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Electrical properties and interfacial reactions of rapidly annealed Ni/Ru Schottky rectifiers on n-type GaN

M. Siva Pratap Reddy [a], V. Rajagopal Reddy [a,*], Chel-Jong Choi [b]

[a] Department of Physics, Sri Venkateswara University, Tirupati 517502, India
[b] School of Semiconductor and Chemical Engineering, Semiconductor Physics Research Center (SPRC), Chonbuk National University, Jeonju 561-756, Republic of Korea

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ABSTRACT

The effect of annealing temperature on the electrical and structural properties of Ni/Ru Schottky rectifiers have been investigated by current–voltage (*I–V*), capacitance–voltage (*C–V*), X-ray diffraction (XRD) and secondary ion mass spectrometer (SIMS) measurements. The measured Schottky barrier height for as-deposited Ni/Ru/n-GaN Schottky diode is 0.66 eV (*I–V*) and 0.79 eV (*C–V*). It is observed that the barrier height of the Ni/Ru Schottky contacts increases with an increase in annealing temperature. When the contact is annealed at 600 °C for 1 min in N_2 ambient, a maximum barrier height is achieved and the corresponding values are 0.79 eV (*I–V*) and 0.98 eV (*C–V*). It is observed that significant improvement in the electrical properties of Ni/Ru Schottky rectifier after annealing temperatures as compared to the as-deposited contact. Based on the results of SIMS and XRD, the formation of gallide phases at the Ni/Ru/n-GaN interface could be the reason for the increase of Schottky barrier height at elevated temperatures. Atomic force microscopy results showed that the surface morphology of the Ni/Ru Schottky contact is fairly smooth with a root-mean-square (RMS) roughness of 2.12 nm even after annealing at 600 °C. These results indicate that the Ni/Ru contacts could be attractive for high-temperature device applications.

1. Introduction

Recently, rapid progress has been found in the fabrication of GaN-based optoelectronic and microelectronic devices such as GaN-based light-emitting diodes [LEDs] [1], laser diodes [LDs] [2], Solar-blind MSM-photodetectors [3], metal–semiconductor field effect transistor [MESFETs] [4], modulation-doped field effect transistor [MODFETs] [5], heterostructure field effect transistors [HFETs] [6] and high electron mobility transistors [HEMTs] [7,8]. Especially, the performance and reliability of these devices have been improved with high quality ohmic and Schottky rectifiers. However, larger leakage current through Schottky rectifiers adversely affects the operation, power consumption, noise and reliability of devices. Hence, Schottky rectifier for GaN with ultra low leakage current is one of the challenges to their use in optoelectronic and electronic devices. Therefore, it is necessary to obtain a high Schottky barrier height at the metal/semiconductor interface to reduce the leakage current and to improve break down voltage in Schottky diodes.

Many researchers have explored various metal schemes for the fabrication of Schottky contacts on n-type GaN, e.g., Rh/Au [9], Ir/Pt [10], Au and Ti/Au [11], Rh [12], Ru/Au [13], Pt [14], Pd, Pt and Ni [15], Au [16], Al/Ni/Au [17], Pt/Mo [18], Ni/AlN/GaN/AlN [19], Ni [20], Ni/Mo [21], Ir/Pt [22], etc. Dobos et al. [11] investigated the structural and electrical properties of Au and Ti/Au contacts to n-GaN and found that the contacts were rectifying up to 700 °C and the highest barrier height of 1.07 eV was obtained for Au single layer. They also observed that the barrier height of the contacts decreased due to the formation of several intermetallic phases after annealing at 900 °C. Tian and Chor [12] studied the electrical and thermal stability of Rh-based Schottky contacts on n-GaN. They found that the Ni/Rh/Au contact exhibits better electrical performance, including a higher Schottky barrier height, a lower leakage current and better thermal stability compared to the Rh/Au and Ni/Au contacts. Reddy et al. [13] investigated the electrical and structural properties of Ru/Au/n-GaN Schottky contacts at different temperatures. They showed that the formation of interfacial gallide phases at the interface could be the reason for the enhancement of barrier height at elevated temperatures. Fang et al. [17] studied the thermal behaviour of Al/Ni/Au multilayer Schottky contacts on n-GaN. They found that the Al/Ni/Au contact exhibits high quality Schottky contact with a barrier height of 0.875 eV and the lowest reverse-bias leakage current respectively after annealing at 450 °C for 12 min in N_2 ambient. Reddy et al. [18] investigated the electrical and structural properties of Pt/Mo Schottky contacts to n-GaN as a function of annealing temperature. They showed that the

* Corresponding author.
E-mail address: reddy_vrg@rediffmail.com (V.R. Reddy).

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Pt/Mo Schottky contacts exhibits better electrical characteristics after annealing at elevated temperatures. Chuah et al. [19] studied the electrical properties of Ni/AlN/GaN/AlN Schottky photodiode in the temperature range of 500–700 °C. They showed that the barrier height increased with an increasing in annealing temperature. Menard et al. [20] fabricated Ni-based Schottky diodes with different thickness (20 nm, 100 nm and 300 nm) on n-type GaN. They reported that the Ni Schottky contact with a 300 nm thickness shows good rectifying behaviour after annealing at 450 °C during 3 min under Argon. Recently, Jyothi et al. [21] studied the electrical and structural characteristics of Ni/Mo Schottky rectifiers on n-GaN in the temperature range 300–600 °C. They reported that the Ni/Mo Schottky rectifier exhibits excellent electrical properties even after annealing at 600 °C. Very recently, Chang et al. [22] fabricated GaN-based Schottky barrier photodiode with Ir/Pt metallization scheme. They found that the barrier height increased from 0.91 eV to 1.03 eV and the ideality factor decreased from 1.58 to 1.16 after annealing at 600 °C in O_2 atmosphere. The interfacial reactions at the metal–semiconductor junction play a vital role in evaluating the Schottky barrier heights. Schottky barrier height is highly interface sensitive, however, the exact nature of metal/semiconductor interface is still not completely understood. In this work, Nickel (Ni) has been selected as first layer because it has high work function metal 5.15 eV as well as good adhesion and reactivity with GaN. In the present work, we have investigated the electrical, structural and morphological characteristics of Ni/Ru Schottky rectifiers on n-type GaN ($\sim 4.07 \times 10^{17}$ cm^{-3}) as a function of annealing temperature.

2. Experimental details

2 μm thick Si-doped GaN films used in this study is grown by metalorganic chemical vapor deposition (MOCVD) on c-plane Al_2O_3 sapphire substrate. The carrier concentration obtained by means of Hall measurements (Lake Shore Model-7604) is $\sim 4.07 \times 10^{17}$ cm^{-3}. Prior to metal deposition, the n-GaN layer is first ultrasonically degreased with warm trichloroethylene, acetone and methanol for 5 min each step. This degreased layer is then dipped into boiling aquaregia [HNO_3:HCl = 1:3] for 10 min to remove the surface oxides and then rinsed in deionized water. Ti (25 nm)/Al (100 nm) metals are deposited on a portion of the sample as ohmic contact and the contacts annealed at 650 °C in N_2 ambient for 3 min. Then a thin layer of Schottky metal consisting of Ni (20 nm)/Ru (30 nm) is deposited through a stainless steel mask with a diameter of 0.7 mm under a vacuum pressure of 4×10^{-6} Torr by electron beam evaporation system. The Ni/Ru Schottky rectifiers are sequentially annealed at various temperatures from 300 °C to 600 °C for duration of 1 min in N_2 ambient in a rapid thermal annealing (RTA) system. Current–voltage (*I*–*V*) and capacitance–voltage (*C*–*V*) characteristics of the Schottky diode are measured by Keithley source measuring unit (Model No 2400) and automated DLTS system (DLS-83D) at room temperature. Secondary ion mass spectrometer (SIMS) (Model No: IMS 6F, CAMECA, France) is performed to examine the intermixing of the metals and n-GaN before and after annealing. X-ray diffractometer (Siefert, XRD PW 3710) using Cu Kα radiation is used to characterize the interfacial reactions between the Ni/Ru and GaN layers. The surface morphology of the Ni/Ru Schottky contacts is analyzed by atomic force microscopy (AFM) (tip size < 10 nm) before and after annealing.

The barrier heights (ϕ_b) and ideality factors (n) are obtained from the forward *J*–*V* characteristics according to the thermionic emission theory and is given by [23]

$$J = J_o \exp\left\{\frac{qV}{(nkT)}\right\} \quad (1)$$

$$J_o = A^* T^2 \exp\left\{\frac{-q\phi_b}{(kT)}\right\} \quad (2)$$

where J_o is the saturation current density, n is the ideality factor, k is the Boltzmann's constant, T is the absolute temperature, ϕ_b is the barrier height and A^* is the effective Richardson coefficient. The value of ϕ_b can be deduced directly from the *I*–*V* curves if the effective Richardson's constant A^* is known. The theoretical value of A^* is 26.4 A cm^{-2} K^{-2} based on the effective mass ($m^* = 0.22\, m_o$) of n-GaN [24] and is used here to deduce ϕ_b. The plot of ln *J* versus *V* will give a straight line with a slope of $q/(nkT)$, the intercept with *y*-axis will yield J_o and subsequently barrier height ϕ_b can be obtained using Eq. (2). The ideality factor (n) is determined from the forward characteristics using the relation

$$n = \left(\frac{q}{kT}\right)\left[\frac{dV}{d(\ln J)}\right] \quad (3)$$

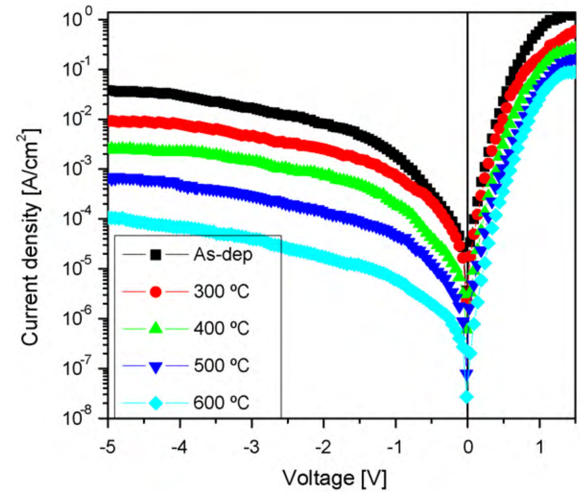

Fig. 1. The reverse and forward *J*–*V* characteristics of the Ni/Ru Schottky contacts on n-type GaN as a function of annealing temperature.

The Norde method is also employed to compare the Schottky barrier height of Ni/Ru contacts because high series resistance can hinder accurate evolution of barrier height from the standard ln (*J*)–*V* plot [25]. In this method, a function *F*(*V*) is plotted against the *V*. The function *F*(*V*) is given by

$$F(V) = \frac{V}{2} - \frac{kT}{q} \ln\left[\frac{I(V)}{AA^{**}T^2}\right] \quad (4)$$

The effective Schottky barrier height is given by

$$\phi_b = F(V_{min}) + \frac{V_{min}}{2} - \frac{kT}{q} \quad (5)$$

where $F(V_{min})$ is the minimum value of $F(V)$ and V_{min} is the corresponding voltage.

The capacitance–voltage (*C*–*V*) relationship for Schottky contact is given by [26]

$$\frac{1}{C^2} = \frac{2(V_{bi} - (kT/q) - V)}{A^2 q N_d \varepsilon_s} \quad (6)$$

where ε_s is the permittivity of the semiconductor ($\varepsilon_s = 9.5\varepsilon_o$), V is the applied voltage and A is the surface area of the diode. The *x*-intercept of the plot of ($1/C^2$) against V is V_o and related to built-in potential V_{bi} by the equation $V_{bi} = V_o + kT/q$, where T is the absolute temperature. The barrier height (ϕ_b) is given by $\phi_b = V_{bi} + V_n$, where $V_n = (kT/q)\ln(N_c/N_d)$. The density of states in the conduction band edge is given by $N_c = 2(2\pi m^* kT/h^2)^{3/2}$, where $m^* = 0.22 m_o$, and its value was 2.6×10^{18} cm^{-3} for GaN at room temperature [27].

3. Results and discussion

The *J*–*V* characteristics of Ni/Ru Schottky contacts are measured as a function of annealing temperature by keeping the sample in a tight box at room temperature. Fig. 1 shows the semilog plot of *J*–*V* characteristics of the Ni/Ru/n-GaN Schottky diodes. The measured reverse-bias leakage current density is about 1.96×10^{-3} A/cm^2 at −1 V for the as-deposited sample. For the samples annealed at 300 °C, 400 °C, 500 °C and 600 °C the leakage current densities are 7.92×10^{-4} A/cm^2, 2.05×10^{-4} A/cm^2, 4.84×10^{-5} A/cm^2 and 6.21×10^{-6} A/cm^2, respectively. It is observed that the leakage current densities decrease with an increase in annealing temperature. This indicates the improvement in electrical characteristics of Ni/Ru/n-GaN Schottky diodes. Calculations show that the Schottky barrier height (SBH) of Ni/Ru Schottky contacts is 0.66 eV for as-deposited contact. The extracted barrier heights of annealed Ni/Ru Schottky contacts are 0.68 eV for 300 °C, 0.72 eV for 400 °C, 0.73 eV for 500 °C and 0.79 eV for 600 °C. These measurements showed that the barrier height increases with an increase in annealing temperature. The ideality factor for the as-deposited Ni/Ru Schottky diode is found to be 1.48. However, the ideality factor decreased to 1.14 upon annealing at 600 °C. The forward current voltage characteristics in each case showed an ideality factor greater than one. This may be due to the transport mechanisms other than thermionic

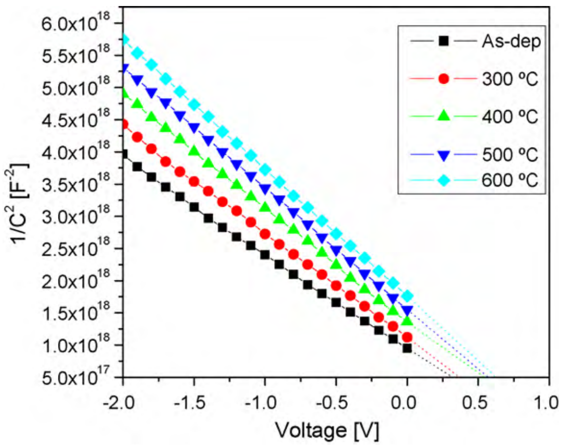

Fig. 2. Plot of $1/C^2$ versus V for the Ni/Ru Schottky contacts annealed at different temperatures.

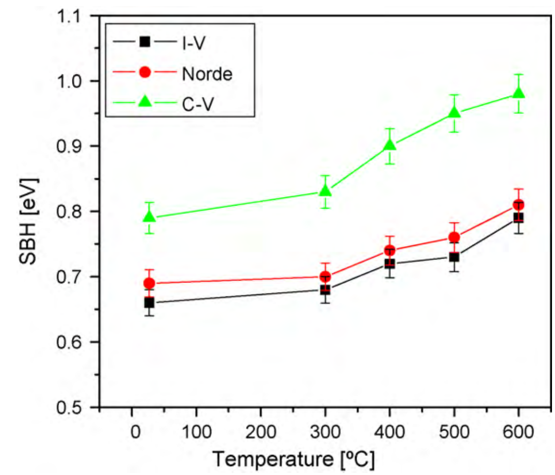

Fig. 3. Plot of barrier heights versus annealing temperatures for the Ni/Ru Schottky contacts on n-type GaN.

emission, such as recombination. Another possibility, independent of the surface preparation the ideality factor (n) decreases, tending to the ideal behaviour predicted by the thermionic emission theory [26]. The series resistance can be estimated from I–V measurements using the method developed by Cheung and Cheung [28]. The calculated series resistance is in the range of $R_s = 445–40\,\Omega$ for the as-deposited and annealed Ni/Ru Schottky contacts. The Norde method is also used to calculate the Schottky barrier height of Ni/Ru Schottky contacts. The extracted Schottky barrier heights are 0.69 eV for as-deposited, 0.70 eV for 300 °C, 0.74 eV for 400 °C, 0.76 eV for 500 °C and 0.81 eV for 600 °C, respectively. These values are in good agreement with those obtained by the I–V method.

Capacitance–voltage (C–V) characteristics of Ni/Ru Schottky contacts are measured as a function of annealing temperature. Fig. 2 shows a plot of $1/C^2$ as a function of bias voltage V at 1 MHz for as-deposited and annealed Ni/Ru Schottky contacts. From the slope of the graph the carrier concentrations (N_d) are calculated and the corresponding values are 9.9×10^{17} cm^{-3} for as-deposited, 9.0×10^{17} cm^{-3} for 300 °C, 8.5×10^{17} cm^{-3} for 400 °C, 7.9×10^{17} cm^{-3} for 500 °C and 7.6×10^{17} cm^{-3} for 600 °C samples. These values are similar to those obtained from the Hall measurement by the Vander Pauw method. In C–V measurements, the net doping concentration is the difference of the electrically active concentration of donors and acceptors. In case of n-type material, the concentration of donors outbalances the concentration of acceptors. If Schottky contacts are leaky, the leakage current will disturb the capacitance measurements and consequently there may be error in estimation of the net doping concentration. This may be one of the reasons for the carrier densities calculated from C–V slightly to be more or less value compared to the value measured from Hall measurements. The measured barrier height of the as-deposited Schottky contact is 0.79 eV. For the samples annealed at 300 °C, 400 °C, 500 °C and 600 °C the Schottky barrier heights are 0.83 eV, 0.90 eV, 0.95 eV and 0.98 eV, respectively. The obtained values of reverse leakage current densities, SBHs and ideality factors of Ni/Ru Schottky diodes as a function of annealing temperature are shown in Table 1. Based on the I–V, Norde and C–V methods, the variation in the Schottky barrier height of Ni/Ru contacts after annealing suggests that Ni/Ru films may react with the GaN, as will be confirmed by SIMS and XRD measurements.

Fig. 3 shows the plot of barrier heights of Ni/Ru/n-GaN Schottky diode as a function of annealing temperature. It can be seen from the plots that the barrier height of the as-deposited Ni/Ru Schottky contact increases with annealing temperature. From Fig. 3, it is evident that the barrier heights ϕ_b obtained from I–V measurements are lower than those obtained from C–V measurements. The difference may be due to the formation of an interfacial layer containing defects. Thus interfacial capacitance and capacitance due to depletion layer are in series thereby decrease in the total capacitance and as a result, C^{-2} increases. This increases the intercept of C^{-2} versus V plot and increases the barrier height. This may be due to the transport mechanism in these diodes and it is not purely thermionic emission in nature. Another possibility is lowering of barrier height by the image force due to current flow across the barrier [29]. The barrier height determined from the I–V method will logically yield lower barrier heights than barrier heights values from C–V method, since the I–V method involves the flow of electrons form semiconductor to metal.

In order to characterize the interfacial reactions between the Ni/Ru and n-GaN layers, SIMS measurement is performed. Fig. 4 shows the SIMS depth profiles of the Ni/Ru Schottky contacts before and after annealing at 600 °C. For the as-deposited Ni/Ru Schottky contact, Fig. 4(a), there is an interface region between the Ni/Ru and the GaN films. It indicates that the possible reaction of Ni/Ru metals with GaN during deposition. It is observed that the both the Ga and N secondary ions have signal overlap with the Ni and Ru secondary ions. For the contact annealed at 400 °C, some amount of Ga out-diffused into the Ni/Ru metal layers as shown in Fig. 4(b). This indicates the possibility of Ga reacts with Ni and Ru, as a result the formation of Ga–Ni and Ga–Ru interfacial phases at the interface.

Table 1
The leakage current densities, Schottky barrier heights (ϕ_b) and ideality factor (n) of Ni/Ru Schottky diodes on n-GaN as a function of annealing temperature.

Sample	Leakage current densities at −1 V	Schottky barrier height (SBH) ϕ_b (eV)			Ideality factor 'n'
		I–V	Norde	C–V	
As-dep	1.96×10^{-3}	0.66(±0.020)	0.69(±0.021)	0.79(±0.024)	1.48(±0.044)
300 °C	7.93×10^{-4}	0.68(±0.020)	0.70(±0.021)	0.83(±0.025)	1.39(±0.041)
400 °C	2.05×10^{-4}	0.72(±0.022)	0.74(±0.022)	0.90(±0.027)	1.26(±0.038)
500 °C	4.86×10^{-5}	0.73(±0.022)	0.76(±0.023)	0.95(±0.029)	1.24(±0.037)
600 °C	6.21×10^{-6}	0.79(±0.023)	0.81(±0.024)	0.98(±0.029)	1.14(±0.034)

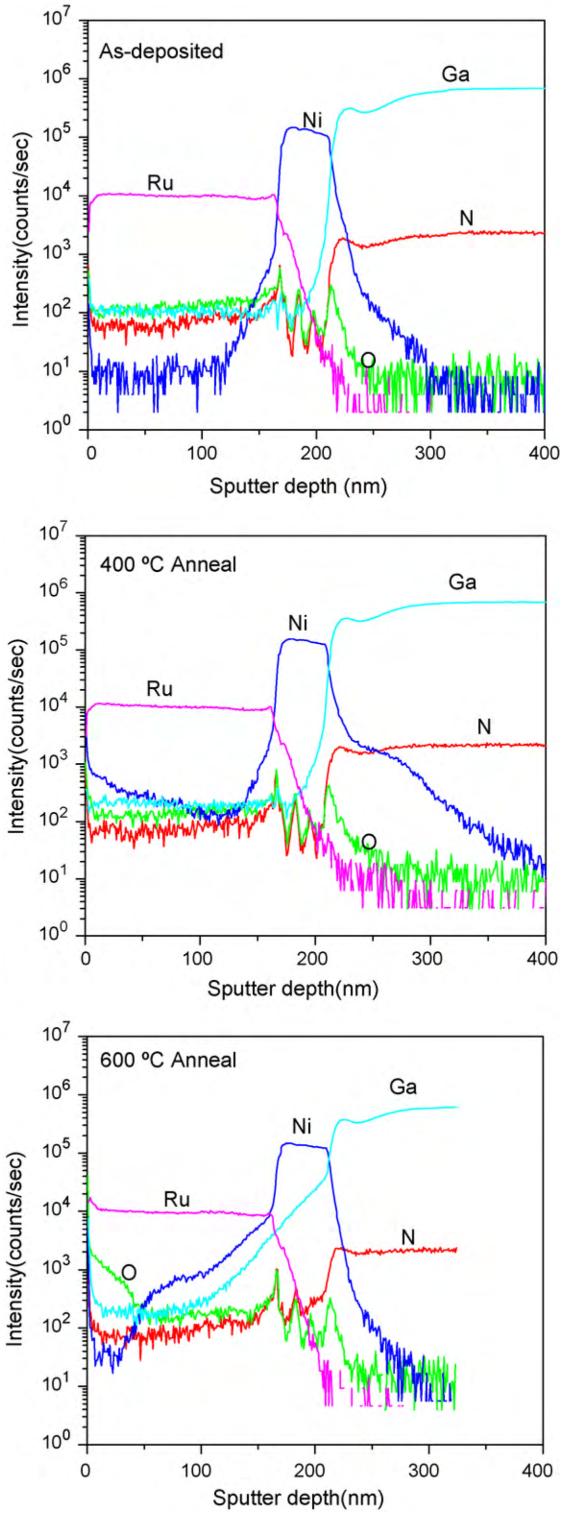

Fig. 4. SIMS depth profiles of the Ni/Ru films on n-type GaN: (a) as-deposited (b) annealed at 400 °C and (c) annealed at 600 °C.

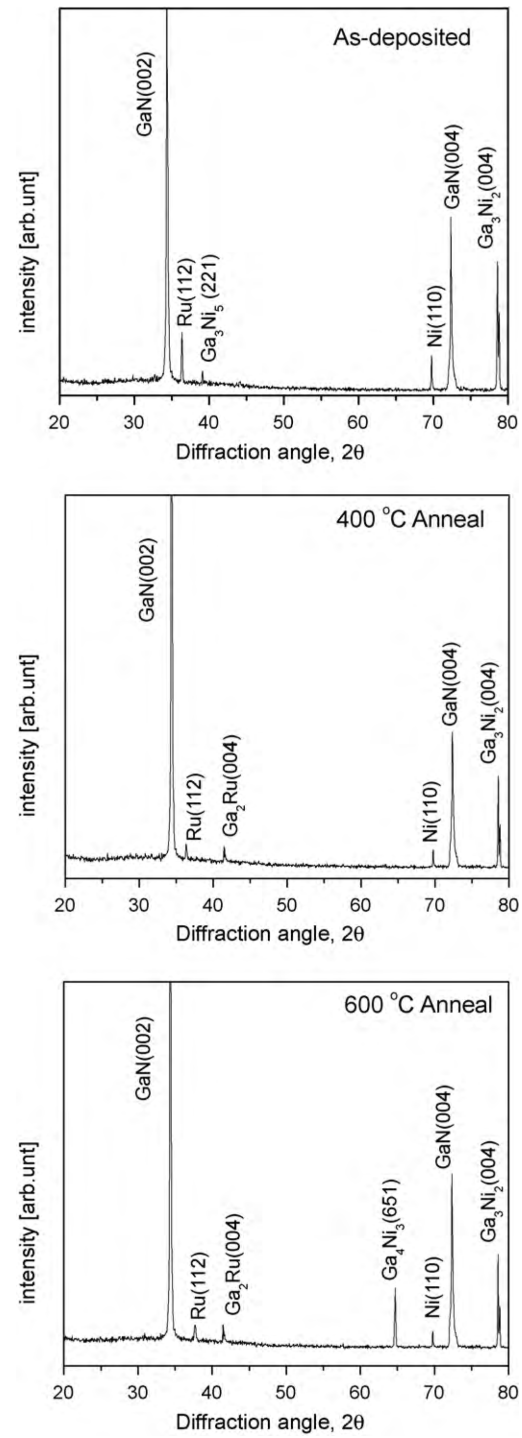

Fig. 5. XRD plots of the Ni/Ru Schottky contacts on n-type GaN: (a) as-deposited (b) annealed at 400 °C and (c) annealed at 600 °C.

When the contact is annealed at temperature 600 °C (Fig. 4(c)), a considerable change in the interface is observed with further out-diffusion of Ga into Ni/Ru films as compared to the contact annealed at 400 °C. It is further noted that there is no clear evidence for out-diffusion of nitrogen into the Ni/Ru layers. Moreover, it is noted that a small amount of oxygen is observed in the interface for all contacts, which may have partially originates from the GaN surface.

In order to investigate the interfacial products which were formed between the metals and GaN before and after annealing at 600 °C, XRD examination is made. Fig. 5(a) shows the XRD plot of the as-deposited sample. In addition to the characteristic peaks of GaN (0 0 2) (0 0 4), Ni (1 1 0), Ru (1 1 2), there are extra peaks which are identified as Ga_3Ni_5 (2 2 1) and Ga_3Ni_2 (0 0 4). For the contact annealed at 400 °C, Fig. 5(b), extra peak is observed as compared to the as-deposited contact. This peak is identified as Ga_2Ru (0 0 4), which indicate the formation of new interfacial phases. When the sample annealed at 600 °C, there is extra peak observed, which is

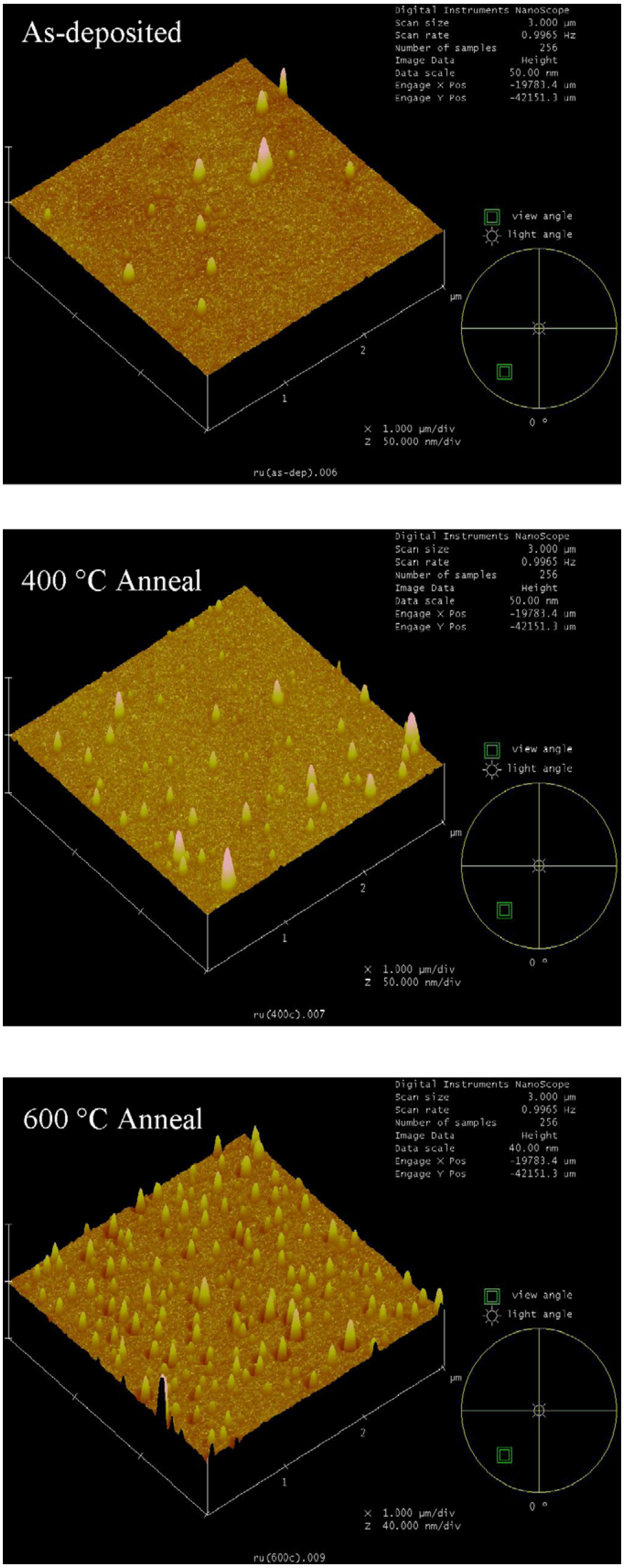

Fig. 6. AFM micrographs of the Ni/Ru Schottky contacts on n-type GaN: (a) as-deposited (b) annealed at 400 °C and (c) annealed at 600 °C.

identified as Ga_4Ni_3 (6 5 1) as expected from SIMS depth profile (Fig. 4(c)).

To investigate the surface morphology of the Ni/Ru Schottky contact, AFM is employed. Fig. 6 shows the AFM images of the samples before and after annealing temperature at 600 °C. The surface morphology of as-deposited sample is fairly smooth with a root-mean-square (RMS) roughness of 1.43 nm (Fig. 6(a)). When the contact is annealed at 400 °C for 1 min in nitrogen ambient, the surface morphology of the Ni/Ru contact is slightly decreased with a RMS roughness of 1.99 nm (Fig. 6(b)) as compared with that of the as-deposited contact. After annealing at temperature 600 °C, the surface morphology of the Ni/Ru contact is slightly decreased further with a RMS roughness of 2.12 nm (Fig. 6(c)). This indicates that the overall surface morphology of Ni/Ru contacts is fairly smooth even after annealing at 600 °C. It is also observed that the Schottky barrier height increases with increasing islands size (either Ni or Ru islands) upon annealing at 600 °C as compared to the as-deposited contact. According to Yang et al. [30] the barrier height decreases with the ratio of the island-area-to-island-periphery. Decreasing barrier heights with decreasing island-area-to-island-periphery ratio have been reported and it is due to increases in recombination in small islands due to increased electric fields at their edges.

It is known that the interface states and chemical reactions between metals and semiconductor interfaces can play an important role in the electrical behaviour of devices. Based on the SIMS and XRD results, the improved barrier height of the Ni/Ru Schottky contact upon annealing temperature could be explained as follows. The SIMS and XRD results showed that the out-diffusion of Ga from the GaN layer into the Ni/Ru layers and participate in the formation of interfacial gallide phases at the interface. As a result, there is an accumulation of Ga vacancies at the GaN surface. The formation of gallide phases at the interface such as Ga_3Ni_5 (2 2 1), Ga_3Ni_2 (0 0 4), Ga_2Ru (0 0 4), Ga_4Ni_3 (6 5 1) (as shown by XRD results Fig. 5(a)–(c)) causes an increase in the barrier height of the Ni/Ru Schottky contact upon annealing temperature. As the reduction of non-stoichiometric defects in the metallurgical interface [31,32] may also be the reason in the increase of Schottky barrier height. The region involving the defects could be reduced due to the interdiffusion of metals in GaN. Accordingly, the consumption of the defect region is followed by an increase in the value of the barrier height calculated from *I–V* characteristics upon annealing temperature. Similar results were also reported by Wang et al. [33], Jyothi et al. [21] and Rao et al. [34]. Wang et al. [33] studied the thermal annealing behaviour of Pt/n-GaN and they observed that the variation of barrier height upon annealing may be attributed to changes of surface morphology and variation of non-stoichiometric defects in the vicinity of the interface. Jyothi et al. [21] studied the barrier height as a function of annealing temperature and reported that the barrier height could be influenced by the interfacial products. Rao et al. [34] reported that the formation of gallide phases at the interface could be the reason for increase of Schottky barrier heights upon annealing temperature. The formation of gallide phases may create Ga vacancies in the GaN at the interface and the Ga vacancies in n-GaN [35] acts as deep acceptors which cause the increase of barrier height upon annealing temperature.

4. Conclusions

We have investigated the electrical and structural properties of Ni/Ru Schottky rectifier on n-type GaN as a function of annealing temperature by *I–V*, *C–V*, SIMS and XRD measurements. Measurements showed that the barrier height of the as-deposited contact is 0.66 eV (*I–V*) and 0.79 eV (*C–V*). However, it is observed that the Schottky barrier height Ni/Ru Schottky rectifiers increase with increasing in annealing temperatures. A maximum barrier height is obtained after annealing at 600 °C and the corresponding values are 0.79 eV (*I–V*) and 0.98 eV (*C–V*). The Norde method is also used to calculate the barrier heights of Ni/Ru Schottky rectifiers and the values are 0.69 eV for as-deposited, 0.70 eV for 300 °C, 0.74 eV for 400 °C, 0.76 eV for 500 °C and 0.81 eV for 600 °C. These values are in good agreement with those obtained by the *I–V* technique. It is noted that the electrical characteristics of Ni/Ru Schottky rectifiers are improved upon annealing temperatures. The SIMS and XRD results showed the formation of Ga–Ni and Ga–Ru interfacial phases at interface at elevated temperatures. This may be the reason for the increasing in the Schottky barrier height and a corresponding reduction in the reverse leakage current. The AFM results showed that the overall surface morphology of Ni/Ru Schottky contacts on n-GaN is fairly smooth. These results indicate that the Ni/Ru Schottky contact could be a useful rectifier for the fabrication of high-temperature device applications.

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