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Growth of gallium oxide thin films on silicon by the metal organic chemical vapor deposition method

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Abstract

We have deposited gallium oxide (Ga_2O_3) films on Si(1 0 0) substrates by metal organic chemical vapor deposition (MOCVD), by a reaction of a trimethylgallium (TMGa) and oxygen (O_2) mixture. The effect of temperature on growth and structure of films has been investigated at temperatures of 500–600 °C. We revealed that the films were amorphous with very small crystallites. The films were smooth but the surface roughness increased with increasing the growth temperature.

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Keywords: MOCVD; Ga_2O_3; Temperature; Thin film

1. Introduction

Gallium oxide (Ga_2O_3) has recently attracted interests as a new material for a gas sensor [1–3], transparent conductor [4,5], phosphor [6,7], and solar cells [8–10]. For the production of Ga_2O_3 films, various synthetic methods such as oxidation of Ga-containing surfaces [11–13], evaporation [14], sol–gel process [15], sputtering [16–18], pulsed laser deposition [19], molecular beam epitaxy [20], and chemical vapor deposition (CVD) have been studied. In order to use the metal organic CVD method, which has an advantage of more flexibility, good step coverage, producing uniform, pure, reproducible, and adherent films, the precursors of $Ga(hfac)_3$ with O_2 [11], $Ga[OCH(CF_3)_2]_3{\cdot}HNMe_2$ with H_2O [21], and $[Ga(\mu\text{-O-}t\text{-Bu})(\text{O-}t\text{-Bu})_2]_2$ with O_2 [22] have been developed.

Although various material, such as Al_2O_3 [15,19], GaAs [23], CoGa(1 0 0) [12], GaN [13], Ni(1 0 0) [24] have been studied as substrates for the Ga_2O_3 growth, there are rare reports on the growth onto the silicon (Si) substrate [18], which will pave the way for integration of future devices with developed Si integrated circuit technology.

Here, we investigate the possibility of obtaining the Ga_2O_3 films on Si substrates using the MOCVD technique, in the temperature range of 500–600 °C. This paper describes the first CVD growth of Ga_2O_3 films using a simple reaction of a trimethylgallium (TMGa) and oxygen (O_2) mixture. We investigate the effect of deposition temperature on the structural property of thin films.

2. Experimental

The Ga_2O_3 films were deposited on p-type Si substrate with (1 0 0) orientation, which was cleaned with organic solvents and dried before loading into the system. A schematic diagram of the MOCVD reactor used in our experiments was previously described [25].

High-purity Ar (99.999% purity) passed through the TMGa bubbler and delivered TMGa vapor to the reactor. The TMGabubbler was maintained at the temperature of −5 °C. The Ga_2O_3 film was synthesized by supplying O_2 and Ar carrier gases with the flow rate of 30 and 30 sccm, respectively in the temperature range of 500–600 °C for 10 min. As-deposited film exhibited a uniform and smooth surface.

An SEM (Hitachi S-4200, 30 kV) was used to observe the structure morphology of Ga_2O_3 film and an XRD (Philips, CM20T, 200 kV, Cu Kα1 $\lambda = 1.5405$ Å) was used to characterize the structural quality of the films. An AFM (Nanoscope III, Digital Instruments) was used to evaluate the surface roughness of the films.

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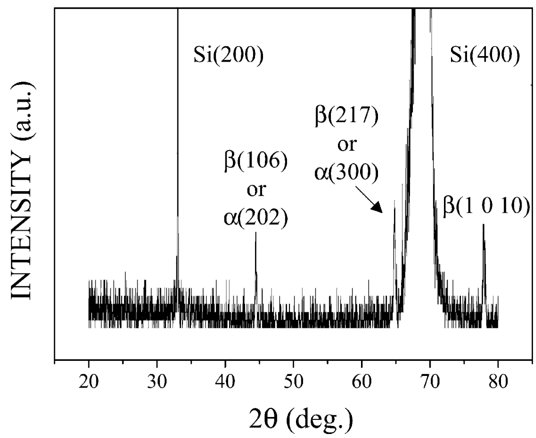

Fig. 1. XRD patterns recorded from Ga_2O_3 deposits at 550 °C. Indexing of weak diffraction peaks corresponds to a monoclinic structure of β-Ga_2O_3 and/or a hexagonal corundum structure of α-Ga_2O_3.

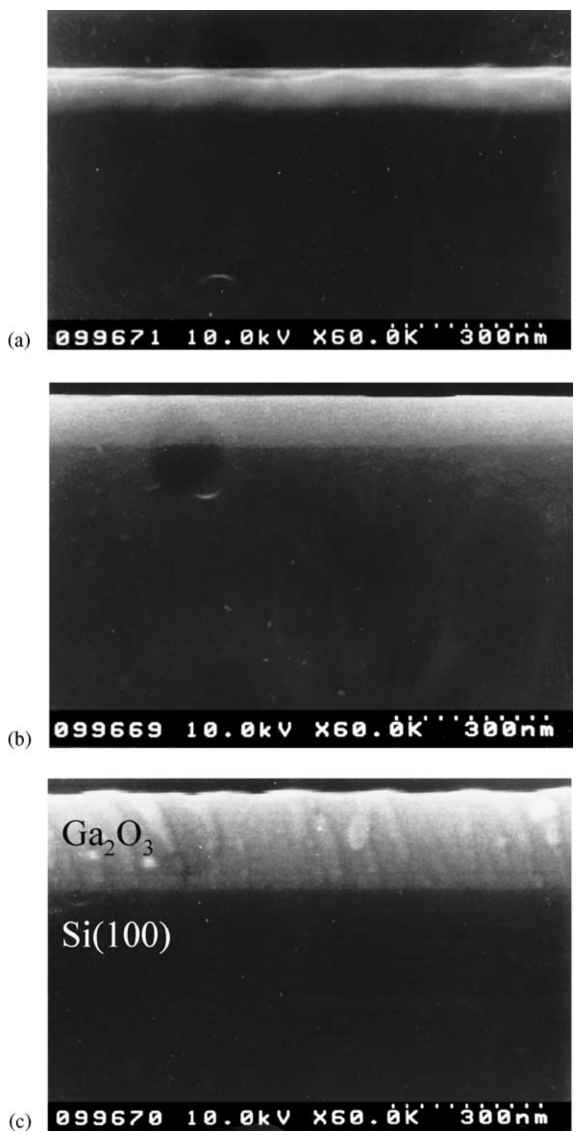

Fig. 2. Cross-sectional SEM images of Ga_2O_3 films at a growth temperature of (a) 500 °C, (b) 550 °C, and (c) 600 °C.

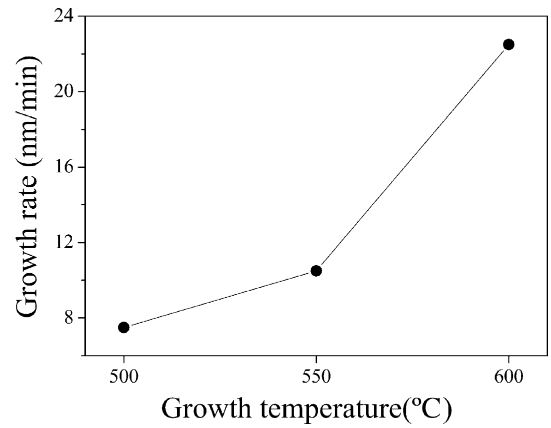

Fig. 3. Dependence of film growth rate on the substrate temperature, indicating that the growth rate increases with increasing the growth rate.

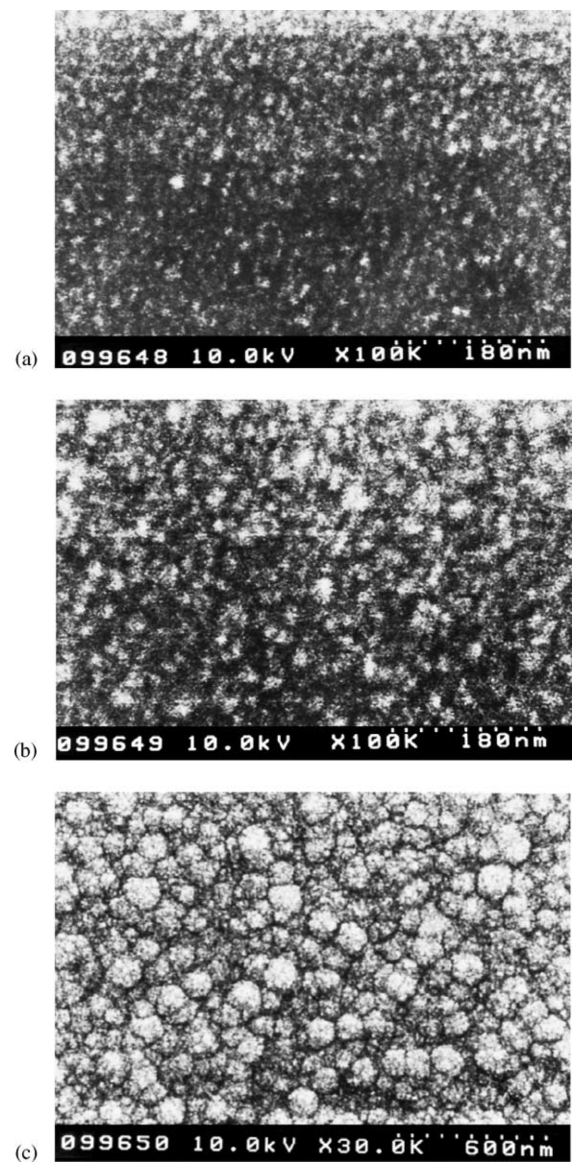

Fig. 4. Plain-view SEM images of Ga_2O_3 films at a growth temperature of (a) 500 °C, (b) 550 °C, and (c) 600 °C.

3. Results and discussion

Fig. 1 shows the XRD patterns of deposits on Si substrates at the growth temperature of 550 °C. The θ–2θ scan data of deposits exhibit strong 2θ peaks at 32.96°and 69.13°, respectively, corresponding to the (0 0 2) and (0 0 4) peaks of Si. Although we suppose that the deposits are close to the amorphous phase due to the absence of a strong Ga_2O_3 diffraction peak, there are weak diffraction peaks, corresponding to the Ga_2O_3 structure. Within experimental error, the lines observed in this diffractogram are found to coincide with (2 0 2) peak of α-Ga_2O_3 or (1 0 6) peak of β-Ga_2O_3, (3 0 0) peak of α-Ga_2O_3 or (2 1 7) peak of β-Ga_2O_3, and (1 0 10) peak of β-Ga_2O_3 (JCPDS 11-370). The α-Ga_2O_3 is metastable and has a hexagonal corundum structure and the stable β-Ga_2O_3 phase has a monoclinic structure. The existence of the Ga_2O_3 peaks indicates the production of Ga_2O_3 deposits on Si substrates. We infer from XRD data that the deposits are crystallographically amorphous or contain very small crystallites.

Fig. 2 shows the cross-sectional SEM images at growth temperatures of 500, 550, and 600 °C. According to SEM images, we are not able to clarify the grain boundaries at the temperature of 500–550 °C. However, the film structure is composed of unknown defects at the temperature of 600 °C, possibly representing the grain boundaries. Fig. 3 shows the dependence of film growth rate on the substrate temperature, indicating that the growth rate increases with

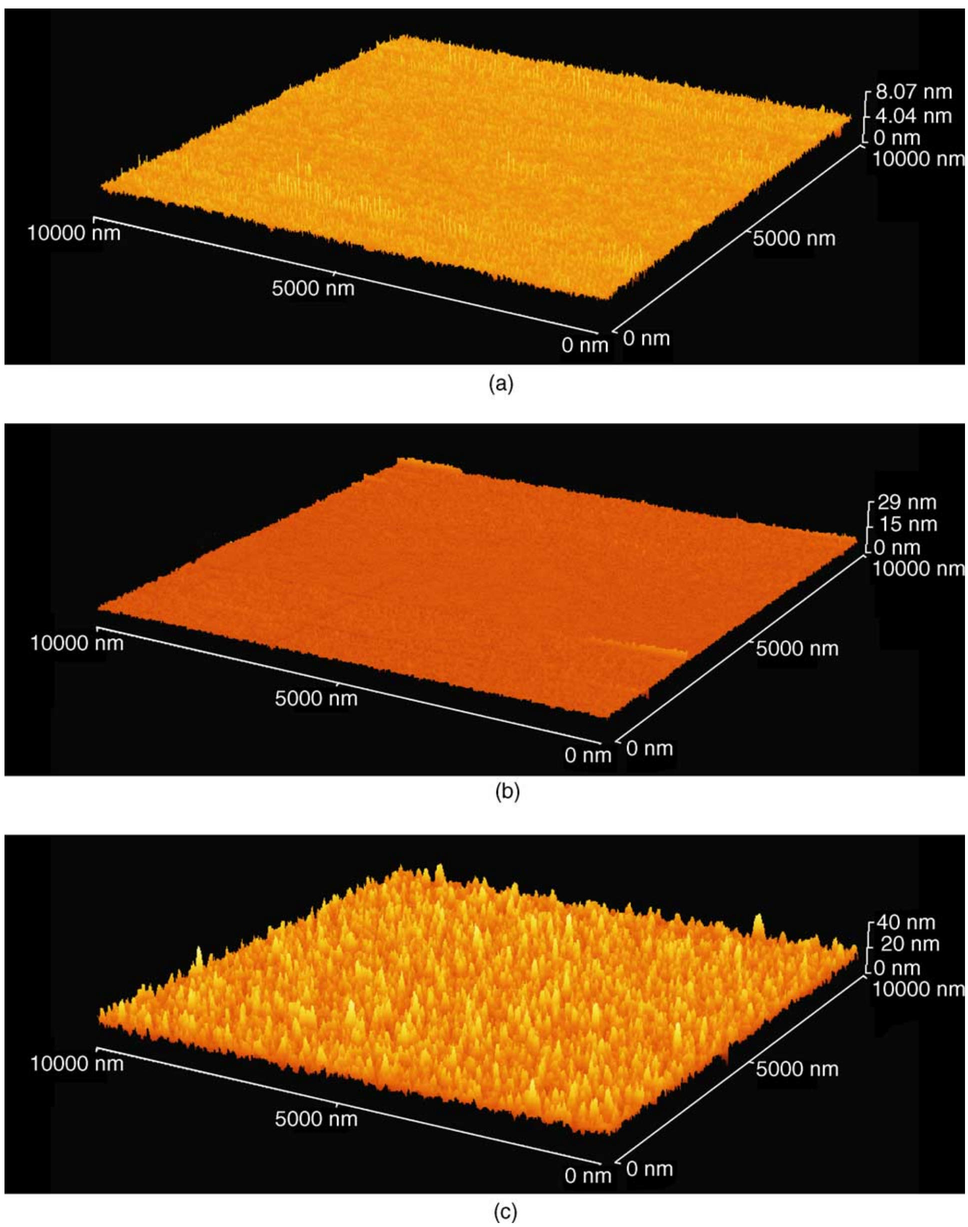

Fig. 5. AFM images of Ga_2O_3 films at a growth temperature of (a) 500 °C, (b) 550 °C, and (c) 600 °C.

increasing the growth rate, from 75 Å/min at 500 °C to 225 Å/min at 600 °C.

Fig. 4 shows the plan-view SEM image of Ga_2O_3 thin films. We surmise that the grains appeared on top of the film at a growth temperature of 600 °C. Previous researcher also reported that the crystallinity of Ga_2O_3 thin films was enhanced with increasing the substrate temperature [11] and we surmise that at high temperature, the atoms have enough diffusion activation energy to occupy the correct site in the crystal lattice and crystallization from the amorphous phase can be initiated. More systematic study is underway in order to reveal the temperature effect on the initiation of crystallization and grain growth in the Ga_2O_3 thin film.

In order to investigate the effect of substrate temperature on the surface roughness of Ga_2O_3 films, we have performed an AFM measurement on the Ga_2O_3 film deposited in the temperature range of 500–600 °C (Fig. 5). The root mean square (RMS) surface roughness of the films deposited at 500, 550, and 600 °C, respectively, are 0.38, 0.60, and 3.91 nm, revealing that surface roughness increases with increasing growth temperature. This observation on the surface roughness may be related to the observation of SEM, indicating that the grain size of Ga_2O_3 films increases with increasing substrate temperature. The high substrate temperature may cause the grain to overgrow and induce the rough surface. Further studies are necessary to reveal the detailed mechanism on the formation of Ga_2O_3 thin films.

4. Conclusions

We have demonstrated the deposition of Ga_2O_3 thin films on Si substrates by MOCVD using the TMGa as a precursor in the presence of oxygen. The XRD data and the SEM images reveal that amorphous Ga_2O_3 films are deposited on Si substrate. The growth rate of Ga_2O_3 thin films increases with increasing growth temperature at 500–600 °C. The AFM analysis indicates that the surface roughness increases with increasing the growth temperature. The first production of Ga_2O_3 thin films on Si using the conventional source will shed light on the potential application of Ga_2O_3 films.

Acknowledgements

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