Research Article

The Achievement of a Zinc Oxide-Based Homojunction Diode Using Radio Frequency Magnetron Cosputtering System

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(Al + N)-codoped *p*-type zinc oxide (ZnO)/undoped *n*-type ZnO homojunction structure was deposited onto Si (100) substrate by using radio frequency (rf) magnetron cosputtering system. Transparent indium tin oxide (ITO)-ZnO cosputtered film was employed as the ohmic contact electrode to the *n*-type ZnO film, and the specific contact resistance was optimized to $2.9 \times 10^{-6} \Omega \text{ cm}^2$ after treating by a rapid thermal annealing (RTA) process at 400°C for 5 min under vacuum ambient. The ohmic contact behavior between the metallic Ni/Au and *p*-ZnO film also was improved to $3.5 \times 10^{-5} \Omega \text{ cm}^2$ after annealing at 300°C for 3 min under nitrogen ambient. The interfacial diffusion of these ohmic contact systems which led to the optimization of the specific contact resistances by the RTA process was investigated by the Auger electron spectroscopy (AES) depth profile measurements. The diode characteristics of the resulting *p*-ZnO/*n*-ZnO homojunction structure realized with these ohmic contact electrodes were confirmed by current-voltage (*I*-*V*) measurement, which performed a forward turn-on voltage of 1.44 V with a reverse current of $1.1 \times 10^{-5} \text{ A}$ at -2 V .

1. Introduction

Zinc oxide (ZnO) with a wide and direct band gap ($\sim 3.37 \text{ eV}$) is one of the most promising materials used in short-wavelength optoelectronics applications [1–5]. The excellent physical and chemical properties, resistance to radiation damage, and possibility of wet-etching processes for the ZnO material are expected to be brighter than the current state-of-the-art gallium nitride-based (GaN-based) material. Besides, the critical advantage of the ZnO over GaN material is the technologies required for depositing quality ZnO films, such as sputtering, pulse laser deposition, and hydrothermal method, are relatively simple and cost effective. Unfortunately, since ZnO material is natural *n*-type as a consequence of the compensation effect associated with the intrinsic oxygen vacancy (V_{O}) and zinc interstitial (Z_{I}) donors, the preparation of the *p*-type ZnO film with a high carriers is comparatively difficult, thereby limiting the development of the ZnO-based homojunction device. As a result,

the heterojunction structure using other quality *p*-type materials, such as ZnTe, Cu_2O , and GaN [6–8], currently becomes the main research target. While most ZnO-based optoelectronic devices achieved from the heterojunction structure are in principle superior to those employing homojunction structure, because of the obstacles in obtaining quality and stable *p*-type ZnO with high conductivity and mobility, there still has been much progress in the preparation of the ZnO-based homojunction structure especially for the light emitting diode applications. Until now, although there have been some reports on the achievement of a ZnO-based homojunction structure fabrication using metalorganic chemical vapor deposition, molecular beam epitaxy, pulse laser deposition, atomic layer deposition, or hydrothermal method [9–12], few studies have realized ZnO film-based homojunction structure purely employing sputtering technology, which has the availability of large area deposition at a relative low temperature [13, 14]. In our previous study, we demonstrated an (Al + N)-codoped *p*-type ZnO film

with a hole concentration higher than 10^{18} cm^{-3} using the radio frequency (rf) magnetron cosputtering system [15], which is beneficial for developing ZnO-based homojunction structure. Except for the achievement of a quality *p*-type ZnO film, it also is essential to optimize the ohmic contact electrodes to *n*-type and *p*-type layers, respectively, since the major loss in device performance is frequently due to high-resistance ohmic contact in case of *p-n* homojunction structure. For the electrode contact to *n*-type ZnO layer, most metal structures show ohmic contact to *n*-type ZnO with a specific contact resistance lower than $10^{-5} \Omega\text{-cm}^2$ [16–18]. Furthermore, a transparent conductive oxide (TCO) film also is employed as ohmic contact electrode to substantially enhance the device transparency [19, 20]. By contrast, similar to the metal structures ohmic contact to *p*-GaN, such as Ni/Au, Pt/Au, Pd/Au, Pt/Ni/Au, and Pd/Ni/Au [21–25], only a few bilayer and multilayer metallization schemes based on high work function metals are adaptive to obtain ohmic contact behavior as contact to *p*-type ZnO. The metal structures incorporated with Ni thus have been currently employed as a promising ohmic contact electrode to phosphorous-doped *p*-type ZnO layer owing to the favorable transformation of the Ni to *p*-NiO during annealing process [26–28].

In this study, with the aim to prepare a ZnO-based homojunction structure with quality diode performance purely using the sputtering technology, the contact resistances for the transparent cosputtered electrode contact to *n*-type ZnO and Ni/Au metallic system contact to (Al + N) codoped *p*-type ZnO films, respectively, were firstly optimized by the rapid thermal annealing (RTA) process. The mechanism responsible for the ohmic contact formation was investigated and discussed. A quality ZnO-based homojunction diode based on these ohmic contact systems thus was achieved as evidence of its rectifying property.

2. Experiment

A 300 nm thick, undoped ZnO layer was deposited on the silicon substrate by a radio frequency (rf) magnetron cosputtering system at room temperature and then annealed at 700°C for 30 minutes under oxygen ambient to improve the crystalline structure (hereafter denoted as *n*-ZnO). A 200 nm thick, cosputtered indium tin oxide (ITO)-ZnO film at an atomic ratio of 33% [$\text{Zn}/(\text{Zn} + \text{In}) \text{ at.}\%$] was deposited on the *n*-ZnO layer as the ohmic contact electrode using the transmission-line method (TLM). The detailed deposition parameters to prepare the cosputtered ITO-ZnO film by the rf magnetron cosputtering system, using ITO and ZnO targets, are available elsewhere [29]. Meanwhile, a 500 nm thick (Al + N) codoped ZnO film at a theoretical Al atomic ratio of 10% [$\text{Al}/(\text{Al} + \text{Zn}) \text{ at.}\%$] also was deposited onto another set of the silicon substrates by the rf magnetron cosputtering system at room temperature, using ZnO and aluminum nitride (AlN) targets, and then annealed at 450°C for 30 min under nitrogen ambient to activate the doping acceptors (hereafter denoted as *p*-ZnO) [15]. Ni/Au (30 nm/10 nm) metallic system was evaporated on the *p*-ZnO film using the TLM. All the contact patterns were formed through standard lift-off technology.

FIGURE 1: Schematic structure of the *p*-ZnO/*n*-ZnO homojunction diode.

With the aim to optimize the contact resistance, the ITO-ZnO/*n*-ZnO and Au/Ni/*p*-ZnO contact systems were then processed by a rapid thermal annealing (RTA) treatment at temperatures ranging from 100°C to 500°C under vacuum and nitrogen ambient, respectively. Finally, a *p*-ZnO/*n*-ZnO homojunction structure was fabricated by employing these ITO-ZnO/*n*-ZnO and Au/Ni/*p*-ZnO ohmic contact systems, as shown in Figure 1.

Film thickness was confirmed by a surface profile system (Dektak 6M). The carrier concentration and Hall mobility of the *n*-ZnO, *p*-ZnO, and ITO-ZnO films were measured using the van der Pauw method (Ecopia HMS-5000) at room temperature. The radiative emission of the *n*-ZnO and *p*-ZnO layers was determined from the photoluminescence (PL) spectra measured at room temperature using the He-Cd laser ($\lambda = 325 \text{ nm}$) as the excitation source. Auger electron spectroscopy (AES) depth profiles of the Ni/Au metallic electrode contact to *p*-ZnO film after the RTA treatment were performed on a scanning Auger nanoprobe (Ulvac-PHI, PHI 700). The current-voltage (*I-V*) properties of the ITO-ZnO/*n*-ZnO and Au/Ni/*p*-ZnO contact systems as well as the *p*-ZnO/*n*-ZnO homojunction diode were measured using a semiconductor parameter analyzer (HP4156C).

3. Results and Discussions

Table 1 illustrates the electrical properties of the as-deposited, annealed ZnO, and (Al + N) codoped ZnO films and the ITO-ZnO cosputtered film. The as-deposited ZnO film behaved as an insulator with a resistivity higher than $10^5 \Omega\text{ cm}$, whereas the as-deposited (Al + N) codoped ZnO film exhibited *n*-type conduction with electron carriers of $2.4 \times 10^{19} \text{ cm}^{-3}$ due to the activation of the Al_{Zn} donors [30]. The annealed ZnO film showed *n*-type conduction with an electron concentration of $3.1 \times 10^{16} \text{ cm}^{-3}$ and a mobility of $19.1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ as a consequence of the native donors activation and crystalline growth. By contrast, owing to the fact that the annealing process on the (Al + N) codoped ZnO film was favorable to activate the N_{O} acceptors, the (Al + N) codoped ZnO film converted into *p*-type conduction with a high hole concentration of $1.9 \times 10^{18} \text{ cm}^{-3}$. In addition, the cosputtered ITO-ZnO film employed as the transparent ohmic contact electrode to the *n*-ZnO layer behaved as an *n*-type degenerated semiconductor

TABLE 1: Electrical properties of the as-deposited, annealed ZnO, and (Al + N) codoped ZnO films, and the ITO-ZnO cosputtered film.

Sample	Concentration (/cm ³)	Mobility (cm ² /V _s)	Resistivity (Ω cm)	Type
As-deposited ZnO	N/A	N/A	>10 ⁵	N/A
Annealed ZnO	3.1×10^{16}	19.1	10.6	<i>n</i>
As-deposited (Al + N) codoped ZnO	2.4×10^{19}	1.2	0.2	<i>n</i>
Annealed (Al + N) codoped ZnO	1.9×10^{18}	3.4	0.9	<i>p</i>
ITO-ZnO	8.2×10^{20}	13.5	5.6×10^{-4}	<i>n</i>

FIGURE 2: Room temperature PL spectra of the (a) as-deposited and (b) annealed ZnO films.

FIGURE 3: Room temperature PL spectra of the (a) as-deposited and (b) annealed (Al + N) codoped ZnO films.

with a low resistivity of $5.6 \times 10^{-4} \Omega \text{ cm}$. Figures 2(a) and 2(b) show the room temperature PL (RTPL) spectra of the as-deposited and annealed ZnO films, respectively. For the PL spectrum of the as-deposited ZnO film, two broad emissions were observed. The UV and blue luminescence emerged from the near band edge emission (NBE) located at 3.24 eV and the radiation associated with the native shallow defect transitions of zinc vacancy (V_{Zn}) and Zn_I at approximately

3.05 and 2.90 eV, respectively, while the deep level emission at green-yellow wavelength ($\sim 2.10 \text{ eV}$) was attributed to the native V_O transition [31–33]. As the sample annealed at 700°C under oxygen ambient, the radiative transitions that resulted from the above-mentioned native defects almost were absent in the PL spectrum, as shown in Figure 2(b), revealing the improvement on the crystal structure. The PL spectrum therefore was dominated by the NBE emission. The RTPL

FIGURE 4: I - V curves of the transparent ITO-ZnO cosputtered electrode contacts to the n -ZnO layer as a function of the annealing temperatures for 1 min under vacuum ambient.

FIGURE 5: Specific contact resistance of the 400°C-annealed ITO-ZnO/ n -ZnO contact system as a function of the annealing time under vacuum ambient (the inset figure shows the I - V curves of the contact system annealed for 5 min).

spectra of the as-deposited and annealed (Al + N) codoped ZnO films are shown in Figures 3(a) and 3(b). Compared to the as-deposited ZnO film, the spectrum of the as-deposited (Al + N) codoped ZnO film shown in Figure 3(a) composed a sharp UV emission, a weak blue emission, and a broad tail extending to long wavelength. In agreement with the reports [30, 34, 35], the UV and blue emissions approximately at 3.18 eV and 2.86 eV were, respectively, identified as the NBE and $\text{Al}_{\text{Zn}}\text{-V}_{\text{Zn}}$ radiative transitions. For the codoped film annealed at 450°C under nitrogen ambient, another radiation located at about 1.87 eV predominated over the PL spectrum other than the NBE emission. This red luminescence was ascribed to be the $\text{N}_{\text{O}}\text{-V}_{\text{O}}$ radiative transition originated from the activation of the N_{O} acceptors and thereby caused the codoped film exhibited p -type conduction.

FIGURE 6: I - V curves of the Ni/Au metallic electrode contact to the as-deposited and annealed (Al + N) codoped ZnO film.

Figure 4 shows the I - V curves of the transparent ITO-ZnO cosputtered electrode contacts to n -ZnO layer as a function of the annealing temperatures for 1 min under vacuum ambient, measured from the contact spacing of 15 μm . The as-deposited ITO-ZnO/ n -ZnO contact system exhibited the rectifying property, while a linear I - V characteristic was achieved from the contact system annealed at 200°C. The ohmic contact behavior of the ITO-ZnO/ n -ZnO contact system was improved with increasing the annealed temperature and a lowest specific contact resistance approximately of $4.1 \times 10^{-5} \Omega \text{ cm}^2$ was obtained as the contact system annealed at 400°C. The specific contact resistance of the 400°C-annealed ITO-ZnO/ n -ZnO contact system as a function of the annealing time under vacuum ambient is highlighted in Figure 5 (the inset figure shows the I - V curves of the contact system annealed for 5 min). It can be seen that the specific contact resistance of the ITO-ZnO/ n -ZnO system was gradually improved with the annealing time increasing and then decreased significantly as the annealing time reached 7 min. The ohmic contact resistance of the ITO-ZnO/ n -ZnO system, as shown in the inset figure, can be further optimized to $2.9 \times 10^{-6} \Omega \text{ cm}^2$ after annealing at 400°C for 5 min under vacuum ambient. The appearance of the homologous $\text{Zn}_2\text{In}_2\text{O}_5$ compounds between the ITO-ZnO/ n -ZnO interfaces as a consequence of the outdiffusion of the oxygen atoms was demonstrated to be responsible for the ohmic contact optimization [36]. The I - V curves of the Ni/Au metallic electrode contact to the as-deposited and annealed (Al + N) codoped ZnO film, respectively, are showed in Figure 6. Since the as-deposited (Al + N) codoped ZnO film exhibited n -type conduction the Ni/Au metallic system with a high work function contacted to the n -type (Al + N) codoped ZnO film thus showed the rectifying property with low leakage current. By contrast, the Ni/Au electrode contacted to the annealed (Al + N) codoped ZnO film which exhibited p -type conduction with a high hole concentration performed ohmic contact behavior with a specific contact resistance $2.7 \times 10^{-3} \Omega \text{ cm}^2$. With the aim to further improve the contact performance, the Au/Ni/ p -ZnO contact system

FIGURE 7: (a) I - V characteristic and (b) specific contact resistance of the Ni/Au metallic electrode contacts to the p -ZnO layer as a function of the annealing temperatures for 1 min under nitrogen ambient.

was processed by the RTA treatment. Figure 7(a) shows the I - V characteristic of the Ni/Au metallic electrode contacts to the p -ZnO film as a function of the annealing temperatures for 1 min under nitrogen ambient. It can be found that the slope of these I - V curves is increased with the annealing temperature raising, indicating the reduction in the contact resistance. The corresponding specific contact resistances of these annealed Au/Ni/ p -ZnO contact systems are illustrate in Figure 7(b). The lowest specific contact resistance of $3.5 \times 10^{-5} \Omega \text{ cm}^2$ was obtained from the 300°C-annealed Au/Ni/ p -ZnO contact system, and then this value was apparently increased to $1.7 \Omega \text{ cm}^2$ for the sample annealed at 400°C for 1 min under nitrogen ambient, revealing that the annealed temperature was crucial to optimize the Ni/Au metallic electrode ohmic contact to the p -ZnO film. An investigation on the interfaces between the metallic Ni/Au and p -ZnO layer therefore was carried out to elucidate the evolutions of the specific contact resistance on the RTA treatment. Figures 8(a)–8(c), respectively, give the elemental depth profiles of the as-deposited, 300°C-, and 400°C-annealed Au/Ni/ p -ZnO contact system, conducted by AES measurements. For the as-deposited sample (Figure 8(a)), the Au, Ni, and p -ZnO layers are well-defined and a little interdiffusion was observed from the Au/Ni and Ni/ p -ZnO interfaces. The interlayer between Ni and p -ZnO was presumed to form p -type NiO [37–40] and thus caused the as-deposited Au/Ni/ p -ZnO contact system to possess the ohmic contact behavior as shown in Figure 6. By contrast, in the case of the sample annealed at 300°C, the outdiffusion of Ni and O through Au layer is clearly observed in Figure 8(b), indicating that the RTA treatment on the sample is greatly favorable to the formation of p -type NiO layer at the surface. Accordingly, an obvious reduction in the contact resistance as compared to the as-deposited sample was obtained from the 300°C-annealed Au/Ni/ p -ZnO contact system. When the sample annealed

at 400°C, the outdiffusion of O is even more remarkable and no individual Ni layer can be observed in Figure 8(c). It is noteworthy that though the appearance of the O and Ni atoms is beneficial for the formation of p -type NiO and thus improved the Au/Ni/ p -ZnO ohmic contact behavior, the oxygen atoms that outdiffuses from the p -ZnO layer surface also is likely to induce the V_O donors and led to the decrease in the hole concentration at the p -ZnO surface. As a result, apparently degradation on the specific contact resistance was obtained from the 400°C-annealed Au/Ni/ p -ZnO contact system which implied an exceeding outdiffusion of the O atoms at the p -ZnO surface. The I - V characteristic of the p -ZnO/ n -ZnO homojunction structure using the Ni/Au and ITO-ZnO ohmic contact electrode is illustrated in Figure 9. The ohmic contacts of the Au/Ni/ p -ZnO and ITO-ZnO/ n -ZnO systems confirmed by the fairly linear I - V relationship are given in the inset figure. A diode behavior was achieved from the p -ZnO/ n -ZnO homojunction structure prepared using the rf magnetron cosputtering system. The forward turn-on voltage appeared at about 1.44 eV and yielded a rectifying ratio approximately of 20 at ± 2 V. The ideality factor, n , extracted from the logarithmic plot of this I - V curve by adopting the thermionic theory was about 3.8. Since such p -ZnO/ n -ZnO homojunction structure was achieved purely by sputtering technology, the dominant current transparent mechanism which resulted in the large deviation from the idea case ($n = 1$) thus was ascribed to be the defects in the interface and imperfections in the formation of the abrupt p - n homojunction originated from the ion-bombardment damage during film deposition. Accordingly, no electroluminescence was observed, possibly because of no radiative recombination emerging from the above-mentioned transitions at the interface of the homojunction structure which also showed evidence of the low forward turn-on voltage.

FIGURE 8: Elemental depth profiles of the (a) as-deposited, (b) 300°C-annealed, and (c) 400°C-annealed Au/Ni/p-ZnO contact systems, conducted by AES measurements.

4. Conclusions

The *n*-ZnO with dominated NBE emission and *p*-ZnO with dominated NBE emission and N_O - V_O deep level radiation at 1.87 eV was produced using the rf magnetron cosputtering system, followed by an adequate annealing process. For the transparent ITO-ZnO/*n*-ZnO contact system annealed at 400°C for 1 min under vacuum ambient, the specific contact resistance was optimized to $2.9 \times 10^{-6} \Omega \text{ cm}^2$ due to the formation of the homologous $Zn_2In_2O_5$ compounds associated with the outdiffusion of the oxygen atoms at the *n*-ZnO layer surface. By contrast, the Au/Ni/*p*-ZnO contact system was optimized at a specific contact resistance of

$3.5 \times 10^{-5} \Omega \text{ cm}^2$ after annealing at 300°C for 1 min under nitrogen ambient. The mechanism responsible for the improvement of the Au/Ni/*p*-ZnO contact system by an RTA treatment was attributed to the *p*-NiO interlayer formed between Ni and *p*-ZnO interface, whereas the degradation on the contact resistance was ascribed to be the reduction in the hole carriers at the *p*-ZnO surface as a consequence of the considerable outdiffusion of the oxygen atoms. By taking advantage of these ohmic contacts' behavior, a quality diode performance with a forward turn-on voltage of 1.44 eV and a reverse current of $1.1 \times 10^{-5} \text{ A}$ at -2 V was therefore obtained from the *p*-ZnO/*n*-ZnO homojunction structure prepared purely using the rf magnetron cosputtering system.

FIGURE 9: I - V characteristic of the p -ZnO/ n -ZnO homojunction structure (the ohmic contacts of the Au/Ni/ p -ZnO and ITO-ZnO/ n -ZnO systems are given in the inset figure).

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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