Effect of Homo-buffer Layers on the Properties of Sputtering Deposited Ga$_2$O$_3$ Films

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Abstract. β- Ga$_2$O$_3$ films were grown by radio-frequency magnetron sputtering method. The influence of Ga$_2$O$_3$ buffer layers and annealing treatment on the structural, optical, morphological and electrical properties of Ga$_2$O$_3$ films was studied. The results revealed an improvement of crystalline quality and transmittance of annealed β- Ga$_2$O$_3$ films prepared with homo-buffer layers. Ga$_2$O$_3$ film UV photodetectors were fabricated with a new B and Ga co-doped ZnO films (BGZO)/Au interdigitated electrode. A good ohmic contact was formed between the film and the electrode. For the detector based on Ga$_2$O$_3$ films with buffer layers, a higher value of photo response and faster response times was obtained.

1. Introduction

Ga$_2$O$_3$ has five different crystalline phases depending on ambient conditions, including α (corundum-hexagonal), β (monoclinic), γ (cubic), δ (cubic), and ε (orthorhombic) Ga$_2$O$_3$. Among them, monoclinic structured β- Ga$_2$O$_3$ is the most stable form [$1$]. β- Ga$_2$O$_3$ presents outstanding properties such as wide band gap (~4.9 eV), high mechanical strength, high breakdown field strength, high chemical and thermal stability [$2$, $3$]. Therefore, β- Ga$_2$O$_3$ has recently attracted much attention for its potential applications in optoelectronic devices [$4$], gas sensors [$5$], field effect transistors [$6$], transparent conductors [$7$] and solar-blind photodetectors [$8$].

As is known to all, the performance of the optoelectronic device is closely related to the film crystalline quality [$9$]. However, high quality β-Ga$_2$O$_3$ films were hard to be prepared by depositing directly on commonly used substrates such as glass, sapphire, Al$_2$O$_3$ and Si because of the lattice mismatch and different coefficient of thermal expansion [$2$, $10$]. In order to improve the films crystalline quality, the introducing of a buffer layer (hetero-buffer or homo-buffer layer) to decrease the lattice mismatch between films and substrates is a well-known method [$11$-$12$].

Up to now, there are no reports on the preparation of Ga$_2$O$_3$ films with homo-buffer (HB) layers. In this paper, attempts were made to utilize Ga$_2$O$_3$ HB layers to obtain high quality Ga$_2$O$_3$ films by using radio-frequency (RF) magnetron sputtering method. The influence of HB layers and annealing treatment on the performances of Ga$_2$O$_3$ films and Ga$_2$O$_3$ film solar-blind UV photodetectors were investigated in detail.

2. Experimental

Ga$_2$O$_3$ films were deposited by RF magnetron sputtering with Ga$_2$O$_3$ ceramic targets (purity of 99.99%). Quartz glasses with area of 20×20 mm$^2$ and the thickness of 1.5mm were used to be the substrate for
depositing the Ga₂O₃ film. First of all, cleaning the quartz glasses with acetone and alcohol about 15 min by the ultrasonic then being dried by N₂. Before the films preparation, the vacuum chamber was evacuated until the residual gas pressure less than 5x10⁻⁷ Torr. For all the films deposition, the sputtering gas was pure argon (Ar) and the sputtering pressure was 6mTorr.

The growth of Ga₂O₃ films were prepared by a two-step process. Firstly, the Ga₂O₃ HB layer with a thickness of 30 nm was prepared on quartz glasses substrates at low temperature of 200 °C and low sputtering power of 80W. Then, Ga₂O₃ films were deposited with a thickness of 300nm at high temperature of 500 °C and high sputtering power of 150W on the HB layers. The films were annealed at 1000 °C for 60 min in nitrogen. Ga₂O₃ films solar-blind UV photodetectors were fabricated with new B and Ga co-doped ZnO films (BGZO)/Au (50nm / 100nm) interdigitated electrodes deposited by RF magnetron sputtering and e-beam evaporation, respectively.

The structure of the Ga₂O₃ films was characterized by X-ray diffraction (XRD, D/MAX-2200V PC, CuKα1). The surface morphology of the Ga₂O₃ films was measured by scanning electron microscope (SEM, Apollo 300). The optical properties of the films were investigated by using a UV-visible spectrophotometer (Shimadzu UV-2501PC). The photoelectric properties of the Ga₂O₃ UV detectors were measured by Keithley 2400 and a PTI optical system.

3. Results and Discussion

Figure 1 shows XRD patterns of as-grown and annealed Ga₂O₃ films prepared with and without HB layers. For the Ga₂O₃ film directly deposited on substrates without HB layer, no distinct peaks were observed with the exception of a broad peak originating from the substrates, indicating the formation of amorphous Ga₂O₃ film. After annealing treatment, five peaks located at 31.94°, 33.08°, 37.82 °, 44.08° and 64.48° corresponding to monoclinic structured β-Ga₂O₃ (002), (111), (402), (601) and (512) appear (JCPDS No. 43-1012), indicating the initial amorphous structure evolved into the polycrystalline structure [13]. For as-grown Ga₂O₃ films with HB layers, three peaks corresponding to β-Ga₂O₃ (402), (601) and (512) are observed. For all the prepared Ga₂O₃ films, annealed Ga₂O₃ films with HB layers show the stronger diffraction intensity of (002) peak. The results indicate that the HB layer and annealing treatment can improve the polycrystalline quality of Ga₂O₃ films significantly.

Figure 2 shows SEM images of as-grown and annealed Ga₂O₃ films prepared with and without HB layers. The as-grown Ga₂O₃ film without HB layers shows a uniform and dense surface with small grain
size. After annealing treatment, the grain size of Ga$_2$O$_3$ films grows slightly larger. It can be obviously found, compared with samples without HB layers, the Ga$_2$O$_3$ films grown on HB layers show larger grain size, indicating the improvement of film crystalline quality which is consistent with the XRD results.

Figure 2. SEM images of Ga$_2$O$_3$ films: (a) annealed without HB layers, (b) annealed with HB layers, (c) as-grown without HB layers and (d) as-grown with HB layers

The transmittance spectra and absorption spectra in the wavelength range 200-800 nm for annealed Ga$_2$O$_3$ films with and without HB layers are shown in Figure 3 (a). Both samples (including the substrates) show high average transmittance over 85%. It can also be found that the transmittance of Ga$_2$O$_3$ films with HB layers is slightly higher than those without HB layers, which may be due to the better crystallinity and good uniformity of the films with buffer layers. From the absorption spectra, the fundamental absorption edges of the films can be found clearly. Figure 3(b) shows the plots of $(\alpha h\nu)^2$ versus $h\nu$ for annealed Ga$_2$O$_3$ films with and without HB layers, where $\alpha$ is the optical absorption coefficient and $h\nu$ is photon energy. As a direct band gap semiconductor, the optical energy band gap ($E_g$) values of the Ga$_2$O$_3$ films can be calculated using Tauc's formula [14]:

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

(1)

Where $C$ is a constant. The $E_g$ can be obtained by extrapolating the linear region of the plot $(\alpha h\nu)^2$ versus $h\nu$ and taking the intercept on the $h\nu$-axis [14]. And it is obvious shown in Figure 3(b) that the optical energy band gap of films with and without homo-buffer layers is 4.95eV and 4.87eV, respectively.
The transmission spectra and the absorption spectra (a) and the $(ah\nu)^2$ vs. $h\nu$ plots for Ga2O3 films (b).

Figure 3. The transmission spectra and the absorption spectra (a) and the $(ah\nu)^2$ vs. $h\nu$ plots for Ga2O3 films (b).

Figure 4. shows the room temperature $I$-$V$ characteristics of Ga$_2$O$_3$ films detectors with and without homo-buffer layers in dark. To evaluate the contact characteristics, the $I$-$V$ plots were fitted using non-linear function: $I(V) = aV^b$, where $a$ is constant, $b$ is an ohmic resistance coefficient. If $b=1$, the contact is ohmic [15]. The calculated value $b$ is about 1.0010 which is much close to 1. The results confirm the ohmic contacts between BGZO/Au and Ga$_2$O$_3$ films. It can also be found clearly that the dark current of detectors with homo-buffer layer reduce 3 times due to the improved crystalline quality with less grain boundaries and defects.

Figure 4. shows the room temperature $I$-$V$ characteristics of Ga$_2$O$_3$ films detectors with and without $HB$ layers measured under bias of 20 V with turning on and off as 250 nm light irradiated on the device. Upon UV illumination, the photocurrent for detectors with and without $HB$ layers increased to about 490 and 230 nA, respectively. As turn the UV light off, it decreased dramatically to the initial value about 12 nA and 30 nA, respectively. The detectors exhibit a nearly identical response after multiple illumination cycles indicating that the device has good reproducible characteristics. For Ga$_2$O$_3$ detectors with and without $HB$ layers, the UV light to dark current ratio is about 41 and 8, respectively.

The energy of the UV (250nm) photons ($\sim 5eV$) is larger than the optical energy band gap of Ga$_2$O$_3$ films. When UV light illuminates the detectors, the UV photons are absorbed in Ga$_2$O$_3$ films, generating electron-hole pairs, responsible for the photocurrent in detectors. In the sputtered polycrystalline Ga$_2$O$_3$ films, there are inevitably many defects and grain boundaries acting as the trapping centers which is also named carrier trapping effect [16]. Before being separated and collected by electrodes, the generated carriers in Ga$_2$O$_3$ film detectors could be seized by those trapping centers. Therefore, the film crystalline quality strongly affects the properties of detectors, which is the reason for difference in UV light to dark.
current ratio of Ga₂O₃ UV detectors. The better crystalline quality of Ga₂O₃ films with HB layers resulting in the higher UV light to dark current ratio of detectors. Rise time (tᵣ) and fall time (tᵢ) is defined as the time taken to vary between 10% and 90% of the maximum photocurrent. From Figure 5, tᵣ is about 0.1s and 0.15s, and tᵢ is about 0.2s and 0.3s for detectors with and without HB layers respectively. The faster response times for detectors with HB layers are mainly attributed to the higher crystalline quality of Ga₂O₃ films.

4. Summary
A two-step growth regime was used to achieve an epitaxial growth of β- Ga₂O₃ films by RF magnetron sputtering method. The influence of homo-buffer layers and annealing treatment on the properties of Ga₂O₃ films was investigated. The results indicate that the homo-buffer layers and annealing treatment can improve the polycrystalline quality of Ga₂O₃ films. The films with homo-buffer layers show larger grain size and higher transparency. The optical band gap of films with homo-buffer layers is about 4.95eV. The higher value of UV light to dark current ratio and better time-dependent characteristic of the Ga₂O₃ detectors with homo-buffer layers manifest that the film crystalline quality plays an important role on the performance of detectors.

5. References
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