characterization of high- κ LaLuO₃ thin film grown on AlGaIn/GaN heterostructure by molecular beam deposition

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We report the study of high-dielectric-constant (high- κ) dielectric LaLuO₃ (LLO) thin film that is grown on AlGaIn/GaN heterostructure by molecular beam deposition (MBD). The physical properties of LLO on AlGaIn/GaN heterostructure have been investigated with atomic force microscopy, x-ray photoelectron spectroscopy, and TEM. It is revealed that the MBD-grown 16 nm-thick LLO film is polycrystalline with a thin (~ 2 nm) amorphous transition layer at the LLO/GaN interface. The bandgap of LLO is derived as 5.3 ± 0.04 eV from O1s energy loss spectrum. Capacitance-voltage (C-V) characteristics of a Ni-Au/LLO/III-nitride metal-insulator-semiconductor diode exhibit small frequency dispersion ($<2\%$) and reveal a high effective dielectric constant of ~ 28 for the LLO film. The LLO layer is shown to be effective in suppressing the reverse and forward leakage current in the MIS diode. In particular, the MIS diode forward current is reduced by 7 orders of magnitude at a forward bias of 1 V compared to a conventional Ni-Au/III-nitride Schottky diode.

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Wide bandgap group-III nitride (III-nitride) based electronic devices, especially in the form of high-electron-mobility transistors (HEMTs), are capable of delivering superior performance in high-frequency power amplifiers and high-voltage power switches.^{1–3} The conventional AlGaIn/GaN and InAlN/GaN HEMTs feature Schottky gates, resulting in relatively high reverse leakage current and a small forward turn-on voltage (V_{on}). By inserting a dielectric layer with large bandgap between the metal gate and semiconductor surface, ultralow gate leakage has been demonstrated in AlGaIn/GaN metal-insulator-semiconductor high-electron-mobility transistors (MISHEMTs). High-dielectric-constant (high- κ) materials are desirable for the gate dielectric as they enable tight channel control even with relatively large thickness. For AlGaIn/GaN MISHEMTs, Al₂O₃ ($\kappa \sim 9$)⁴ prepared by atomic layer deposition (ALD) has been predominately used. HfO₂ ($\kappa \sim 20$)⁵ has also been investigated although large leakage and poor stability have been reported. Recently, LaLuO₃ (LLO) has been studied and considered as a promising candidate for next-generation gate dielectric in Si,^{6–10} Ge-on-insulator (GeOI),¹¹ and GaAs (Ref. 12) metal-oxide-semiconductor field-effect transistors (MOSFETs) owing to its high dielectric constant (~ 30) and high crystallization temperature ($\sim 1000^\circ\text{C}$). La₂O₃ has also been used in AlGaIn/GaN MISHEMTs.¹³ Despite the high dielectric constant (~ 30) (Ref. 14) of La₂O₃, it is hygroscopic and could be leaky electrically. On the other hand, Lu₂O₃ has a larger bandgap, better hygroscopic immunity but lower crystallization temperature.¹⁵ Thus, by combining the benefits of La-based and Lu-based oxide, LLO thin film which also has the benefit of large electric-breakdown field of 2 MV/cm

(Ref. 16) is emerging as an attractive high-performance high- κ gate dielectric for AlGaIn/GaN MISHEMTs.

In this letter, we report the study of LLO thin film on AlGaIn/GaN heterostructure grown by molecular beam deposition (MBD) technique.⁶ It is revealed that the LLO film is initially amorphous at the LLO/III-nitride interface and then changes to polycrystalline after ~ 2 nm. The bandgap of the LLO film is 5.3 eV, determined from O1s energy loss spectrum obtained in x-ray photoelectron spectroscopy (XPS). Electrical characterizations performed on Ni-Au/LLO/AlGaIn/GaN MIS diode yield a dielectric constant of ~ 28 and illustrate effective suppression of gate leakage.

The AlGaIn/GaN heterostructure used in this work consists of a 1.8 μm GaN buffer, a ~ 18 nm undoped Al_{0.26}Ga_{0.74}N barrier, and a 2 nm GaN cap grown by metal organic chemical vapor deposition on (111) silicon substrate. LLO film was deposited on AlGaIn/GaN heterostructure by MBD at 450 $^\circ\text{C}$, and the growth details can be found in Lopes *et al.* (2007).¹⁰ Post-deposition annealing (PDA) was carried out at 400 $^\circ\text{C}$ in oxygen for 10 min, in order to compensate the possible oxygen vacancies in the film⁸ and to suppress moisture absorption, since it is considered that healing the oxygen vacancies in the film can effectively suppress moisture absorption.¹⁷ For the characterization of electrical properties, LLO MIS diode was fabricated. Ohmic contact region was opened by selectively etching LLO using BCl₃/Ar plasma in an inductively coupled plasma reactive ion etching (ICP-RIE) system,¹¹ followed by e-beam deposition of Ti/Al/Ni/Au (20 nm/150 nm/50 nm/80 nm). Then, the ohmic contact was annealed at 850 $^\circ\text{C}$ for 35 s. Then, Ni/Au (20 nm/240 nm) Schottky metal was deposited on LLO through a shadow mask by e-beam evaporation, followed by annealing at 350 $^\circ\text{C}$ in nitrogen ambient for 5 min to improve the interface between Ni/Au and LLO. The diameter of the circular Schottky metal was measured as 196 μm .

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FIG. 1. (Color online) AFM image of the surface of the LLO film.

The surface morphology of LLO was characterized by atomic force microscopy (AFM) over a $2 \times 2 \mu\text{m}^2$ scan region, as shown in Fig. 1. The root mean square (rms) and average roughness of the 16 nm LLO film is 0.58 nm and 0.43 nm, indicating decent surface quality.

In XPS measurements, monochromatic Al $K\alpha$ x-ray source (1486.6 eV) was used. The binding energy (BE) measurement was calibrated by correcting C1s to be 285 eV. Fig. 2(a) shows the La3d core-level XPS spectrum of LLO. Two doublets of La3d_{3/2} (b, b') and La3d_{5/2} (a, a') are shown, in which the main peaks a, b are 834.6 eV and 851.3 eV while the shake-up satellites a', b' on the high energy side of the two main peaks are 838.8 eV and 855.7 eV, respectively. The shake-up satellites are generally found in rare earth oxide and are believed to result from a core hole with electron transferred from the O_{2p} valence band to an empty 4f orbital.^{18,19} Lu4d_{3/2} and Lu4d_{5/2} core peaks are determined to be 196.8 eV and 206.7 eV (not shown here). From La3d_{3/2}, Lu4d_{3/2} and O1s core peaks, the ratio of oxygen to metal atoms near LLO surface can be deduced as 2.55:1, compared to the nominal ratio of 1.5:1. The excess oxygen was introduced during PDA in oxygen ambient and considered to suppress degradation due to the hygroscopic feature of LLO.

The bandgap of LLO can be derived from O1s energy loss signal in XPS spectrum, based on the fact that photoelectrons experience inelastic loss before band-to-band (from valence band to conduction band) transition, which corresponds to the bandgap energy. The bandgap of dielectric film is generally defined as the intercept of the linear extrapolation of the leading edge to the background level.²⁰ As shown in Fig. 2(b), the bandgap of LLO is determined to be 5.3 eV, which is in good agreement with the internal photoemission and photoconductivity measurement results 5.2 eV.⁶

FIG. 2. (Color online) (a) La3d core-level and (b) O1s energy loss spectra of LLO.

FIG. 3. (a) Cross sectional TEM micrograph of the 16-nm-thick LLO thin film on AlGaN/GaN heterostructure. (b) Higher magnification image for illustration of a thin amorphous LLO layer adjacent to GaN. (c) Metal atomic (Lu/La) ratios at 5 locations in the vertical direction of the LLO film from EDX profiles.

For transmission electron microscopy (TEM) study of the LLO film, sample was prepared by conventional polishing procedure and ion milling. The kinetic energy of electron beam used in TEM was 200 keV. Energy dispersive x-ray (EDX) profiles were also recorded to study the atomic ratio of Lu to La in the film. Fig. 3(a) shows the cross sectional TEM micrograph of LLO film deposited on AlGaN/GaN heterostructure. The physical thickness of LLO film is determined to be 16 nm and the LLO film consists of a polycrystalline layer over an amorphous layer on GaN. Fig. 3(b) provides a magnified image of LLO adjacent to GaN, the irregular contrast indicating amorphous phase over a thin layer (2–4 nm) near LLO/GaN interface. The thin amorphous layer at the LLO/GaN interface is formed since the lattice mismatch between LLO and GaN increases the activation energy barrier to nucleating LLO crystalline. After the growth of the thin amorphous LLO layer, the mismatch is relaxed so that polycrystalline layer start to grow.²¹ The thin amorphous layer is effective to suppress vertical leakage current in this work. The atomic ratio of Lu to La at 5 locations vertically across the LLO film was measured by EDX and was plotted in Fig. 3(c). It is found that the Lu/La ratio is considerably large as 1.74:1 near the surface (within 4 nm), while being uniform below the surface. The loss of La atoms near the surface is due to La atoms being absorbed by moisture in air. In the region 4 nm below the surface, the Lu/La ratio varies from 1.02 to 1.13, close to the nominal ratio of 1:1.

Agilent B1505A semiconductor device analyzer was used for capacitance-voltage (C-V) and current-voltage (I-V) characterization. Figure 4 shows C-V characteristics of AlGaN/GaN Schottky diode and LLO/AlGaN/GaN MIS

FIG. 4. (Color online) C-V characteristics of AlGaIn/GaN Schottky and LLO/AlGaIn/GaN MIS diode.

diodes. Frequency dispersion of less than 2% in the two-dimensional electron gas (2DEG) accumulation region between 100 kHz and 1 kHz can be observed. The voltage where the capacitance is 90% of the plateau capacitance is chosen as the flat band voltage V_{FB} , at which 2DEG starts to accumulate at AlGaIn/GaN interface. The determined V_{FB} at 10 kHz is shifted from -1.9 V for Schottky diode to -3.2 V for MIS diode. Given the capacitance of Schottky and MIS diode is, respectively, 481 nF/cm² and 370 nF/cm² at 10 kHz, applying series capacitance equation in the 2DEG accumulation region $1/C_{MIS-diode} = 1/C_{Sch-diode} + 1/C_{LLO}$, the capacitance of the 16 nm thick LLO is determined to be 1603 nF/cm². Therefore, the dielectric constant of 16 nm thick LLO is deduced to be ~ 28 . As shown in Fig. 5(a), it is observed that LLO insulator can remarkably reduce leakage current density at negative ($V = -4$ V) and forward bias ($V = 1$ V) by 1 and 7 orders of magnitude, respectively. The resistivity of the LLO film at room temperature is estimated to be $\sim 10^5$ Ω -cm from the I-V characteristics measured between two circular Ni/Au contacts on the film. In addition, V_{on} of diode is increased from 0.9 V to 2.0 V after inserting LLO between Schottky metal and AlGaIn/GaN, where V_{on} is defined as intercept of the linear extrapolation of the leakage leading edge to the background level under linear scale, as shown in Fig. 5(b). Larger V_{on} leads to enlarged gate swing.

FIG. 5. (Color online) (a) I-V characteristics of AlGaIn/GaN Schottky and LLO/AlGaIn/GaN MIS diode. (b) Comparison of V_{on} between AlGaIn/GaN Schottky and LLO/AlGaIn/GaN MIS diode.

In conclusion, high- κ dielectric LLO deposited on AlGaIn/GaN have been investigated by employing AFM, XPS, TEM, and electrical characterization. The 16 nm thick LLO film exhibits polycrystalline layer over a thin amorphous layer on GaN. Large bandgap of 5.3 ± 0.04 eV is derived. The electrical characteristics of LLO MIS diode reveal high dielectric constant of ~ 28 , low frequency dispersion ($< 2\%$), and effective leakage suppression capability at both negative and forward bias. Therefore, LLO could be a promising high- κ candidate for AlGaIn/GaN MISHEMTs.

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