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Growth and characterization of β -Ga₂O₃ thin films by molecular beam epitaxy for deep-UV photodetectors

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The growth of high quality epitaxial beta-gallium oxide (β -Ga₂O₃) using a compound source by molecular beam epitaxy has been demonstrated on c-plane sapphire (Al₂O₃) substrates. The compound source provides oxidized gallium molecules in addition to oxygen when heated from an iridium crucible in a high temperature effusion cell enabling a lower heat of formation for the growth of Ga₂O₃, resulting in a more efficient growth process. This source also enabled the growth of crystalline β -Ga₂O₃ without the need for additional oxygen. The influence of the substrate temperatures on the crystal structure and quality, chemical bonding, surface morphology, and optical properties has been systematically evaluated by x-ray diffraction, scanning transmission electron microscopy, x-ray photoelectron spectroscopy, atomic force microscopy, spectroscopic ellipsometry, and UV-vis spectroscopy. Under optimized growth conditions, all films exhibited pure $\bar{2}01$ oriented β -Ga₂O₃ thin films with six-fold rotational symmetry when grown on a sapphire substrate. The thin films demonstrated significant absorption in the deep-ultraviolet (UV) region with an optical bandgap around 5.0 eV and a refractive index of 1.9. A deep-UV photodetector fabricated on the high quality β -Ga₂O₃ thin film exhibits high resistance and small dark current (4.25 nA) with expected photoresponse for 254 nm UV light irradiation suggesting that the material grown using the compound source is a potential candidate for deep-ultraviolet photodetectors. *Published by AIP Publishing.* [<http://dx.doi.org/10.1063/1.4985855>]

I. INTRODUCTION

Recently, wide bandgap semiconducting oxides including tin oxide (SnO₂), indium oxide (In₂O₃), and gallium oxide (Ga₂O₃) have attracted much interest for high-power electronic devices and ultraviolet optoelectronic applications.¹ The unique properties of beta-gallium oxide (β -Ga₂O₃) make it a promising material for next-generation deep-ultraviolet (DUV) photodetectors and high-power devices due to its excellent thermal and chemical stability. Because of its large direct bandgap of ~ 4.9 eV, a high breakdown electric field of 8 MV/cm is expected for electronic devices fabricated from such a material. Compared to other wide bandgap semiconductor substrates such as GaN and SiC, β -Ga₂O₃ is more suitable for mass production of single crystal β -Ga₂O₃ substrates using simple and low-cost melt-growth methods.^{2–5} The large bandgap of β -Ga₂O₃ also makes it transparent from the visible to UV wavelengths while providing tunable bandgaps when alloying with Al₂O₃ (Ref. 6) or In₂O₃.⁷ Moreover, its maximum bulk electron mobility is projected to be 300 cm²/V s resulting in a Baliga's figure of merit (BFOM) higher than that of 4H-SiC and GaN^{1,4,8} in power device application. In addition, by doping with Si or Sn, it shows a controlled electron concentration of 10¹⁶–10¹⁹ cm^{−3}.⁴ These attractive attributes of β -Ga₂O₃ make it an ideal candidate for potential

applications that include high temperature sensors, luminescent phosphors, antireflection coatings, and UV optoelectronics and low loss, high-voltage switching devices such as high-breakdown Schottky diodes.⁸ Important requirements for these applications include crystallinity, a high degree of purity, and controlled doping when thin films are grown. Some of the techniques that have been used to deposit epitaxial thin films of β -Ga₂O₃ include pulsed laser deposition,⁹ metalorganic chemical vapor phase epitaxy,¹⁰ and ozone or plasma-assisted molecular beam epitaxy (MBE).^{1,8}

MBE is a conventional technique to grow ultrapure epitaxial films since the ultrahigh vacuum environment, pure source materials, and growth processes allow for the control of the crystal quality. With plasma-assisted MBE (PAMBE), epitaxial films with lower unintended impurity and defects levels can be achieved making it suitable for the investigation of the unexplained conductive mechanism of the thin oxide films in particular the role of oxygen vacancies in a leakage mechanism. This growth technique also results in reproducible doping control with improvement in the crystalline quality.^{11,12} In a conventional PAMBE technique, plasma-activated oxygen flux is used to oxidize the evaporated gallium (Ga) flux on a heated substrate to form a layer of Ga₂O₃. Typically, oxygen-rich conditions are used to provide an

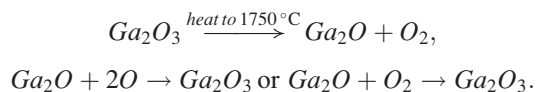
overpressure of oxygen to the surface for stoichiometry control and growth of β -Ga₂O₃, whereas Ga-rich growth conditions can be used for reducing Ga vacancy formation which act as compensating acceptors in β -Ga₂O₃.^{3,13} The reactions for the formation of the film growth and suboxide are: $2Ga + 3O \rightarrow Ga_2O_3$ and $2Ga + 1O \rightarrow Ga_2O$. According to Vogt and Bierwagen,³ the competition between accumulation of Ga₂O₃ and desorption of volatile Ga₂O determines the growth rate of the film. Besides, the growth rate depends on the availability of oxygen species which constitutes a big challenge for the growth of thick layers of β -Ga₂O₃ by PAMBE.³ To reduce the formation and desorption of Ga₂O, an increase of oxygen flux is required. Moreover, the introduction of a more reactive oxygen species such as ozone or plasma can reduce the desorption of Ga₂O and consequently increase the growth rate of the β -Ga₂O₃ film. However, there is a limitation on the partial pressure of oxygen that can be introduced into an MBE chamber, due to the risk of oxidation of filaments as well as exposure of the source materials in the effusion cells. To avoid this problem, the use of a Ga₂O₃ compound source would provide oxidized Ga species to the growing surface in the form of Ga₂O which would remove the requirement for a high oxygen pressure necessary to oxidize metallic Ga at the surface. An analysis of the heats of formation suggests that the use of Ga₂O molecules and oxygen would result in the growth of the crystalline Ga₂O₃ thin films at a much lower temperature and in comparison with using an elemental Ga source, the Ga₂O₃ compound source provides for a relatively higher growth rate.⁸ A similar technique was used for the growth of thick BaSnO₃ by MBE where a SnO₂ source has been used instead of elemental Sn to overcome the high overpressure and difficulty of incorporation of Sn into the ternary film.¹⁴ Prior reports on the growth of an oxide thin film using a Ga₂O₃ compound source with the films being amorphous^{15,16} were aimed for gate dielectric applications resulting in Fermi Level unpinning of GaAs surfaces. Therefore, an in-depth study of β -Ga₂O₃ thin film properties grown using a Ga₂O₃ compound source will be useful for future investigation.

In this paper, we explore the growth and properties of high quality β -Ga₂O₃ epitaxial thin films on c-plane sapphire (Al₂O₃) substrates as a function of MBE growth parameters using a Ga₂O₃ compound source instead of using an elemental Ga source. In addition, because of the presence of oxidized Ga (in the form of Ga₂O) and oxygen from the source, the growth of single phase β -Ga₂O₃ using the compound source without the use of any additional oxygen will be investigated. In this approach, the required oxygen need to form Ga₂O₃ will be obtained through the decomposition of the polycrystalline Ga₂O₃ source and transported to the substrate surface along with the Ga₂O molecules. To the best of our knowledge, there are no reports on the growth of crystalline β -Ga₂O₃ films without the use of an oxygen source by MBE. The properties of the thin Ga₂O₃ films will be demonstrated by fabricating and measuring the performance of a DUV photodetector.

II. EXPERIMENTAL

Heteroepitaxial β -Ga₂O₃ films were grown on single-side c-plane sapphire substrates by MBE. In the present

work, a compound polycrystalline Ga₂O₃ source (99.999%) evaporating from an iridium crucible in a high temperature effusion cell has been used to investigate the influence of the growth parameters including substrate temperature and pressure with activated oxygen generated through an RF plasma source. The heteroepitaxial growth of β -Ga₂O₃ using a compound source follows the following reactions:



The thermal decomposition of the Ga₂O₃ compound source provides both Ga₂O and oxygen molecules and it is expected that oxygen deficiency would be reduced when using this source to grow Ga₂O₃ thin films since there is no requirement to oxidize metallic Ga atoms. A set of growths were carried out at various substrate temperatures ranging from 500 to 850 °C. The crystallinity of the films was monitored *in-situ* using reflection high energy electron diffraction (RHEED) and confirmed by *ex-situ* x-ray diffraction measurements. The chamber pressure was 1×10^{-5} Torr during growth which was limited by the partial pressure of the Ga₂O and O₂ species resulting partially from the decomposition of the compound source. In addition, an oxygen flow rate of 1.2 sccm with the input RF plasma power of 300 W was used to supply additional oxygen. The detailed procedure to deposit β -Ga₂O₃ epilayers on a sapphire substrate has been reported in Ref. 8. β -Ga₂O₃ thin films were also deposited without the use of additional oxygen at a substrate temperature of 700 °C with the chamber pressure of 5×10^{-6} Torr to demonstrate that the compound source provides sufficient oxygen for the nucleation of a crystalline Ga₂O₃ film.

The crystal structure and preferred orientation of the thin films were investigated using X-ray diffraction measurements on a Rigaku SmartLab system with a Cu rotating anode. Four-bounce channel-cut Ge (220) crystals were used to obtain Cu K α radiation and eliminate vertical divergence. An aberration-corrected JEOL JEM-ARM200CF scanning transmission electron microscope (STEM) equipped with a cold field-emission source and an Oxford X-max 100TLE windowless SDD X-ray detector were utilized for crystal structure, microstructural and interfacial analysis at atomic resolutions. Epitaxial relationships between Ga₂O₃ and Al₂O₃ and the quality of interface were determined by both high-angle annular dark-field (HAADF) and annular bright-field (ABF) images acquired at 200 kV, with a collection semi angle from 90 to 170 mrad and 11 to 22 mrad, respectively.¹⁷ The cross-sectional sample was polished using an Allied High Tech MultiPrep polishing system and then thinned down to electron transparency (<100 nm) using a Fischione IonMill. The surface morphology and roughness were measured using a Park XE7 atomic force microscope (AFM). Chemical compositions and Ga oxidation states were investigated using x-ray photoelectron spectroscopy (XPS) with a Mg x-ray source. Refractive indices and film thicknesses were measured by variable angle spectroscopic ellipsometry (SE) using a J.A. Wollam, M-2000 ellipsometer

and optical transparency was determined using a Shimadzu UV-2501 UV-Vis spectrometer.

To determine the electrical properties of the deposited thin films, a metal-semiconductor-metal (MSM) Deep-Ultraviolet (DUV) photodetector was fabricated by depositing Au (100 nm)/Ti (50 nm) circular electrodes on the β -Ga₂O₃ thin film through a shadow mask by e-beam evaporation followed by 10 min thermal annealing at 450 °C in an Ar atmosphere.

The current-voltage (I-V) characteristics of the β -Ga₂O₃ thin film based photodetector were measured at room temperature by means of a Keysight B1500A semiconductor device analyzer. A low-pressure mercury (wavelength of 254 nm) UV lamp and a black light (wavelength of 365 nm) were used as the sources at a fixed distance of ~ 5 cm away from the devices to evaluate the UV photoresponse of β -Ga₂O₃ thin films.

III. RESULTS AND DISCUSSION

The out-of-plane 2θ - ω x-ray diffraction (XRD) scan of the Ga₂O₃ thin films grown at various substrate temperatures using the compound Ga₂O₃ source with a fixed oxygen pressure is shown in Fig. 1(a). The spectrum taken from the Ga₂O₃ layer deposited without any additional oxygen is also shown. At a growth temperature of 500 °C, the Ga₂O₃ film appears to be amorphous as evidenced from the XRD spectrum in which the only diffraction peaks are those from the sapphire substrate and a diffuse RHEED pattern during growth. At deposition temperatures ≥ 600 °C, the XRD spectra exhibit a set of diffraction peaks at 18.98°, 38.48°, 59.28°, and 82.30° which can be indexed to ($\bar{2}01$), ($\bar{4}02$), ($\bar{6}03$), and ($\bar{8}04$) planes of β -Ga₂O₃ indicating pure single ($\bar{2}01$) oriented thin films growth on the c-plane sapphire substrate. This is in agreement with previous reports where ($\bar{2}01$) oriented β -Ga₂O₃ thin films were grown on c-plane sapphire as well as a-plane sapphire and GaN substrates.^{8,18,19} During the growth of β -Ga₂O₃, the gallium atoms bond to the interfacial oxygen atom layer of the sapphire substrate resulting in optimized lattice matching between the ($\bar{2}01$) plane of β -Ga₂O₃ and the (0001) planes of the sapphire.¹² It is worth noting that there is a possibility of α -Ga₂O₃ phases to exist since the (0006) diffraction peak of α -Ga₂O₃ overlaps with the ($\bar{4}02$) diffraction peak of β -Ga₂O₃ located at 38.48° in the XRD spectrum. According to Oshima *et al.*²⁰ this can be clarified by examining the ratio of the intensity between the ($\bar{2}01$) and ($\bar{4}02$) peaks in the spectrum where the peak corresponding to ($\bar{4}02$) is expected to be higher than the peak corresponding to ($\bar{2}01$), if α -Ga₂O₃ was present. Typically, α -Ga₂O₃ would be observed at low growth temperatures.^{9,20} At 750 °C, the intensity ratio(($\bar{2}01$)/($\bar{4}02$)) was experimentally determined to be 2.17 but as growth temperature was decreased, this ratio gradually decreased to a value of 0.37 at 600 °C. Taking into account the Powder Cell 2.4 calculation (mentioned in Ref. 20), the ideal value of this ratio for a pure β -Ga₂O₃ phase is 2.2. While at higher growth temperatures this value is close to the ideal intensity ratio, the deviation to lower values when the growth temperature is reduced suggests a partial formation of the α -Ga₂O₃ phase.^{21,22}

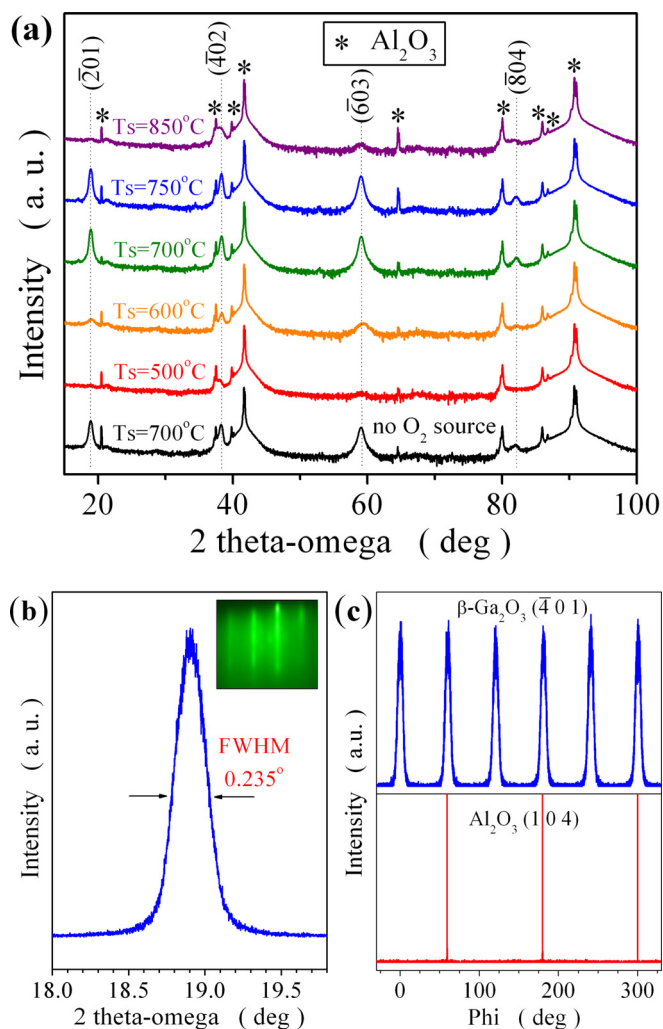


FIG. 1. (a) XRD 2theta-omega (2θ - ω) spectra of Ga₂O₃ thin films grown at various substrate temperatures on a sapphire substrate. Peaks marked as \square belong to sapphire (Al₂O₃) substrates; (b) High resolution X-ray rocking curve for the ($\bar{2}01$) plane of the β -Ga₂O₃ thin film with the inset showing the RHEED pattern of the β -Ga₂O₃ thin film grown at 700 °C substrate temperature; (c) XRD Φ -scan for the $\{401\}$ diffraction peak of β -Ga₂O₃ grown at 700 °C substrate temperature with the Φ -scan for the $\{104\}$ diffraction peak of the Al₂O₃ substrate.

An analysis of the spectra in Fig. 1(a) shows a strong dependency of the thickness and phase of the Ga₂O₃ films on growth temperatures above 600 °C. Higher substrate temperatures are required for the growth of stable single phase β -Ga₂O₃ due to insufficient surface adatom energy and mobility present at lower temperatures. With a growth temperature of 700–750 °C, the high intensity of the planes of β -Ga₂O₃ suggested that enough thermal energy is supplied to the impinging molecules to increase the surface mobility leading to a highly ordered ($\bar{2}01$) crystal orientation. The intensity of the ($\bar{2}01$) peak and higher order diffraction peaks of β -Ga₂O₃ decreased when the growth temperature was increased to 850 °C due to a decrease in film thickness caused by the reduction of the sticking coefficient of the volatile Ga₂O molecules.³ The decrease in film thickness at this elevated growth temperature was confirmed by spectroscopic ellipsometry measurements. High resolution XRD scans around the ($\bar{2}01$) plane of β -Ga₂O₃ were performed on the

films grown at 700 and 750 °C substrate temperatures. The corresponding full width half maxima (FWHM) are 0.235° and 0.233°, respectively, with Fig. 1(b) showing the high resolution ($\bar{2}01$) XRD peak for the film grown at 700 °C with the FWHM comparable to reported values.^{9,18} The crystallinity was further confirmed during growth by monitoring RHEED patterns exhibiting a streaky threefold reconstruction [Fig. 1(b) inset] indicative of epitaxial growth.

To determine the epitaxial relationship between β -Ga₂O₃ and the Al₂O₃ substrate, in-plane XRD Φ (phi) scan was measured for the $\{401\}$ planes of the β -Ga₂O₃ thin film and the $\{104\}$ planes of the Al₂O₃ substrate shown in Fig. 1(c) for growth at 700 °C. The Φ scan for the $\{401\}$ diffraction planes shows six peaks which are 60° apart from each other indicating the presence of a sixfold in-plane rotational symmetry for the β -Ga₂O₃ thin film. However, monoclinic β -Ga₂O₃ $\{401\}$ diffraction planes have twofold in-plane rotational symmetry and due to the threefold rotational symmetry of the c-plane sapphire (Al₂O₃) surface, it can be concluded that the originally twofold β -Ga₂O₃ films grew epitaxially in the three different directions at the same rates, resulting in the sixfold rotational symmetry.^{9,20} Also, shown in Fig. 1(a) is an XRD spectrum for Ga₂O₃ grown at 700 °C using the compound oxide source with no additional oxygen, suggesting that the decomposition of the compound oxide source provides sufficient oxygen necessary for the crystallization of the Ga₂O₃ layer. An examination of the XRD spectra indicates that the crystal quality of the layers grown with and without oxygen appears to be comparable.

Figure 2(a) shows the low-magnification HAADF image of the β -Ga₂O₃ thin film grown on an Al₂O₃ substrate at 700 °C. The magnified HAADF image [Fig. 2(b)] shows that the Ga₂O₃ thin film is single-crystal and the β phase of Ga₂O₃ has been further confirmed by the selected-area electron diffraction patterns of the Ga₂O₃ thin film along the $[010]$ axis shown in Fig. 2(d) indicating the epitaxial growth of the film with the orientation relationship of ($\bar{2}01$)

β -Ga₂O₃||(0001) Al₂O₃. The atomic-resolution HAADF and ABF (allows observation of oxygen atoms) images [Figs. 2(e) and 2(c)] are obtained simultaneously and show a sharp interface between Ga₂O₃ and Al₂O₃ without any precipitation. The energy dispersive spectroscopy (EDS) line scan profile of the yellow arrow line shown in Fig. 2(e) across the Ga₂O₃/Al₂O₃ interface was obtained to investigate the compositional distribution of the Ga₂O₃ thin film [Fig. 2(f)]. From the EDS line scan, along with the HAADF image, it can be seen that Ga atoms are uniformly distributed in the film and there is no noticeable phase separation or metal agglomerated areas that can be detected. Additionally, the analysis of the STEM images indicates no evidence of α -Ga₂O₃ for the film grown at 700 °C.

X-ray photoelectron spectroscopy (XPS) was employed to analyze the stoichiometry and chemical valence state (i.e., oxidation state) of β -Ga₂O₃ films deposited on the sapphire substrate using the compound Ga₂O₃ as the source material in the two growth schemes: with and without additional oxygen. The Ga 3d spectra shown in Figs. 3 and 4 were taken immediately after deposition and without exposure to air since the sample can be transferred to the spectroscopy chamber under UHV. In this way, any artifacts due to air exposure can be eliminated.

The deconvolution of XPS spectra was carried out by employing a Gaussian function on a Shirley background. From Fig. 3(a), it is clear that the measured Ga 3d peak can be fitted with a single peak at the binding energy (BE) of 19.89 eV assigned to Ga₂O₃ when grown at a substrate temperature of 750 °C using oxygen plasma. The additional peak observed at 23.61 eV represents the O 2s peak. While the growth of crystalline Ga₂O₃ can be accomplished without the use of additional oxygen when using the compound source, there is some evidence of metallic Ga in the film as evidenced from the Ga 3d spectrum in Fig. 4(a). This spectrum is composed of two Ga-related peaks apart from the O 2s peak (centered at 23.22 eV) which are attributed to Ga₂O₃

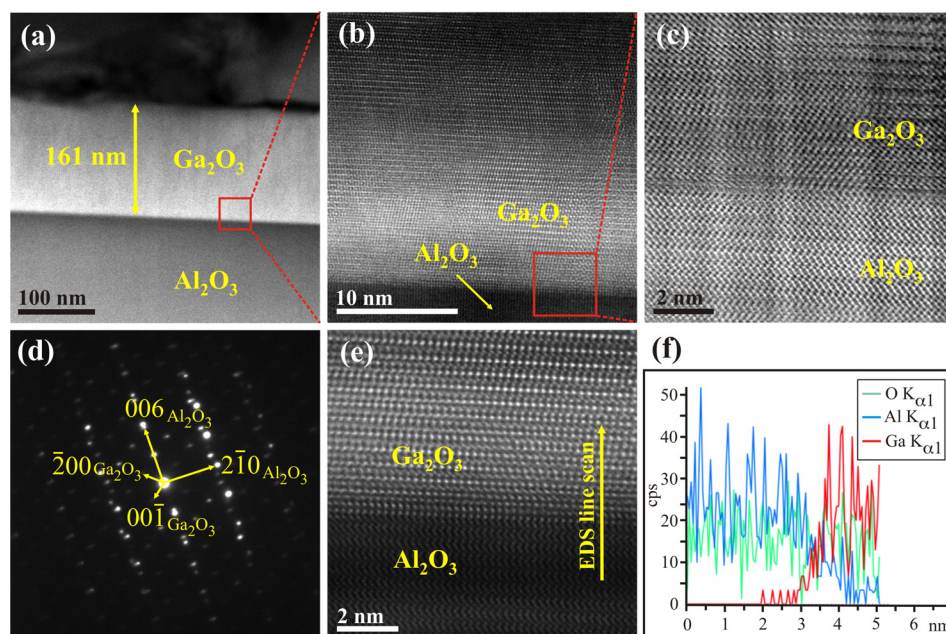


FIG. 2. (a) Low magnification cross-sectional STEM-HAADF image of the Ga₂O₃/Al₂O₃ heterostructure; (b) STEM-HAADF image of the Ga₂O₃/Al₂O₃ interface in high magnification; (c) Atomic-resolution HAADF images of the Ga₂O₃/Al₂O₃ interface taken from (b) as marked by a red small square; (d) The SAED pattern of the Ga₂O₃ thin film; (e) STEM-HAADF image of the Ga₂O₃/Al₂O₃ interface; (f) The EDS line scan profile of the yellow arrow line shown in (e).

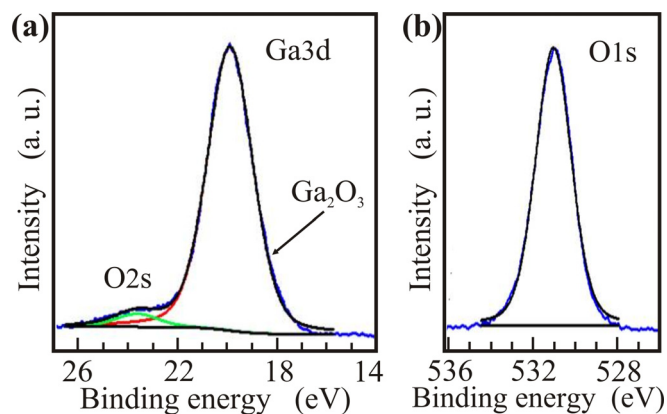


FIG. 3. (a) Ga 3d core level high resolution spectrum of β -Ga₂O₃ thin films grown with 750 °C with additional oxygen which shifts towards higher BE compared to metallic Ga represents the Ga³⁺ state. (b) O 1s core level spectra of Ga₂O₃ thin films. Blue lines denote experimental data and fitting results represented by black lines. Both fitted to a single peak indicates stoichiometric composition.

recorded at 20.05 eV and metallic Ga at 18.44 eV. In both cases, a positive shifting in BE, caused by the electronic charge redistribution around the constituent atoms of the crystal, is observed for Ga₂O₃ films indicative of a Ga³⁺ oxidation state. Hence, changes in Ga chemical bonding lead to a shifting in the BE which can be utilized to extract information about the oxidation state of Ga-oxide phases in the grown films.^{15,23} Based on the XPS analysis [Fig. 3(a)], it can be said that it is possible to grow stoichiometric Ga₂O₃ thin films on a sapphire substrate employing a Ga₂O₃ compound source under optimum growth condition which includes the use of the oxygen source because in this growth scheme, oxidized Ga is provided through the use of the compound Ga₂O₃ source. On the other hand, when growing using the compound source without additional oxygen, the films were observed to be slightly Ga rich as expected [Fig. 4(a)] as there is insufficient oxygen to sustain the $Ga_2O + 2O \rightarrow Ga_2O_3$ reaction resulting in an extra peak assigned to Ga-Ga bonding.

Figures 3(b) and 4(b) show core level photoelectron spectra of O 1s measured from the grown films. These

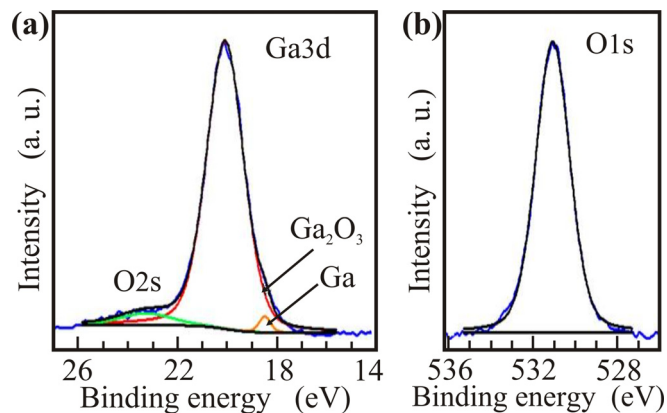


FIG. 4. (a) Ga 3d core level high resolution spectrum of β -Ga₂O₃ thin films grown without an oxygen source which is a convolution of two peaks represents the Ga³⁺ state and Ga⁰ state. (b) O 1s core level spectra of Ga₂O₃ thin films which are fitted to a single peak. Blue lines denote experimental data and fitting results represented by black lines.

spectra consist of a single peak indicative of films which are composed of Ga with a predominantly single oxidation state.

In order to visualize the growth temperature influence on surface morphologies and roughness of β -Ga₂O₃ thin films grown by a Ga₂O₃ compound source, AFM micrographs were taken on various samples grown at different substrate temperatures shown in Fig. 5. AFM images were obtained over $5 \times 5 \mu\text{m}^2$ scanning area at 1.5 Hz scanning frequency. The surfaces exhibit a flat granular shape morphology with relatively low root mean square (rms) roughness (Table I) without noticeable cracks or discontinuities. Several authors reported a strong dependency of surface morphology on the growth temperature and oxygen partial pressure.^{8,9,24} Based only on a growth temperature variation analysis at a fixed oxygen pressure, the surface morphology did not change considerably in the samples, Figs. 5(a)–5(d). However, at the highest substrate temperatures (850 °C), the surface roughness decreased significantly. This can be understood in terms of an increased mobility of adatoms on the surface accompanied by a decrease of the film thickness and growth rate due to reduction of the sticking coefficient of Ga₂O molecules.³ The sample grown at the lowest temperature (500 °C) exhibited a lower roughness but lacked a good structural quality according to the XRD characterization shown in figure (Fig. 1).

In order to investigate the optical properties of the β -Ga₂O₃ thin film, UV-Vis measurements were carried out.

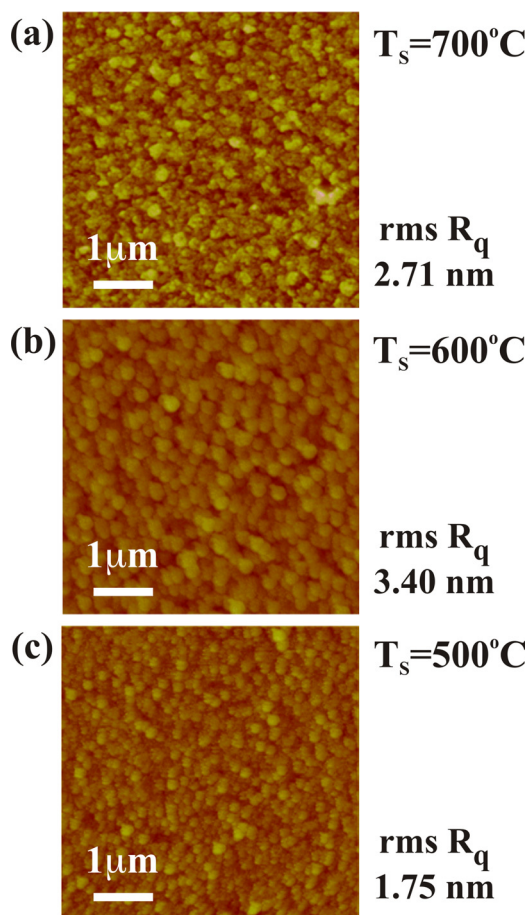


FIG. 5. $5 \times 5 \mu\text{m}^2$ surface AFM images of β -Ga₂O₃ epitaxial films grown at various substrate temperatures.

TABLE I. Material characteristics of Ga₂O₃ thin films grown at various substrate temperatures.

Substrate temperature (°C)	Bandgap (eV)	Thickness (nm)	Refractive index at 632 nm	RMS Roughness (nm)
850	4.98	15.89	1.79	0.88
750	5.03	150.5	1.89	3.07
700	5.02	161.0	1.91	2.71
600	5.05	33.75	1.90	3.40
500	5.10	26.23	1.92	1.75

Figure 6(a) shows a plot of the transmittance spectrum vs. wavelength of the film prepared at a substrate temperature of 700 °C showing a good optical transparency and exhibits clear fringes in the visible and UV regions. The amorphous Ga₂O₃ films (grown at 500 °C) exhibit (not shown in figure) increased transparency over the whole spectra except at the band edge where absorption of the incident radiation becomes more apparent.²⁴ This is a very important feature for specific applications such as antireflection and dielectric coatings of solar cells.²⁵ To formulate a quantitative structure-property relationship and to understand the microstructural effect on the optical properties, analysis of the transmission spectra was also performed. Since β -Ga₂O₃ is a direct band gap material^{9,26,27} the absorption spectra obey a power law of the form

$$(\alpha h\nu) = B(h\nu - E_g)^{1/2}, \quad (1)$$

where $h\nu$ is the incident photon energy, α is the absorption coefficient, B is the absorption edge width parameter, and E_g is the band gap. The optical absorption coefficient can be determined as²⁴

$$\alpha = \left[\frac{1}{t} \right] \ln \left[\frac{T}{(1-R)^2} \right], \quad (2)$$

where T is the transmittance, R is the reflectance, and t is the film thickness. A plot of $(\alpha h\nu)^2$ vs $h\nu$ shows (Fig. 4 inset) a

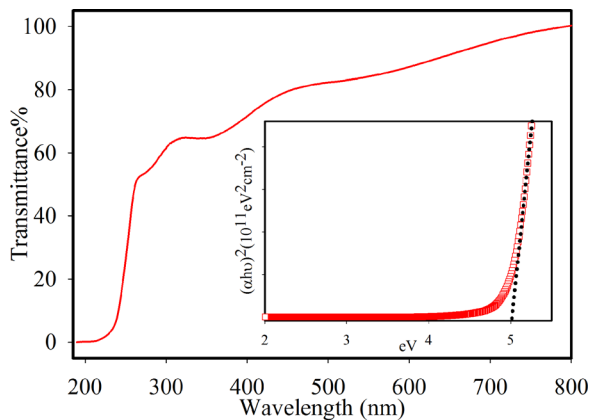


FIG. 6. Transmission spectrum of the Ga₂O₃ thin film grown at 700 °C and the inset is a plot of $(\alpha h\nu)^2$ vs photon energy at different substrate temperatures where α and $h\nu$ represent the absorption coefficient and photon energy, respectively. By extrapolating α to 0, the optical bandgap was obtained.

linear relationship in the high absorption region confirming a direct optical bandgap for the Ga₂O₃ films^{26,27} whose value could be found by extrapolating the linear region of the plot to $h\nu = 0$. The E_g variations for Ga₂O₃ films grown at various substrate temperatures are shown in Table I. The E_g values of the grown Ga₂O₃ films are around 5 eV with the amorphous film grown at 500 °C showing the highest E_g value of 5.1 eV. The obtained higher values of E_g at low temperature for Ga₂O₃ films may be due to the non-stoichiometric and/or amorphous nature which could contribute to disorder, leading to a slightly higher band gap measured for the films grown at 500 °C, in agreement with previous studies.²⁴ The calculated bandgap energies obtained in this study are comparable with previously reported values for β -Ga₂O₃.^{12,20,26,27}

The complex index of refraction and thickness of the films were measured by obtaining the ellipsometric parameters Ψ and Δ at room temperature for different wavelengths (200–1000 nm) and incident angles (65°–85° with 5° interval). An optical model based on regression analysis and the Mean Squared Error (MSE) were used to determine the refractive index, film thickness, and surface roughness. The details of the fitting and modelling could be found in our previous work.⁸ The measured film thicknesses and calculated refractive index (n) (at wavelength of 6328 Å) for β -Ga₂O₃ films are both compiled in Table I. The thickness and the modelled surface roughness of the films showed consistency with the measured results from a profilometer and AFM measurements, respectively. The resulting refractive indexes are closely related to the film growth temperature with an average value of around 1.90 for growths at temperatures <850 °C. The lowest value (1.79) obtained for the growth at 850 °C is probably due to the accuracy in measuring such a thin film decrease in film thickness. It is worth noting that the refractive indexes in this work are consistent with the previously reported literature for β -Ga₂O₃.^{12,20,23}

The room temperature current-voltage (I-V) characteristics of the DUV photodetector are shown in Fig. 7. In Fig. 7(a), it is clearly observed that with increasing bias voltage, current increases linearly under both dark and illuminated conditions suggesting that Au/Ti forms a good ohmic contact for Ga₂O₃ films. The possible reason behind this is the large surface states at the surface of Ga₂O₃ and hence, carriers can easily tunnel through the surface metal/oxide barrier.²⁸ The dark current was measured to be 4.25 nA at the bias voltage of 10 V shown in Fig. 7(b). When black light is used to illuminate the detector, the current increased to 56 nA which is not very significant compared to the dark current indicative of the non-sensitivity of β -Ga₂O₃ thin films to 405 nm wavelength radiation. Rather, this small increase in current with black light is probably due to the phonon response of the light and sample interaction.²⁰ Conversely, the detector shows a significant increase of current to a value of 5.6 μ A at the bias voltage of 10 V when illuminated under a UV lamp with 254 nm wavelength with the photo-to-dark current ratio of the detector >1000. This characteristic implies that under 254 nm light irradiation, the resistance of the β -Ga₂O₃ thin film based photodetector has decreased considerably.

To examine the current conduction mechanism of the MSM-DUV photodetector, the I-V characteristics were

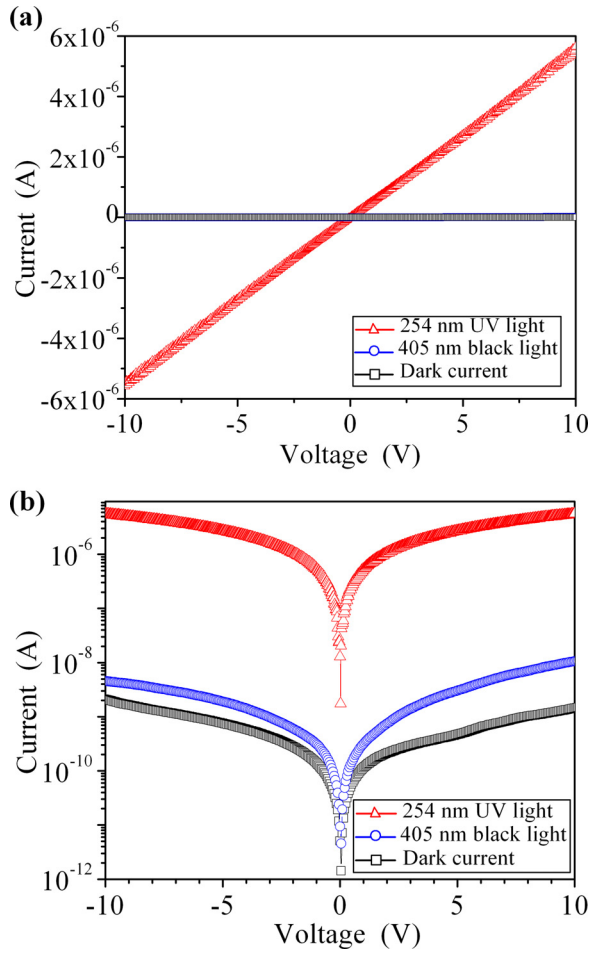


FIG. 7. The I-V characteristic curve of the β -Ga₂O₃ based DUV photodetector in the dark, under black light (wavelength of 405 nm), and under 254 nm light irradiation at room temperature where (a) and (b) plots represent the linear and logarithmic coordinate, respectively.

analyzed with various carrier injection methods to find the best fit such as Ohmic conduction ($I \propto V$), Thermionic emission ($\ln I \propto V$), Space charge limited conduction (SCLC) ($I \propto V^a$, where $a \geq 2$), Poole-Frenkel emission [$\ln(I/V) \propto V^{1/2}$], and Fowler-Nordheim tunneling [$I \propto V^2 \exp(-E_a/V)$, where E_a is the kinetic energy of the charge carriers].^{17,29–31} In this device, the conduction mechanisms were determined by fitting the I-V curves for both the negative and positive voltage regions depicted in Figs. 8(a) and 8(b). For 405 nm illumination and dark current, the space charge limited conduction (SCLC) mechanism is primarily responsible for current conduction between ± 2 to ± 10 V where most of the charge carriers are injected from the contact and follow Ohmic conduction in the small 0 to ± 2 V region.²⁹ However, during 254 nm UV illumination it was found that only Ohmic current dominates for the entire voltage range of 0 ± 10 V where most of the charge carriers are generated photoelectrons from β -Ga₂O₃.²⁹ This phenomenon suggests that β -Ga₂O₃ thin films can be potential candidates for high performance photodetector applications.

To evaluate the performance of the detector, the time-dependent photoresponse of the photodetector under 254 nm light illumination was determined by applying a light pulse at 1 V bias voltage with the results shown in Fig. 9(a)

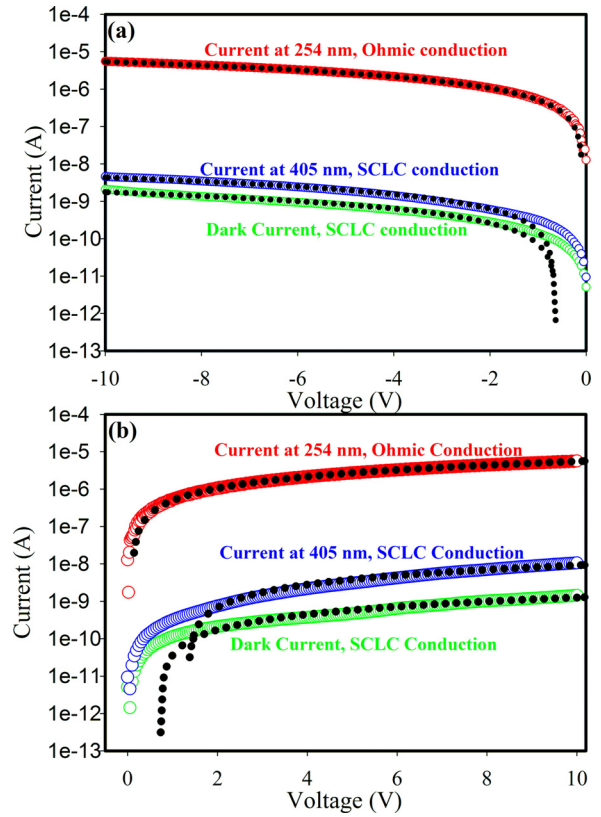


FIG. 8. Carrier injection mechanism of the β -Ga₂O₃ based DUV photodetector in the dark, under black light (wavelength of 405 nm), and under 254 nm light irradiation at room temperature for (a) negative voltage and (b) positive voltage.

exhibiting good reproducibility and high robustness. The photocurrent increases rapidly to a stable value of ~ 7.5 nA and then abruptly decreases to a value of ~ 0.16 nA when the 254 nm illumination is turned on and off, respectively. The approximate dark current (~ 0.16 nA) is low and favorable for practical detectors. Detailed comparative and quantitative analysis of the response and recovery time to 254 nm illumination was investigated by fitting the photoresponse curve with a biexponential relaxation equation given by the following formula:

$$I = I_o + Ae^{\frac{-t}{\tau_1}} + Be^{\frac{-t}{\tau_2}},$$

where I_o is the steady state photocurrent, t is the time, A and B are constants, and τ_1 and τ_2 are two relaxation time constants. Figure 9(b) shows excellent fitting of the photoresponse processes where τ_r is the time constant for the rising edge and τ_d is the time constant for the decay edge. Under 254 nm illumination, the current rise and decay sharply with rise time constant, τ_r of 6 s and the decay time constant, τ_d of 5 s as shown in Fig. 9(b) comparable to other published results.^{12,32–34}

IV. CONCLUSION

High quality epitaxial β -Ga₂O₃ thin films were grown on a (0001) Al₂O₃ substrate at various substrate temperatures using a Ga₂O₃ compound source by a MBE technique which provide oxidized Ga molecules from a high temperature

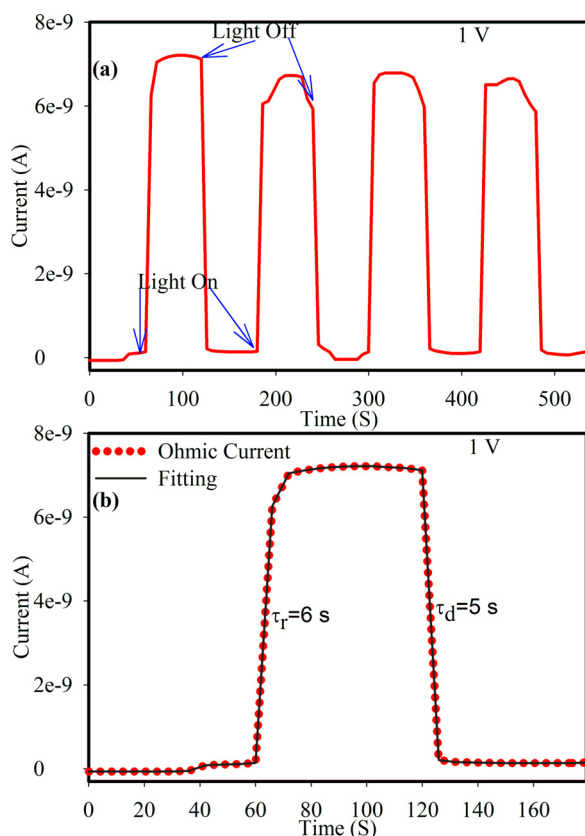


FIG. 9. (a) Time-dependent photo response of the β -Ga₂O₃ thin films photo-detector to 254 nm illumination. (b) Enlarged view of the rise/decay edges and the corresponding exponential fitting.

effusion cell. The effect of the substrate temperature effect on the structural and optical properties of the grown films was systematically investigated with pure phase ($\bar{2}01$) oriented β -Ga₂O₃ films obtained in an optimized temperature range of 700–750 °C. The use of a compound source for the first time to grow a ($\bar{2}01$) oriented β -Ga₂O₃ thin film without additional oxygen source was demonstrated. In-plane XRD Φ -scan revealed the presence of sixfold in-plane rotational symmetry with TEM investigations confirming high quality monoclinic thin films. The grown β -Ga₂O₃ thin films exhibit high transparency in the UV-visible region and were used to fabricate an ohmic type DUV photodetector with an MSM structure. The I-V characterization of the photodetector demonstrated high resistance as well as small dark current with high sensitivity to UV light with 254 nm wavelength. These experimental results suggest excellent structural quality and optical properties for the oxide films grown using polycrystalline Ga₂O₃ as the source material and combined with the I-V characteristics under UV light irradiation β -Ga₂O₃ thin films produced are shown to be a potential candidate for deep-ultraviolet photodetectors.

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