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β-Ga_2O_3 epitaxial films deposited on epi-GaN/sapphire (0001) substrates by MOCVD

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ABSTRACT

Gallium oxide (Ga_2O_3) films were grown on epi-GaN/sapphire (0001) substrates by the metalorganic chemical vapor deposition (MOCVD) method, followed by a post-deposition thermal annealing at different temperatures of 800, 900 and 1000 °C for one hour in air. X-ray diffraction (XRD, both $\theta \sim 2\theta$ and Φ scans) and transmission electron microscopy (TEM) were used to inspect the lattice structure and epitaxial relationship. With the increase of annealing temperature, the film structure changed from amorphous to single crystalline and then to polycrystalline. A single crystalline β-Ga_2O_3 film with the best crystalline quality was obtained under the annealing condition at 900 °C, and a clear epitaxial relationship of β-Ga_2O_3(100)‖GaN(0001) with β-$Ga_2O_3 < 010 >$ ‖GaN $< \bar{1}2\bar{1}0 >$ and β-$Ga_2O_3 < 001 >$ ‖GaN $< \bar{1}010 >$ was determined. The elemental composition and proportion of the 900 °C-annealed film were investigated by the X-ray photoelectron spectroscopy (XPS) measurements and the atomic ratio of Ga/O was about 0.69, which was close to stoichiometric Ga_2O_3. For the sample annealed at 900 °C, the average transmittance in the visible wavelength region was about 78% and the average reflectivity in the 200–800 nm wavelength range was about 18%.

1. Introduction

Metallic oxide materials such as ZnO, TiO_2 and CuO synthesized by different methods are widely used in many fields of photocatalysis, gas sensors and photoelectric devices [1–10]. In recent years, a material with deep ultraviolet emission such as AlGaN, ZnMgO, and diamond have attracted more and more interests owing to the promising applications in solar-blind deep ultraviolet (DUV) photodetectors, quantum-well devices and high-density optical data storage [11,12]. Gallium oxide (Ga_2O_3) is one of the emerging wide band gap materials (*Eg*, 4.2–4.9 eV) due to its excellent thermal and chemical stability [13]. Therefore, Ga_2O_3 shows wide applications in high-temperature gas sensors, UV transparent conducting oxides, and efficient phosphors [14–17].

Among the five phases of Ga_2O_3 (α-, β-, γ-, η- and ε-Ga_2O_3), β-Ga_2O_3 is the most stable one [18,19]. The lattice parameters of the monoclinic structured β-Ga_2O_3 are a = 12.23 Å, b = 3.04 Å, c = 5.8 Å, α = 90̊, β = 103.7̊ and γ = 90̊ (JCPDS 43-1012). The band gap of β-Ga_2O_3 is 4.9 eV, which is wider than most of the transparent conducting oxides (TCOs) [20–22]. The optical transmittance of β-Ga_2O_3 films in UV region is more than 80%. On the premise of the advantage, β-Ga_2O_3 can be used in the field of transparent optoelectronic devices such as vertical solar-blind deep-ultraviolet schottky photodetectors [23]. The main limitations of single crystal β-Ga_2O_3 materials are their high price and poor thermal conductivity. Some literatures about β-Ga_2O_3 materials preparation have been reported. Z.Yu obtained amorphous Ga_2O_3 thin films on GaAs(001) by molecular-beam epitaxy [24]. Z.Yanget al. obtained β-Ga_2O_3 nanorods on Si(111) substrates by annealing the Ga_2O_3/V films [25]. Polycrystalline Ga_2O_3 films were grown on sapphire substrates by MOCVD [26]. β-Ga_2O_3 epilayer grown on Al_2O_3(0001) at low temperature by low-pressure MOCVD was used to fabricate the solar-blind DUV photodetectors, and the effect of annealing on the β-Ga_2O_3 epilayer defects and detector performance were investigated [27]. However, based on literature, depositing monocrystalline Ga_2O_3 films is still a challenging research subject. Based on the calculation of the lattice mismatch between β-Ga_2O_3 and GaN (0001), we find that it is possible to grow monocrystalline β-Ga_2O_3 on GaN(0001). Compared with other substrates such as GaAs, sapphire, and SiC, GaN exhibits excellent luminescence and conduction properties. Therefore, it has been widely used in not only high-temperature/high-power electronic devices, but also optoelectronic devices such as high electron mobility transistors, blue lasers, and optical communication [28]. Combining the unique properties of β-Ga_2O_3 with the GaN technology is a promising method to develop future applications in

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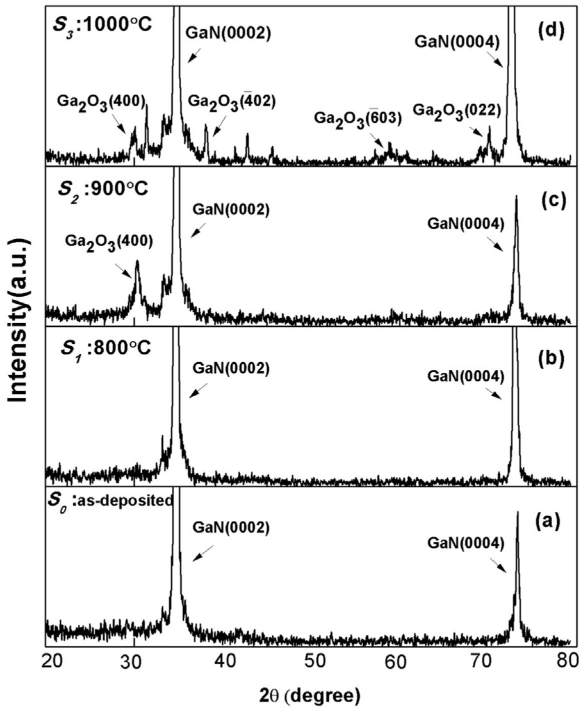

Fig. 1. XRD $\theta-2\theta$ scans of the Ga_2O_3 films before and after annealing at different temperatures.

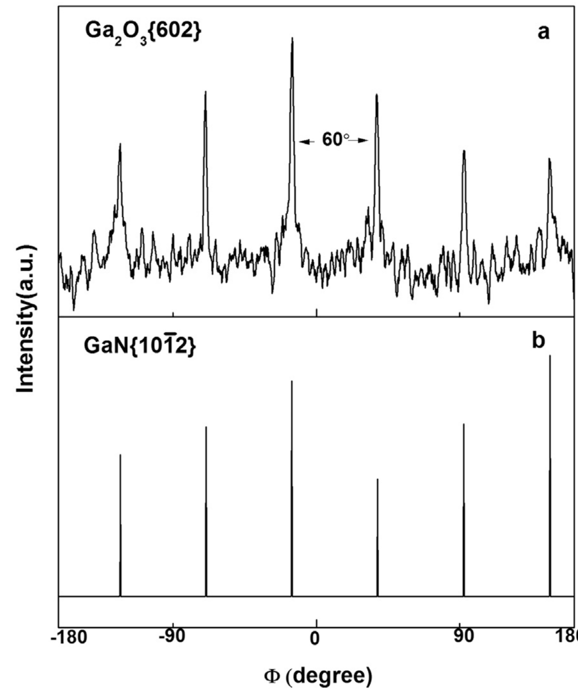

Fig. 3. Off-specular Φ-scans of β-Ga_2O_3{602} and GaN{101()2} planes at a fixed position.

electronic and optoelectronics devices. For example, a metal-oxide-semiconductor field-effect transistor (MOSFET) based on Ga_2O_3/GaN have been studied widely. In this paper, β-Ga_2O_3 films were deposited on epi-GaN/sapphire (0001) substrates by MOCVD and then annealed at different temperatures. High quality single crystalline β-Ga_2O_3 film grown along the [100] direction was obtained and the corresponding epitaxial relationship was identified. The structural, morphological, optical, and compositional properties of the Ga_2O_3 films were investigated and discussed in detail.

2. Experimental details

The deposition of Ga_2O_3 films was deposited using MOCVD system. The O_2 (purity 5 N) and trimethylgallium [$Ga(CH_3)_3$] (purchased from Nanda Opto-electronic materials Co., Ltd., China) served as the oxidant and metalorganic source, respectively. Commercial epi-GaN/sapphire (0001) wafers (GaN epilayer thickness: 4 µm, purchased from Shanghai Daheng Optics and Fine Mechanics Co., Ltd.,China) were used as the substrates. During the deposition, the trimethylgallium was stored in a stainless steel bubbler keeped at −14.5 °C. The metalorganic source vapor and O_2 were transported into the reactor with ultrahigh purity N_2

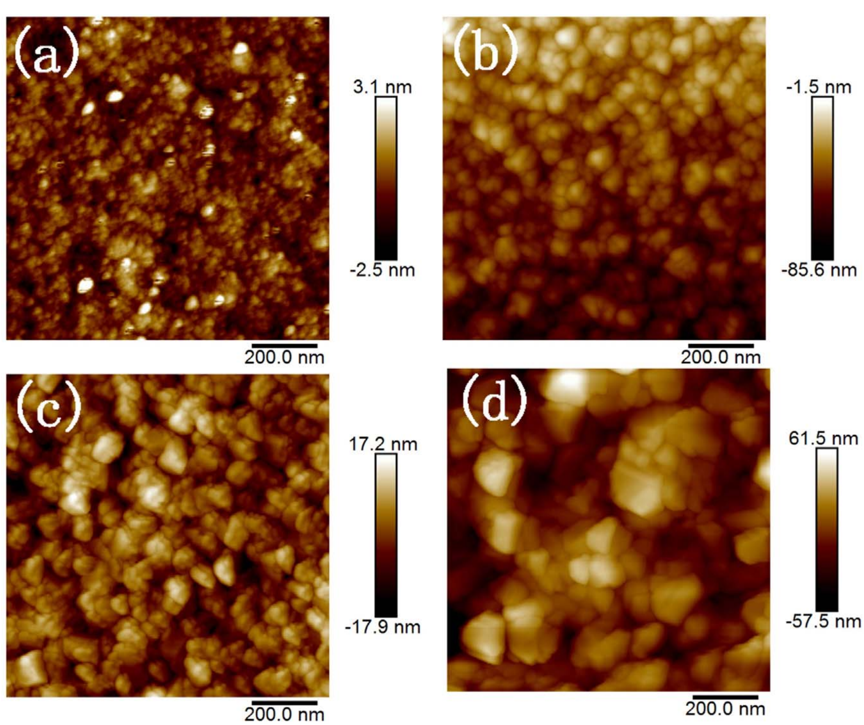

Fig. 2. AFM images of: (a) S_0, (b) S_1, (c) S_2 and (d) S_3.