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Growth of epitaxial ZnO thin films on lattice-matched buffer layer: Application of $InGaO_3(ZnO)_6$ single-crystalline thin film

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Abstract

Effects of lattice-matched substrates on growth of ZnO epitaxial films were studied. ZnO thin films were grown on single-crystalline $InGaO_3(ZnO)_6$ (IGZO) layers, which have small lattice mismatches of ~0.8% and ~2.2% in *a*- and *c*-axes, respectively. Epitaxial ZnO films were grown with the epitaxial relationship between the ZnO film and the single-crystalline IGZO of $[0001]_{ZnO}//[0001]_{IGZO}//[111]_{YSZ}$ and $[11\bar{2}0]_{ZnO}//[11\bar{2}0]_{IGZO}//[1\bar{1}0]_{YSZ}$. The use of the lattice-matched substrate and optimization of film microstructure and post-annealing condition led to atomically flat surfaces at maximum process temperatures as low as 700 °C. A large Hall electron mobility ~80 $cm^2\ (V\ s)^{-1}$ (N_e: ~2.8×10^{18} cm^{-3}) was obtained even if the film thickness was only 150 nm although comparable mobilities have been reported on films having much larger thicknesses (~1000 nm) fabricated at higher temperatures ~1000 °C.
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1. Introduction

Transparent oxide semiconductors (TOSs) are promising for optoelectronic devices because they exhibit simultaneously high optical transparency in visible region and good controllability of carrier concentration from $<10^{15}$ to 10^{21} cm^{-3} [1,2]. Concretely, optoelectronic devices such as transparent *pn* junction rectifier, ultraviolet light-emitting diode (UV-LED) and transparent field-effect transistors have been demonstrated to date [3–5]. Especially, zinc oxide (ZnO), a representative n-type TOS, has been intensively studied for developing high performance devices because of its good optoelectronic properties such as large electron mobility (>100 $cm^2\ (V\ s)^{-1}$ for bulk single-crystal) and sharp UV luminescence originating from room-temperature-stable exciton [6–8].

In general, it is known that device performance strongly depends on crystal quality of active layers including hetero-interface structures. Defects such as grain and/or domain boundaries may cause significant deterioration of device performance because carriers are trapped or scattered by the defects and carrier mobility is seriously reduced [9]. Therefore, it is vital to grow a high-quality epitaxial film without any boundaries to fabricate high performance devices. In addition, an atomically flat surface is more favorable to suppress the carrier scattering at a channel-gate insulator interface especially in field-effect transistors.

Use of a lattice-matched substrate is effective for growing high-quality epitaxial films because domain/grain boundaries are easily generated due to large lattice mismatching. Thus it is thought much favorable to use lattice-matched layer/substrate for growing high-quality epitaxial films.

Compounds in R_2O_3–M_2O_3–ZnO system (e.g., R=In, Sc and rare earth elements such as Lu; M=In, Ga, Al and Fe) represented by the chemical formula $RMO_3(ZnO)_m$

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(m=integer) are well-known as n-type TOSs [10,11]. They may offer a good lattice-matched material for ZnO because their crystal structures are similar to that of wurtzite-type ZnO. They have layered structures composed of RO_2^- and $MO^+(ZnO)_m$ layers stacked along the c-axis, in which the $MO^+(ZnO)_m$ layer has a distorted wurtzite-type structure. Recently we succeeded to develop a technique, reactive solid-phase epitaxy (R-SPE), to grow high-quality single-crystalline films of $RMO_3(ZnO)_m$ at high throughput and good reproductively [12–15]. The a-axis lattice constant of single-crystalline $InGaO_3(ZnO)_6$ (IGZO) is 0.328 nm, which corresponds to an in-plane lattice mismatch value of ~0.8% with ZnO (a-axis lattice constant is 0.324 nm, taken from JCPDS #36-1451) (Fig. 1(a)). Furthermore, the

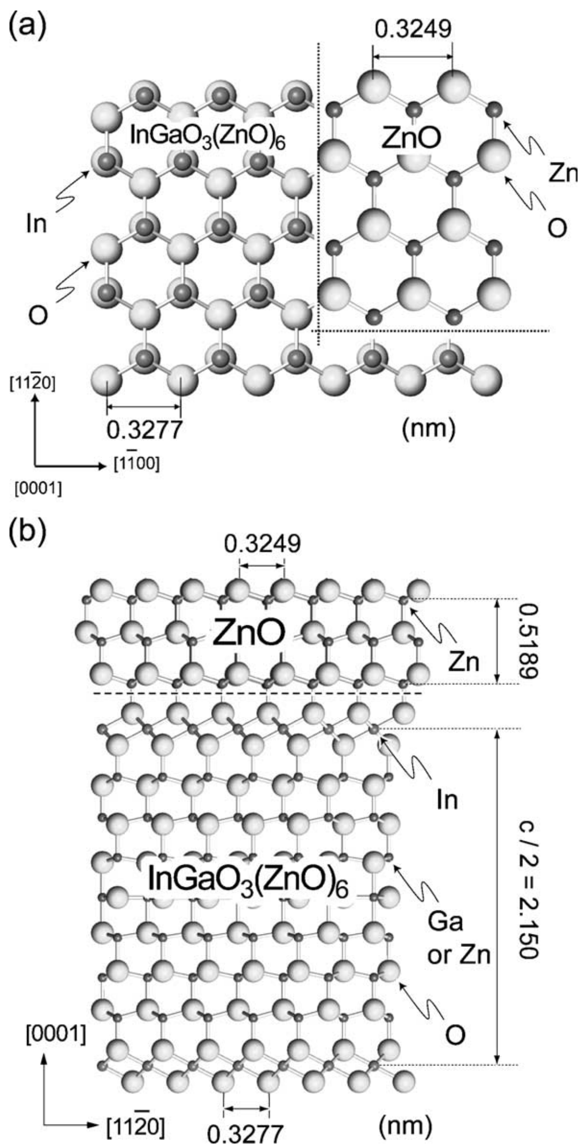

Fig. 1. Crystal structure of $InGaO_3(ZnO)_6$. (a) Atomic configurations of O and In in (0001) IGZO and O and Zn in (0001) ZnO. (b) Atomic configurations of O and In in $(11\bar{2}0)$ IGZO and O and Zn in $(11\bar{2}0)$ ZnO. The structure of ZnO epitaxial layer is illustrated on the top of the $InGaO_3(ZnO)_6$ layer assuming that the Zn^{2+} ions in the ZnO layer occupies tetrahedral Zn^{2+} sites at the topmost surface of the $InGaO_3(ZnO)_6$ layer.

surface of the single-crystalline IGZO film has an atomically flat terraces-and-steps structure, where the step height corresponds to the distance between adjacent InO_2^- layers (d_{0003}) in the IGZO crystal. The step height is ~2.1 nm, which matches with the height of 4 unit cells of ZnO (c-axis lattice constant is 0.5189 nm) (Fig. 1(b)).

Here, we report growth, structure and carrier transport properties of ZnO epitaxial thin films grown on the three-dimensionally lattice-matched IGZO substrates. The films with atomically flat surfaces and large Hall mobilities were achieved by thermal annealing of 600 °C-deposited films at temperatures as low as 700 °C. This mobility value is much improved from that reported on ZnO epitaxial films with similar thicknesses (~51 $cm^2 (V s)^{-1}$ for 250-nm-thick films annealed at 1000 °C [16]), indicating the present process is effective to reduce fabrication temperature by keeping large mobility and atomically flat surfaces.

2. Experimental details

The lattice-matched IGZO buffer layers were fabricated on YSZ (111) substrates by a reactive solid-phase epitaxy (R-SPE) (see Refs. [12] and [14] for detail). ZnO films were grown on the IGZO layers by a pulsed laser deposition (PLD) technique using a KrF excimer laser (wavelength=248 nm, pulse duration=20 ns, repetition frequency=10 Hz). The background pressure of the growth chamber was 1.0×10^{-6} Pa, while the oxygen pressure of 1.0×10^{-3} Pa was maintained during the film growth. The energy density of the laser pulse was ~0.5 J cm^{-2} $pulse^{-1}$. The thickness of the ZnO thin films was controlled to be ~150 nm. Crystalline quality and orientation were evaluated by high-resolution X-ray diffraction measurements (HR-XRD, ATX-G, Rigaku). Surface morphologies of the ZnO films were observed by an atomic force microscope (AFM, SPI-3800N, S.I.I.). Cross-sectional high-resolution transmission electron microscopy (HR-TEM) images were observed using EM-002B (TOPCON). Hall mobility and carrier concentration were measured by the van der Pauw method using an ac magnetic field modulation technique.

3. Results and discussion

We first examined the growth of ZnO thin films on the IGZO layers by PLD at temperatures varied from 600 °C to 900 °C. It was confirmed that all the ZnO films were heteroepitaxially grown on the IGZO layers with the epitaxial relationship; out-of-plane of $[0001]_{ZnO}//[0001]_{IGZO}$ and in-plane $[11\bar{2}0]_{ZnO}//[11\bar{2}0]_{IGZO}$.

Fig. 2 shows topographic AFM images of the ZnO thin films. Granular structures with lateral sizes less than 10 nm were seen in the ZnO films grown at temperatures as low as 600 °C. When the films are grown at >700 °C, on the other hand, hexagonal hillock grains with facets were

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