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Research Article

Structural, Optical, and Electrical Characterization of β-Ga_2O_3 Thin Films Grown by Plasma-Assisted Molecular Beam Epitaxy Suitable for UV Sensing

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β-Ga_2O_3 thin films were grown on c-plane sapphire substrates by plasma-assisted molecular beam epitaxy. The films were grown using an elemental gallium source and oxygen supplied by an RF plasma source. Reflection high-energy electron diffraction (RHEED) was used to monitor the surface quality in real time. Both in situ RHEED and ex situ X-ray diffraction confirmed the formation of single crystal β-phase films with excellent crystallinity on c-plane sapphire. Spectroscopic ellipsometry was used to determine the film thicknesses, giving values in the 11.6–18.8 nm range and the refractive index dispersion curves. UV-Vis transmittance measurements revealed that strong absorption of β-Ga_2O_3 starts at ~270 nm. Top metal contacts were deposited by thermal evaporation for I-V characterization, which has been carried out in dark, as well as under visible and UV light illumination. The optical and electrical measurements showed that the grown thin films of β-Ga_2O_3 are excellent candidates for deep-ultraviolet detection and sensing.

1. Introduction

Ultraviolet radiation (UV) covers the wavelengths shorter than 400 nm in the electromagnetic spectrum. The effect of UV radiation depends on the wavelength and can lead to some benefits as in the case of UV-A (400–320 nm) that stimulates photosynthesis, while exposure to other wavelengths can cause skin cancer (UV-B 320–280 nm) and be a risk factor for some eye diseases. The most energetic part of the UV spectrum, UV-C (280–10 nm) causes damage and degradation of organic materials [1–3]. As a result, the development of miniature, fast, highly sensitive, selective, stable, and robust UV detectors is the subject of intense studies [4].

Gallium oxide (Ga_2O_3) is a wide bandgap (~5 eV) semiconductor material with the β-phase being the most stable form. Ga_2O_3 thin films have been used as gas sensors, dielectric coatings for solar cells, and deep-ultraviolet (UV) transparent oxides, among others [5–11]. Wide bandgap semiconductors present advantages over the polymers currently used for UV detection because of their intrinsic visible blindness, temperature stability, and enhanced radiation hardness [12, 13]. A wide variety of techniques have been used to deposit or grow Ga_2O_3 thin films. Among them is molecular beam epitaxy (MBE) [14–18].

In this work, we report on the preparation and characterization of β-phase Ga_2O_3 thin films grown by plasma-assisted molecular beam epitaxy (PAMBE) at various growth conditions. The quality of the grown films was inferred from reflection high-energy electron diffraction (RHEED) and X-ray diffraction (XRD). Current-voltage (I-V) measurements were

(a) (b)

Figure 1: RHEED pattern of (a) c-plane sapphire substrate and (b) grown Ga_2O_3 film.

carried out in dark, as well as under visible and UV light illumination to study the possibility of using β-phase Ga_2O_3 in deep-UV sensors.

2. Materials and Methods

β-Ga_2O_3 thin films were grown on single-side polished c-plane sapphire substrates by PAMBE in an oxide MBE chamber. The substrates were degreased in acetone and thermally cleaned for 15 min at 850°C in the chamber followed by an oxygen plasma exposure for 30 minutes. A chamber base pressure of $<5\times10^{-10}$ Torr was obtained by a combination of turbomolecular and cryogenic pumps. During the growth, the oxide chamber cryopanels were cooled with liquid nitrogen. The films were grown using a well-known method from evaporated Ga and oxygen provided from a radio frequency (RF) plasma source equipped with an ion deflector [19]. The oxygen flow rate was varied from 1.0 to 1.5 sccm using RF plasma power of 200 and 300 W. The elemental evaporated Ga was provided from an effusion cell with a fixed temperature of 800°C (thermocouple temperature). The substrate temperature was varied from 650 to 750°C as measured using a thermocouple. All samples were grown for 60 minutes with the surface quality monitored in real time in situ using RHEED.

The as-grown samples were analyzed by XRD (Bede System diffractometer) to determine the crystalline quality of the films. The thickness and the optical constants of the films were determined by spectroscopic ellipsometry measurements using a J. A. Woollam M-2000 variable angle ellipsometer. Transmission spectra of the samples were measured by the UV-Vis spectrometer Shimadzu 2600. For electrical characterization, a group of samples with 70×70 μm Al contacts with spacing of 30 μm between them was prepared by thermal evaporation. I-V measurements of lateral current between two adjacent contacts were carried out in dark as well as under UV-C (254 nm), UV-A (365 nm), and visible light illumination (532, 650 nm) using a Keithley 4200 Semiconductor Characterization System.

3. Results and Discussion

Real-time RHEED was used to monitor the crystallinity and surface quality during the film growth. Figure 1(a) shows a RHEED pattern of pure c-plane sapphire substrate prior to growth presenting a diffuse background. After formation of the Ga_2O_3 film, the RHEED pattern exhibited a streaky threefold reconstruction, indicating high-degree of crystallinity in the grown films as is shown in Figure 1(b).

The film thicknesses were determined by Ellipsometry using the Cauchy model [20, 21]. The Cauchy model assumes that the refractive index n can be represented by a slowly varying function of the wavelength $n = A + B/\lambda^2 + C/\lambda^4$, where A, B, and C are fitting parameters that determine the index. The thicknesses were determined by fitting the experimental results only in the 300–1000 nm range, where the films are transparent. The obtained results for the thickness, roughness, and the refractive index at $\lambda = 632.8$ nm of the grown samples are shown in Table 1. The dependence of the film thickness on the substrate temperature is shown in Figure 2(a), while Figure 2(b) presents the dispersion curves of the refractive index.

The reduction of thickness at high temperature is a result of increased desorption of Ga_2O molecules on the growing surface whereas higher oxygen flux suppresses the desorption resulting in higher growth rates. For films grown at oxygen flux of 1.0 sccm, the refractive index shows a tendency to increase with the substrate temperature, while for films grown at flux of 1.5 sccm, the dependence of n on T is very weak.

XRD results confirmed the formation of the β-phase single crystal Ga_2O_3 on sapphire with excellent crystallinity. The X-ray diffractograms were measured in the 10–70° 2θ range. Figure 3 shows XRD spectra of films grown with an oxygen flux of 1.5 sccm and RF plasma power of 300 W at three different substrate temperatures, 650, 700, and 750°C. Three well-defined peaks at 18.9°, 38.4°, and 59.2° can be distinguished, corresponding to diffractions of (−201), (−402), and (−603) planes of β-Ga_2O_3. Besides the peaks associated to β-Ga_2O_3 and the substrate, no other significant peaks were found in the spectra, thus excluding the existence of another phase. The intensity of the XRD peaks shows a weak dependence on the growth temperature; higher intensity is observed at lower substrate temperature. Similar results were obtained for films grown at the same

Table 1: Thickness, roughness, and refractive index at $\lambda = 632.8$ nm obtained using the Cauchy model.

Growth temperature (°C)	O_2 flux (sccm)	Thickness (nm)	Roughness (nm)	n at 632.8 nm
650	1.0	13.1	0.65	1.864
700	1.0	12.1	0.78	1.868
750	1.0	11.6	1.02	1.876
650	1.5	18.8	0.54	1.875
700	1.5	17.1	0.36	1.879
750	1.5	16.7	0.48	1.873

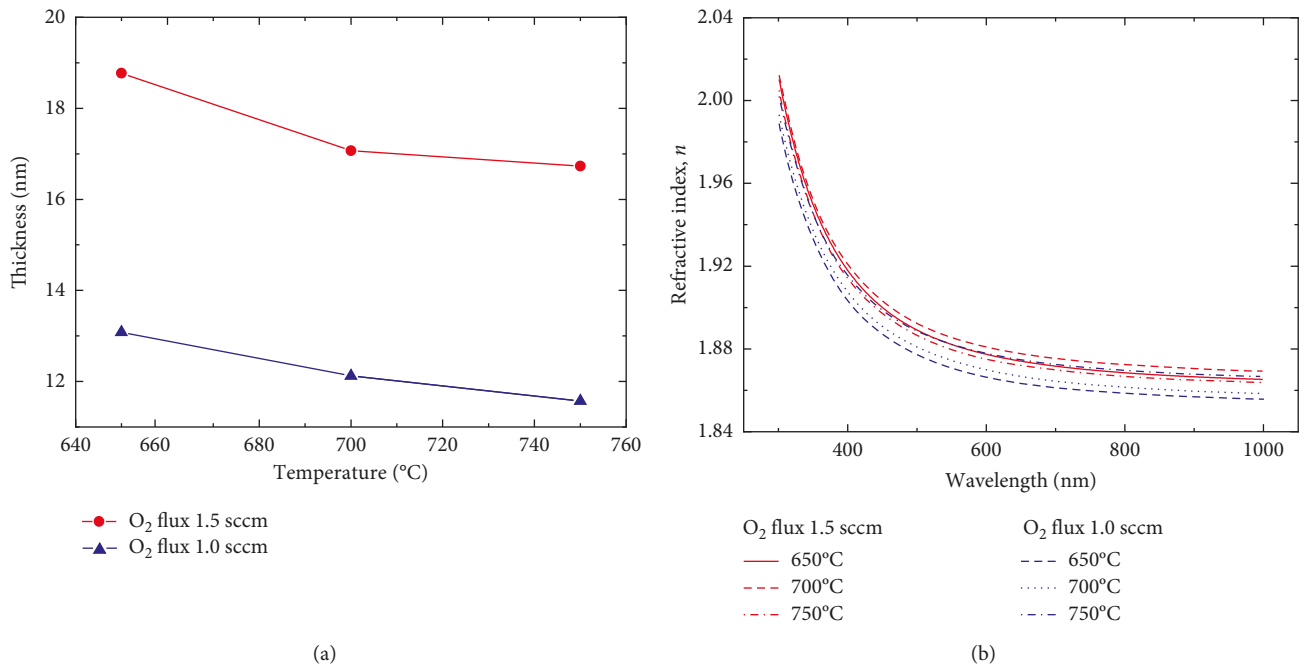

Figure 2: (a) Film thickness dependence on the substrate temperature at fluxes of 1.5 and 1.0 sccm; (b) dispersion curves of the refractive index.

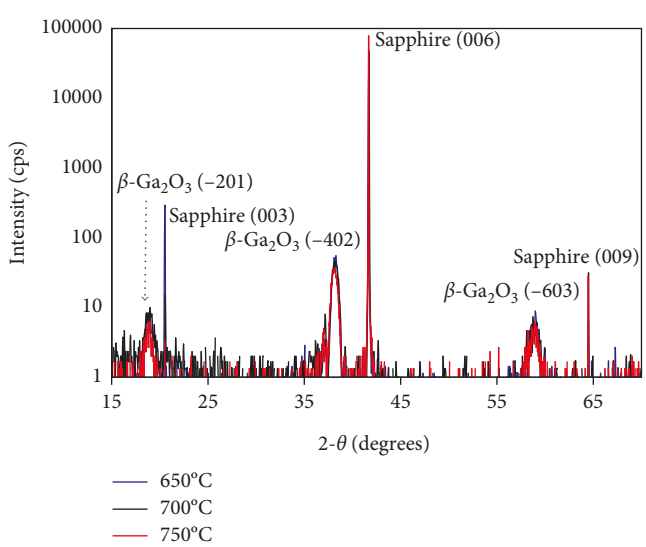

Figure 3: XRD spectra of β-Ga_2O_3 films grown on sapphire at power of 300 W and flux of 1.5 sccm.

plasma power and substrate temperatures and oxygen flux of 1.0 sccm.

High-resolution rocking curves of the peak at 38.4° corresponding to the <−402> plane of β-Ga_2O_3 confirmed the relation between the peak intensity and the temperature. Higher intensities were measured from films grown at 650°C (Figure 4), a result that is in agreement with the thickness dependence on the temperature (Figure 2).

The UV-Vis spectroscopy results revealed that strong absorption of β-Ga_2O_3 starts at ~270 nm, which corresponds to UV-C radiation. Figure 5 shows the UV-Vis spectra of sapphire substrate as well as of β-Ga_2O_3 thin films grown with RF plasma power of 200 and 300 W at two temperatures, 700 and 750°C. Sapphire is a highly transparent material and exhibits higher transmittance compared to the sapphire/β-Ga_2O_3 samples. The samples grown with an RF plasma power of 200 W exhibit a lower transmittance than that of samples grown at 300 W, which does not depend on the deposition temperature. In contrast, the increase of the substrate temperature for samples grown at 300 W leads to higher transmittance. The transmittance values shown in Figure 5 are

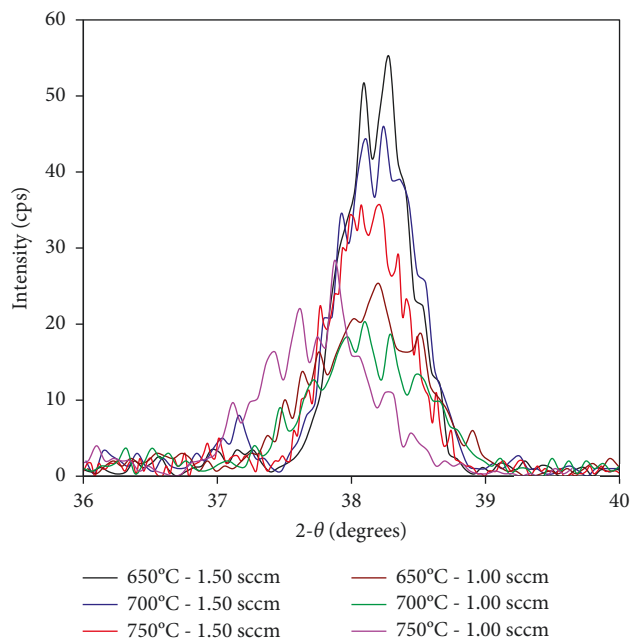

Figure 4: High-resolution rocking curves corresponding to <−402> plane of β-Ga_2O_3 of films grown at 650, 700, and 750°C at fluxes of 1.0 and 1.5 sccm.

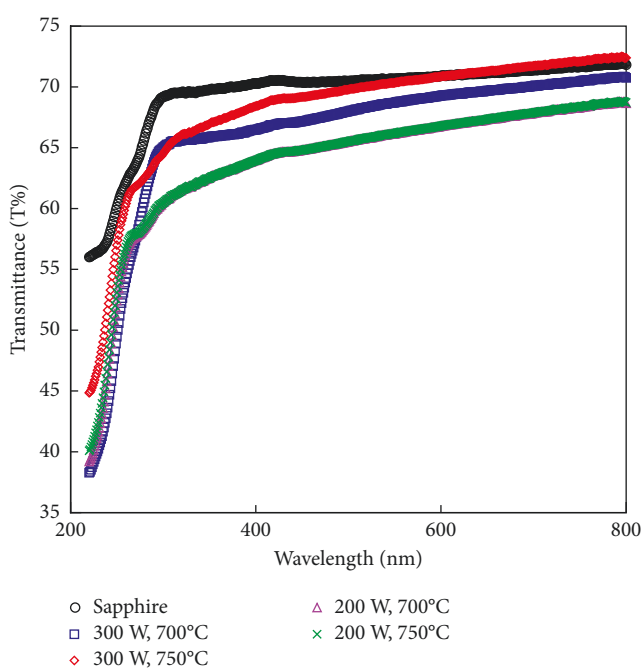

Figure 5: UV-Vis spectra of sapphire substrate (circles) and β-Ga_2O_3 thin films grown at 700°C with RF plasma power of 200 and 300 W (squares and triangles) and at 750°C at the same powers (diamonds and crosses).

lower than previously reported [14, 22, 23] because of light scattering by the unpolished side of the sapphire substrate.

From the UV-Vis results, the optical bandgaps of the films were determined using the power law for direct bandgap semiconductors [22–26]:

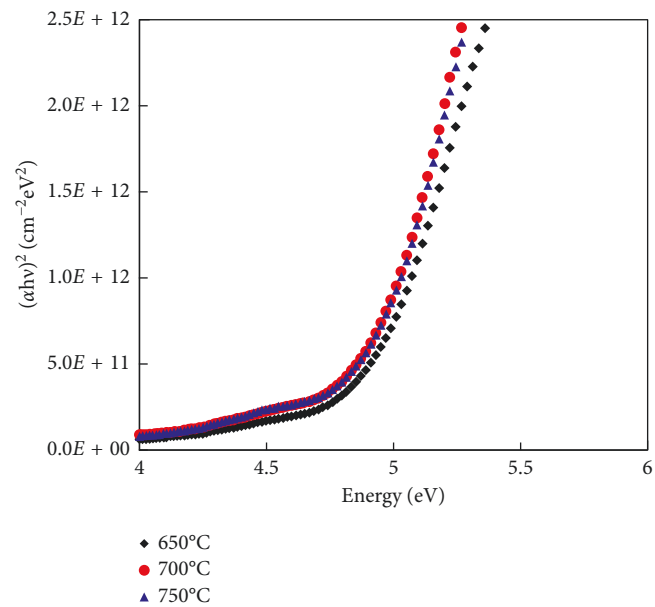

Figure 6: $(\alpha h\nu)^2$ versus $h\nu$ dependence of Ga_2O_3 thin films grown at different substrate temperatures and a 1.5 sccm O_2 flux.

Table 2: Optical bandgaps of β-Ga_2O_3 thin films grown at 300 W at different temperatures and oxygen fluxes.

Growth temperature (°C)	O_2 flux (sccm)	E_g (eV)
650	1.0	4.84
700	1.0	4.73
750	1.0	4.88
650	1.5	4.82
700	1.5	4.84
750	1.5	4.88

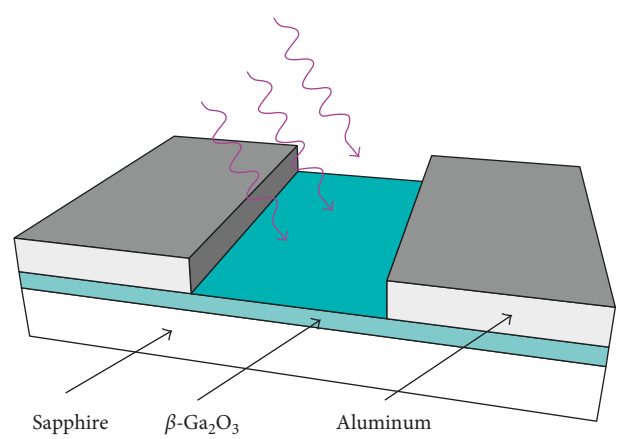

Figure 7: Schematic representation of the structure used in the I-V measurements.

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

where α is the absorption coefficient, $h\nu$ is the photon energy, and E_g is the bandgap. The optical absorption coefficients were calculated using the ellipsometrically determined film thicknesses. E_g was estimated graphically by regression analysis of

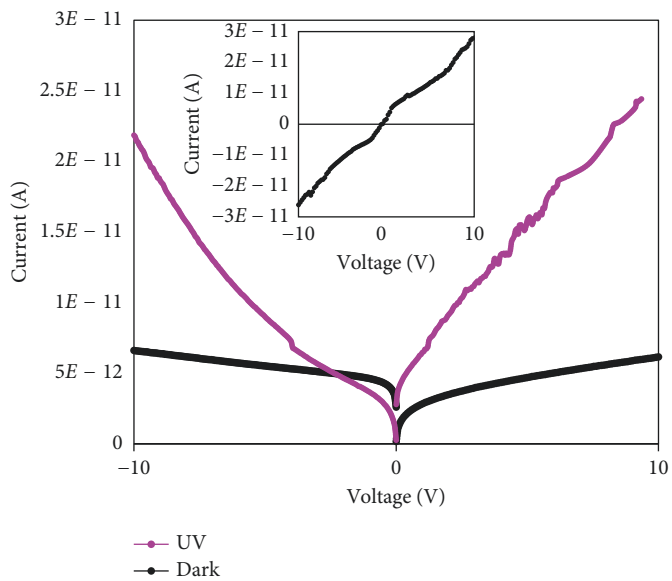

Figure 8: Dark current and photocurrent measured under 245 nm UV illumination. The inset figure shows I-V characteristics measured using SMU1 as "Force" at positive and as "Ground" at negative voltages.

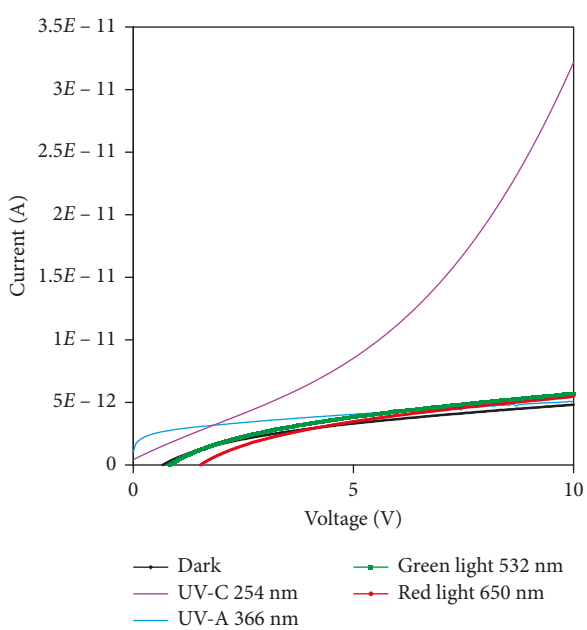

Figure 9: I-V characteristics of Ga_2O_3 thin films in dark and under different illuminations.

the linear part of the $(\alpha h\nu)^2$ versus $h\nu$ dependence (Figure 6). The obtained values for films grown at 300 W are shown in Table 2 and are similar to previously reported values [27–29].

A schematic representation of the structure used in the electrical characterization is shown in Figure 7. I-V measurements were carried out between top Al contacts on Ga_2O_3 using voltage sweeps from 0 to ± 10 V in dark, as well as under UV light illumination. Symmetric characteristics were obtained at positive and negative voltages (Figure 8), excluding the formation of a Schottky contact between the Al contact and the thin films. The slight asymmetry is an instrumentation effect, which most likely results from the probe station or from small variation of the characteristics of the two source-monitor units (SMUs) of Keithley 4200 SCS. The results in Figure 8 were obtained using SMU1 as a "Force" and SMU2 as a "Ground" for measurements at positive and at negative voltages. By changing the SMU1 to "Ground" and SMU2 to "Force," the absolute values of the current measured at negative voltages coincide with the current at positive voltages (inset in Figure 8). In the voltage range used, the photocurrent under 254 nm UV illumination reaches values ~5 times higher than the dark current in both directions.

In order to prove that the observed photocurrent is exclusively due to UV-C excitation, the samples were exposed to illumination with light sources of 650 nm (red), 532 nm (green), and 366 nm (UV-A). In Figure 9, a plot of the electrical behavior of β-Ga_2O_3 thin films under different illuminations is shown proving that the photocurrent was generated only under UV-C exposure. The I-V results are in agreement with the UV-Vis measurements where strong absorption occurs at wavelengths shorter than 270 nm.

4. Conclusions

β-Ga_2O_3 thin films with high crystal quality were grown successfully on c-plane sapphire substrate by plasma-assisted molecular beam epitaxy. In this work, the combined effect of substrate temperature, RF plasma power, and oxygen flux was studied showing that at a fixed O_2 flux the thickness of the grown film decreases with the increase of the temperature. Ellipsometry measurements revealed that, at oxygen flux of 1.0 sccm, the refractive index increases with the substrate temperature, while at flux of 1.5 sccm this dependence is very weak. The samples grown at 300 W RF plasma power exhibited higher transmittance than those grown at 200 W. Optical measurements showed that β-Ga_2O_3 starts to

absorb at wavelengths lower than ~270 nm, in the range of UV-C radiation. The electrical measurements under UV-C illumination showed that the grown thin films of β-Ga_2O_3 are excellent candidates for deep-ultraviolet sensing.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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