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Growth and characterization of nonpolar (10-10) ZnO transparent conductive oxide on semipolar (11–22) GaN-based light-emitting diodes

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We have grown thin films of nonpolar m-plane (10-10) ZnO on a semipolar (11–22) GaN template by atomic layer deposition (ALD) at low growth temperatures (<200 °C). The surface morphology of the ZnO film is found to be an arrowhead-like structure, which is a typical surface structure of the semipolar (11–22) GaN films. On increasing the growth temperature of the ZnO films, the concentration and mobility of the charge carriers in the ZnO film are increased. However, the optical transmittance decreases with an increase in the growth temperature. Based on these results, we have fabricated semipolar (11–22) GaN-based light-emitting diodes (LEDs) with nonpolar m-plane ZnO film as a transparent conductive oxide (TCO) to improve the light extraction efficiency. In spite of a decrease in the optical transmittance, the operation voltage of semipolar (11–22) GaN-based LEDs is found to decrease with an increase in the growth temperature, which might be due to the improvements in the electrical properties and current spreading effect, resulting in an increase in the optical output power.

1. Introduction

Commercially available III-nitride-based optoelectronic devices typically consist of epitaxial multilayer thin films that are grown along the polar c-axis of the wurtzite crystal structure, and they suffer from the quantum confinement Stark effect owing to the existence of strong piezoelectricity and spontaneous polarization [1,2]. The build-up of the electric fields along c-direction causes a spatial separation of electron and hole wave functions, thereby reducing the light emission efficiency and causing a blue-shift phenomenon of the emission wavelength with an increase in the injection current owing to the carrier-induced screening effect [3,4]. To solve these issues, there have been many research efforts that focus on nonpolar a-plane (11–20) and m-plane (10-10) GaN-based light-emitting diodes (LEDs) [5,6]. Other approaches to decrease or potentially eliminate the polarization effects by the growth of (11–22), (10-10) and (10-1-3) semipolar GaN-based LEDs on various substrates have also been examined [7–9]. Although there have been many efforts to achieve high quality nonpolar and semipolar GaN-based LEDs by epitaxial growth, the efficiencies of these LEDs are still not comparable with that of the conventional polar c-plane GaN-based LEDs owing to the anisotropic crystallographic-induced crystal defects and arrowhead-like surface defects in the nonpolar and semipolar GaN structures [10,11]. Therefore, many researchers have focused on the reduction of crystal defects in the heteroepitaxial growth of semipolar GaN epilayers.

Besides the epitaxial growth, there are various effective techniques in improving the external quantum efficiency of LEDs such as in the chip design, fabrication, and package process [12–14]. We have focused on the use of a current spreading layer on semipolar GaN-based LEDs to improve the light extraction efficiency with a nonpolar ZnO transparent conductive oxide (TCO). Regardless of polar, nonpolar, and semipolar GaN-based LEDs, indium tin oxide (ITO) has been widely used as a TCO material to increase the current spreading length in the LEDs because of its unique optical and electrical properties [15,16]. ZnO has also attracted much attention for the use of TCO in GaN-based LEDs due to its advantages over ITO such as the low growth temperature, low cost, low lattice mismatch with GaN, and good stability. There are several reports in the

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literature on the growth of nonpolar m-plane (10-10) and semipolar (10−11) ZnO films on m-plane sapphire substrates [17–22]. However, the applications of nonpolar ZnO TCO films grown on semipolar (11−22) GaN-based LEDs have not been reported yet. In this study, we investigate the optical and electrical properties of semipolar (11−22) GaN-based LEDs with nonpolar m-plane (10-10) ZnO as a TCO material that is grown by atomic layer deposition (ALD).

2. Experimental procedure

100 nm-thick ZnO films were grown on m-sapphire, c-sapphire and semipolar (11−22) GaN templates by an ALD system using high-purity diethylzinc (DEZn) and distilled water (H_2O) as precursors for Zn and oxygen, respectively. In a typical ALD cycle, DEZn and H_2O were fed into the chamber through separate inlet lines and N_2 gas was purged [23]. The times of the DEZn and H_2O exposures, and the N_2 purging were 1.0 s and 5.0 s, respectively and the flow rate of all the samples were fixed at 1600 sccm. 100 nm-thick ZnO films were grown on a semipolar (11−22) GaN-based LED epitaxial structure that consisted of a 2.0 μm-thick undoped GaN layer, a 3.0 μm-thick Si-doped n-type GaN layer, 5 periods of $In_{0.15}Ga_{0.85}N$/GaN quantum wells (QWs) as an active layer, a 20 nm-thick p-type $Al_{0.2}Ga_{0.8}N$ as an electron blocking layer and a 80 nm-thick Mg-doped p-type GaN layer. To evaluate the effects of the growth temperature of the ZnO films on the optical and electrical properties of semipolar GaN-based LEDs, the growth temperature was varied from 115 to 195 °C. After preparing the semipolar (11−22) GaN-based LED wafers with nonpolar ZnO film as the TCO material, we fabricated conventional mesa-structured LED chips (300 μm × 300 μm) with lateral electrode structures.

The crystal planes of the ZnO films were examined by high-resolution X-ray diffraction (HR-XRD). The electrical properties of ZnO films were measured using the van der Pauw method. The optical transmittances of ZnO films were evaluated by UV–visible spectroscopy in the wavelength range from 350 nm to 750 nm. Light-current–voltage (L–I–V) measurements on the semipolar GaN-based LEDs with nonpolar ZnO TCO were carried out using HP 4155B to determine the optical power and the operation voltages. The electroluminescence (EL) spectra of the samples were measured using an USB 4000-UV-VIS miniature fiber optic spectrometer.

3. Results and discussion

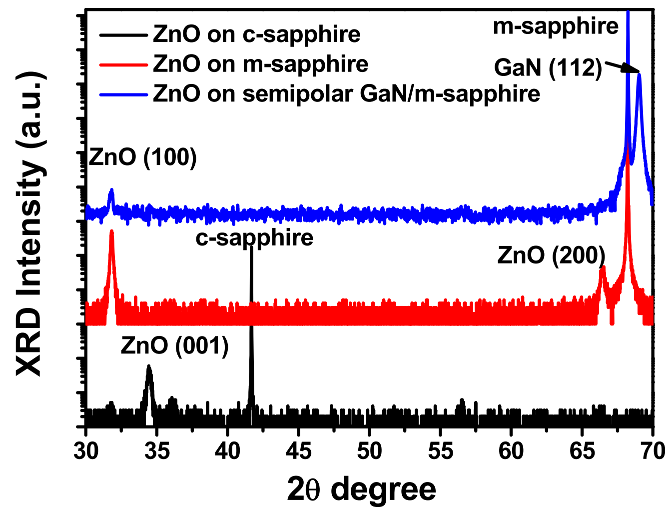

Fig. 1. HR-XRD ω-2θ scans of ZnO films grown on c-plane, m-plane sapphire, and semipolar (11−22) GaN template.

Fig. 1 shows the HR-XRD ω/2θ scans of the ZnO films grown on c-sapphire, m-sapphire and semipolar (11−22) GaN/m-sapphire. ZnO film grown on the c-sapphire substrate exhibits two peaks, at 41.67° and 34.4° which correspond to the (006) c-plane of the sapphire and the polar c-plane (002) of the ZnO film, respectively. This result indicates that the crystallographic plane of ZnO film grown on c-sapphire is the polar c-plane. In the case of ZnO film grown on m-sapphire, three peaks were observed corresponding to the nonpolar (100), (200) planes of ZnO and (30-30) of the m-sapphire. We observed four peaks in the case of ZnO film grown on semipolar (11−22)/m-sapphire, which correspond to nonpolar (100), (200) of ZnO, (300) of m-sapphire and semipolar (11−20) GaN, respectively [24]. These XRD results indicate that the crystallographic plane of the ZnO film is significantly affected by that of the templates on which the films are grown. In particular, we find that polar (001) c-plane and nonpolar (100) m-plane ZnO films were grown on (001) c-plane and (100) m-plane sapphire substrates, respectively, which indicates that the crystallographic plane of the ZnO films is determined by that of the sapphire substrates. However, the crystallographic plane of ZnO film grown on semipolar (11−22) GaN template is not semipolar (11−22) ZnO, but nonpolar (10-10) ZnO. In general, the surface structure of semipolar (11−22) GaN film is found to be arrowhead-like with three crystallographic planes (one {000-1} plane and two {10-10} planes). We surmise that the growth of ZnO is initiated from the m-planes of the arrowhead-like structure, leading to the formation of nonpolar (10-10) ZnO films on semipolar (11−22) GaN films. However, further experiments have to be carried out using the ALD system in order to study the growth mechanism.

Fig. 2 (a) and (b) show the AFM images (1.0 μm × 1.0 μm) of polar (0001) c-plane and nonpolar (10-10) m-plane ZnO films grown on c-plane and m-plane sapphire substrates, respectively. The surface morphology examination of polar (0001) ZnO film shows an isotropic granular-shaped surface whereas nonpolar (10-10) ZnO film exhibits a relatively anisotropic granular-shaped surface owing to the anisotropic lattice structure [25]. Fig. 2 (c) shows the surface morphology of nonpolar (10-10) ZnO film grown on semipolar (110) GaN template. The surface structure of nonpolar (10-10) ZnO film exhibits a preferred orientation towards [11-2-3], which is a typical of arrowhead-like surface structure of semipolar (11−22) GaN film, as shown in Fig. 2 (d). Moreover, the presence of small granular-shaped structures embedded in the arrowhead-like surface structure is similar to the surface granular structure of the nonpolar (10-10) ZnO film grown on m-sapphire. Based on these AFM and XRD results, we believe that the nonpolar (10-10) m-plane ZnO film is grown from the m-planes of the arrowhead-like structure of semipolar (11−22) GaN film, leading to the anisotropic granular shape with preferred orientation towards [11-2-3]. Moreover, as the growth temperature is increased from 115 to 195 °C, the root mean square (RMS) roughness of the nonpolar (10-10) ZnO film grown on the semipolar (11−22) GaN template decreases from 10.02 nm to 6.49 nm, and the surface grain size of the nonpolar (10-10) ZnO film increases from 56 nm to 85 nm not shown here. This can be attributed to the acceleration of nucleation and growth step due to an increase in the surface migration energy with increasing growth temperature, leading to the large surface grain size and low roughness.

We measured the electrical properties of nonpolar (10-10) m-plane ZnO films grown at different growth temperatures by a Van der Pauw method that is used for the Hall measurements at room temperature, and the results are given in Fig. 3 (a). The carrier concentration and mobility of the nonpolar (10-10) ZnO film are