Comparative Study About the Structural Properties of ZnO Films Grown on Si, GaAs, And Al2O3 Substrates and a GaN Template

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Abstract. We report on the structural properties of ZnO films grown on (001) Si, (001) GaAs, (0001) Al_2O_3 substrates and a (0001) GaN template by RF-sputtering. It is observed that all the ZnO films have textured structures with a preferred orientation of the c-axis, irrespective growth conditions, in terms of atomic-force microscope and wide-range X-ray diffractometer. However, it is found that the ZnO films in the ZnO-on-Si and ZnO-on-GaAs are consisted of various domains with different orientations, while the ZnO films in the ZnO-on-Al_2O_3 and ZnO-on-GaN are consisted of a single domain with the same orientation in terms of the φ-scan of X-ray diffractometer. Moreover, it is found that the ZnO-on-GaN has smaller edge dislocation density and screw dislocation density than the ZnO-on-Al_2O_3, which seriously depends on substrate temperature, in terms of the ω-scan of X-ray diffractometer.

1 Introduction

ZnO has attracted an attention as one of the promising materials required to realize the emitters and detectors in the ultraviolet region and the high-speed devices in the information communication system, because it has the outstanding physical properties of a wide-bandgap of 3.37 eV, an exiton-binding energy of 60 meV, a saturation velocity of 3.1×10^7 cm/sec, and a cohesive energy of 1.89 eV [1-3]. However, ZnO has an essential problem where it is still lack of the reliably available homosubstrate that thin film ZnO can be grown on, even if the substrate that a material can be grown on is an essential problem in the thin film technology. Instead, Al_2O_3, GaN, Si, and GaAs substrates, etc have been tried as the second best thing. The ZnO film of a wurzite structure is generally grown on the (0001) Al_2O_3 substrate of a corundum structure, which is the most developed substrate among the materials with the similar crystalline structure to ZnO. However, the ZnO lattice has a lattice mismatch of 30 % between the basal plane of the Al_2O_3 lattice and a lattice mismatch of 18 % between the small cell of Al atoms on the basal plane [4,5]. The GaN template, which is fabricated on the Al_2O_3 substrate by metal-organic chemical-vapor deposition (MOCVD), with the same crystalline structure as ZnO effectively can suppress the structural defects in the subsequently grown ZnO film, because the GaN lattice has a very small lattice mismatch of 1.8 % with the ZnO lattice. However, because the GaN

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template is also grown on the Al_2O_3 substrate, the intrinsic problem derived from the Al_2O_3 substrate itself is remained [5,6]. The Si substrate of a diamond structure and the GaAs substrate of a zinc blende structure, which have the highest crystalline qualities in the surface and bulk among any other developed semiconductor substrates, have the merits of large-area wafer ability and economic feasibility. However, the two materials have the structural mismatches with ZnO, which hampers the epitaxial growth of ZnO films on the both substrates [7-10]. We note that there is not yet systematic result about the difference in the structural properties of the ZnO films grown on each substrate, referred above, though a lot of research results about the growth technique and physical properties of thin film ZnO have been reported until now. In this work, we have a comparative study about the structural properties in the surface and bulk of the ZnO films grown on Si, GaAs, and Al_2O_3 substrates and a GaN template by using RF-sputtering.

2 Experimental details

Four types of ZnO films were prepared on (001) Si, (001) GaAs, and (0001) Al_2O_3 substrates and a (0001) GaN template by RF-sputtering. Each sample was grown under each optimized condition, whose detailed conditions were reported elsewhere [5,8,10]. The ZnO-on-Si structure was grown at a substrate temperature of 500°C and an $O_2/Ar+O_2$ ratio of 80 %. The ZnO-on-GaAs structure was grown at a substrate temperature of room temperature (RT) and an $O_2/Ar+O_2$ ratio of 0 (Ar = 100 %). The ZnO-on-Al_2O_3 structure and the ZnO-on-GaN structure were grown at a substrate temperature of 700°C and an $O_2/Ar+O_2$ ratio of 80 %. The other conditions were fixed at a plasma power of 100 W, a working pressure of 10 mTorr, and a growth time of 1 hr.

The surface properties of ZnO films were measured by the non-contact type mode of atomic-force microscope (AFM). The bulk properties of ZnO films were measured by wide-range X-ray diffractometer (XRD) and high-resolution X-ray diffractometer (HRXRD). The θ-2θ scan of wide range XRD was used in order to investigate the intercompounds between ZnO films and substrates and the ZnO phases with different crystalline directions. The ω scan and φ scan of HRXRD were used in order to investigate the crystalline qualities, dislocation types, and epitaxial growth of ZnO films.

3 Results and discussion

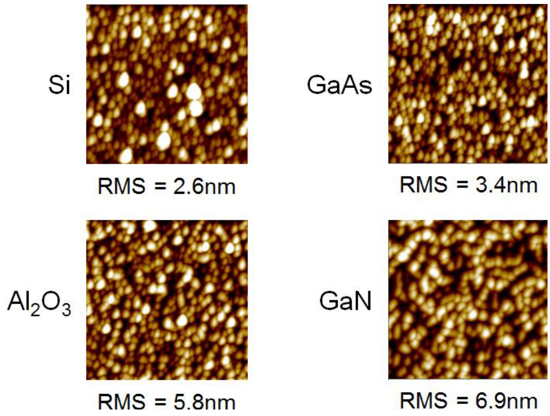

Fig. 1. Surface morphology and surface roughness (RMS) in the ZnO films grown on Si, GaAs, and Al_2O_3 substrates and a GaN template, respectively.

Figure 1 shows the surface morphology and surface roughness in the ZnO films grown on Si, GaAs, and Al_2O_3 substrates and a GaN template, respectively. All the ZnO films commonly have the similar surface morphology composed of nano-sized grains, which indicates a typical evidence of textured growth, though their details are dependent on each substrate. It is reported that the ZnO film with a preferred orientation of the c-axis has the textured surface, which has the nano-scaled surface structures of various pyramidal shapes due to the columnar growth along an axial direction [11,12]. Each ZnO film has the different grain size and surface roughness, which is ascribed to the fact that each optimized growth condition is different. It is known that i) the substrate with a different crystalline structure and a different surface state, ii) the substrate temperature that supplies the activation energy for ZnO film formation, iii) the working gas ratio that manipulates the stoichiometry of ZnO films, and iv) the working pressure and plasma power that control the growth rate of ZnO films by changing sputter yield give influences on the surface structures of the ZnO films [5,8,10]. On the other hand, hexagonal or triangular islands and step or terrace structures sometimes reported in the films grown by metal-organic chemical-vapor deposition (MOCVD) and molecular-beam epitaxy (MBE) were not observed in this research [13-16].

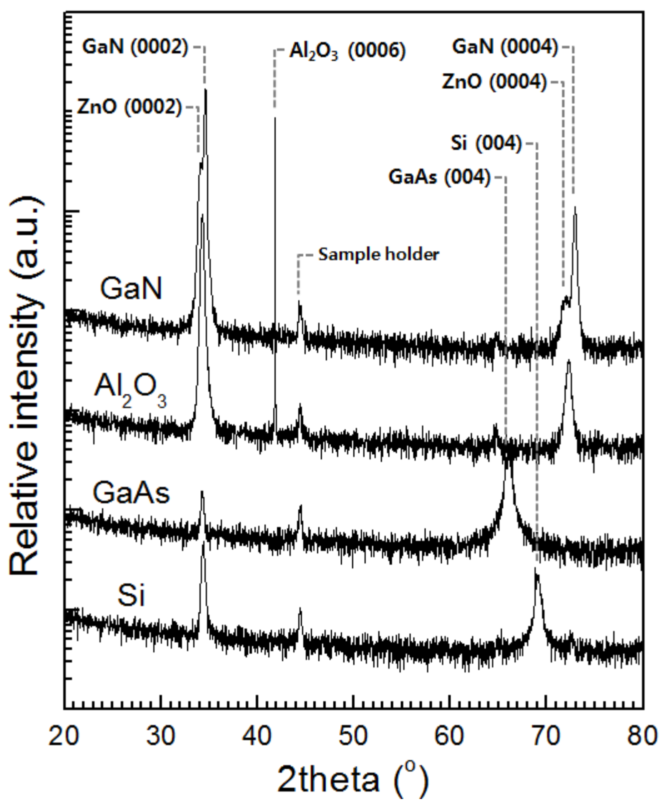

Fig. 2. X-ray θ-2θ scan diffraction curves in the wide range of 20 – 80°C in the ZnO films grown on Si, GaAs, and Al_2O_3 substrates and a GaN template, respectively.

Figure 2 shows the X-ray θ-2θ scan diffraction curves in the wide range of 20 – 80°C in the ZnO films grown on Si, GaAs, and Al_2O_3 substrates and a GaN template, respectively. The peak located at a low angle of 34.3° is assigned to the ZnO (0002) plane and the peak located at a high angle of 72.2° is assigned to the ZnO (0004) plane [5,8,10]. In the ZnO-on-Si a peak at 69.0° is due to the (004) plane of the Si substrate and in the ZnO-on-GaAs a peak at 65.9° is due to the (004) plane of the GaAs substrate. In the both samples, any other diffraction peaks do not exist beside the ZnO (0002) peaks and their crystal planes are parallel to the (004) planes of each substrate, which indicate that the ZnO films were grown with a preferred orientation of the c-axis. The peaks commonly found at 44.5° are due to a sample holder. In the ZnO-on-Al_2O_3 a peak at 41.8° is due to the (0006) plane of the Al_2O_3 substrate and in the ZnO-on-GaN the two peaks at 34.6° and 72.9° are due to the (0002) and (0004) planes of the GaN template, respectively. The peak separation between the ZnO film and the Al_2O_3 substrate is estimated to be 7.5°, which corresponds to the strain of 20.1 %. The peak separations between the ZnO film and the GaN template are estimated to be 0.48° in the (0002) plane and 0.96° in the (0004) plane, which corresponds to the strains of 1.35 and 1.16 %, respectively. In the both samples, the two ZnO films grown on the Al_2O_3 substrate and GaN template have larger diffraction intensities than those on the Si and GaAs substrates and the two ZnO films grown on the Al_2O_3 substrate and GaN template have the diffraction peaks for (0004) planes that are not exposed in those on the Si and GaAs substrates. This is ascribed to the fact that the film and substrate have the similar structure in the former while they have the different structure in the latter, which indicates that the ZnO-on-Al_2O_3 and the ZnO-on-GaN have the improved crystalline qualities, compared to the ZnO-on-Si and the ZnO-on-GaAs.

Figure 3 shows the X-ray φ-scan diffraction curves for asymmetric (10-11) planes in the ZnO films grown on Si, GaAs, and Al_2O_3 substrates and a GaN template, respectively. The φ-scan is that the sample surface rotates as a function of azimuthal angle whose axis is normal to the sample surface, while the ω scan (or θ-2θ scan) is that the sample surface rotates as a function of incident angle of X-ray beam whose axis is parallel to the sample surface [17,18]. In the cases of ZnO-on-Si and ZnO-on-GaAs there no diffraction peaks exist, while in the cases of ZnO-on-Al_2O_3 and ZnO-on-GaN there 6 diffraction peaks as a period of 60° exist. The latter is due to the six-fold symmetry of ZnO lattices, which indicates that the grown ZnO films are single domain epitaxial films with one orientations, while the former is due to the random distribution of various domains with different orientations, which indicates that the ZnO lattices are not randomly stacked along the a-axis, though they are well grown along the c-axis [17,18].

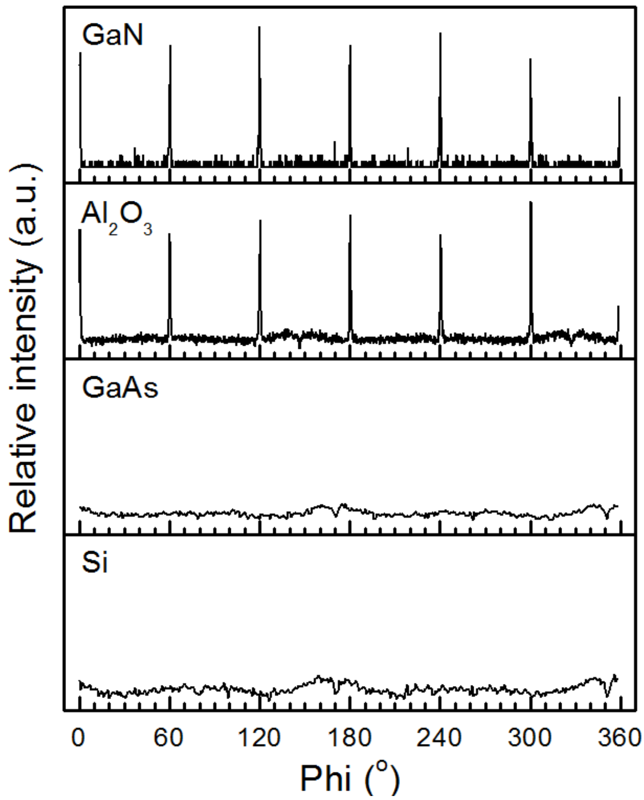

Fig. 3. X-ray φ-scan diffraction curves for asymmetric (10-11) planes in the ZnO films grown on Si, GaAs, and Al_2O_3 substrates and a GaN template, respectively.

Figure 4 shows the X-ray ω-scan diffraction curves for symmetric (0002) planes and asymmetric (10-11) planes in the ZnO films grown on Si, GaAs, and Al_2O_3 substrates and a GaN template, respectively. In the (0002) scan curves, the ZnO-on-GaN with a diffraction linewidth of 0.289° is narrower than the ZnO-on-Al_2O_3 with a diffraction linewidth of 0.480°. In the (10-11) scan curves, the ZnO-on-GaN with a diffraction linewidth of 0.643° is also narrower than the ZnO-on-Al_2O_3 with a linewidth of 0.789°. The ω scan for symmetric planes such as (0002) reflects screw dislocation densities because Burger's vector is parallel to the c-axis, while the ω scan for asymmetric planes such as (10-11) reflects edge dislocation densities because Burger's vector is normal to the c-axis [17,18]. Therefore, the ZnO-on-GaN has smaller edge dislocation density and smaller screw dislocation density than the ZnO-on-Al_2O_3, which is ascribed to the fact that the GaN lattice has a small lattice mismatch of 1.8 % with the ZnO lattice, while the Al_2O_3 lattice a large lattice mismatch of 18 % with the ZnO lattice [4-6]. This phenomenon was more clearly shown in the ZnO film growth at RT without the supply of surface reaction energy: the case of ZnO-on-Al_2O_3 had 5 times larger broadness than the case of ZnO-on-Al_2O_3 in the X-ray ω-scan diffraction curves of (0002) planes.

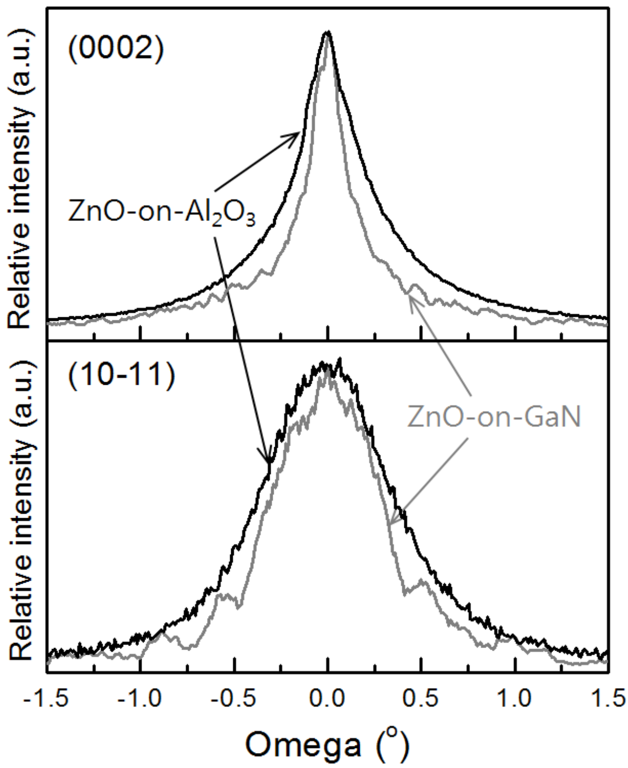

Fig. 4. X-ray ω-scan diffraction curves for symmetric (0002) planes and asymmetric (10-11) planes in the ZnO films grown on Si, GaAs, and Al_2O_3 substrates and a GaN template, respectively.

4 Conclusions

We had a comparative study about the structural properties in the surface and bulk of the ZnO films grown on (001) Si, (001) GaAs, and (0001) Al_2O_3 substrates and a (0001) GaN template by using RF-sputtering. First, all the ZnO films had textured structures with a preferred orientation of the c-axis, irrespective growth conditions. Second, the ZnO films in the ZnO-on-Si and ZnO-on-GaAs were consisted of various domains with different orientations, while the ZnO films in the ZnO-on-Al_2O_3 and ZnO-on-GaN were consisted of a single domain with the same orientation. Finally, the ZnO-on-GaN had smaller edge dislocation density and screw dislocation density than the ZnO-on-Al_2O_3, which seriously depended on substrate temperature.

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