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Preparation of Crack-Free Al_xGa_{1-x}N Films with High Al Composition for Schottky-Type UV Detectors

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High-quality $Al_xGa_{1-x}N$ layers with an Al composition of x=~0.4 were prepared on a GaN/sapphire substrate by a combination of controlling the growth kinetics, resulting in a slow growth rate of 5 mm/min, and introducing a stress-compensating thin AlGaN interlayer between the $Al_xGa_{1-x}N$ and GaN layers. To control the growth kinetic activity, the reactor pressure, TMAI/TMGa flow rate, and H₂/NH₃ flow rate were all adjusted to obtain less-defective and homogeneous $Al_xGa_{1-x}N$ layers with high Al compositions, and optimized at a reactor pressure of 50 torr, TMAI/TMGa flow rate of 70/40 μ mol/min, and H₂/NH₃ flow rate of 6/6 slpm. As a result, the sequential growth of an $Al_xGa_{1-x}N$ layer on a 10-nm-thick AlGaN interlayer grown at 700°C on a 2.0- μ m-thick GaN layer finally produced a crack-free, less defective, homogeneous $Al_{0.33}Ga_{0.67}N$ film with 0.5 μ m thickness. Thereafter, a typical Schottky diode of Pt/ $Al_{0.33}Ga_{0.67}N/LT$ -AlGaN interlayer/n⁺-GaN exhibited a promising electro-optical sensitivity, including a reverse leakage current of 1 nA at -5 V, UV-visible extinction ratio of ~10⁴, and responsivity of 150 mA/W at a radiation wavelength of 280 nm.

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1. Introduction

Schottky barrier-type ultraviolet (UV)-sensitive diodes are attractive as UV photosensors due to their promising performances such as a high UV/visible rejection ratio, low noise, short response time, and easy fabrication process. In particular, since their cutoff optical band was expected tunable from 200 nm to 365 nm by simply controlling the Al mole fraction x in the Al_xGa_{1-x}N layer, diodes based on an Al_xGa_{1-x}N alloy have been considered to be most suitable for application in a solar-blind UV detector requiring a high responsivity within a UV band of 280 ~ 320 nm without any additional filters.⁽¹⁾ The solar blindness of an Al_xGa_{1-x}N-based optical sensor would be essential for the reliable detection of a flame or missile, even under harsh conditions, such as a background of strong daylight.

For the development of high-performance UV photosensors, the growth of thick, crack-free $Al_xGa_{1-x}N$ films with a high Al composition of > 20% was a prerequisite and so, a series of reports on the growth of thick, crack-free $Al_xGa_{1-x}N$ films by inserting either a low-temperature AlN interlayer⁽²⁾ or a low-temperature $Al_xGa_{1-x}N$ interlayer⁽³⁾ between the $Al_xGa_{1-x}N$ and GaN layers were made. In addition, since the growth rate of AlGaN film and the Al incorporation efficiency into the (Ga, N) layer were largely dependent on the parasitic reactions between trimethylaluminum (TMAl) and ammonia (NH₃), some research groups have devised and tested to resolve the prereaction problems using a special gas-injection scheme,⁽⁴⁾ short inlet-susceptor separation system,⁽⁵⁾ and high-gas-velocity method.⁽⁶⁾ However, the reliable control of quality and Al composition is still a task to be resolved mainly due to the large lattice and thermal mismatch of the heterojunction between the $Al_xGa_{1-x}N$ and GaN films.

In this work, high-performance UV sensors are investigated by adjusting the key growth parameters of the MOCVD process as well as by introducing an ultrathin AlGaN interlayer for a high-quality $Al_xGa_{1-x}N$ layer with a high Al concentration. As a result, highly promising Schottky-type diodes were fabricated on an MOCVD-grown $Al_xGa_{1-x}N$ layer surface and evaluated applicable to a visible-blind, UV-sensitive photosensor.

2. Experimental

The Al_xGa_{1-x}N films were grown on a 2" c-plane (0001) sapphire substrate using the metal-organic chemical vapor deposition (MOCVD) system with a high-speed rotatingdisk reactor. Trimethylgallium (TMGa), trimethylaluminum (TMAl), and high-purity ammonia (NH₃) were used as the precursors for the Ga, Al, and N species, respectively. The n-type Si-doping was carried out by employing silane gas and hydrogen (H₂) was used as a carrier gas. For the successful growth of high-quality Al_xGa_{1-x}N layers, the key growth parameters were optimized by tuning the reactor pressure between 50 torr and 150 torr, the TMGa-flow-rate between 30 and 60 μ mol/min, and the H₂/NH₃-flow-rate between 3/3 slpm and 6/6 slpm. Based on the optimized growth conditions, two kinds of device structures, that is, two-layer samples of Al_xGa_{1-x}N/n⁺-GaN and three-layer samples of Al_xGa_{1-x}N/LT-Al_xGa_{1-x}N interlayer/ n⁺-GaN, were grown on the sapphire substrate and tested for potential use as better Schottky-type UV detectors. Al concentration was calculated from the separation between the GaN and Al_xGa_{1-x}N peaks in the high-resolution X-ray diffraction (HRXRD) spectra. The electrical properties of the layer structures were investigated by Hall-effect measurements, the thickness by scanning electron microscopy (SEM), and the surface morphology by atomic force microscopy (AFM). After fabricating the Schottky barrier-type devices, the optical responsivity was then characterized using a commercially available optical testing bench within a band region from UV to visible.

3. Results and discussion

Figure 1(a) shows the series of HRXRD spectra recorded from the two-layer samples of $Al_xGa_{1-x}N/n^+$ -GaN (1.1 μ m thick) when the reactor pressure was 50, 100, and 150 torr with



Fig. 1. (a) HRXRD spectra for less homogeneous $Al_xGa_{1-x}N$ epilayers grown on GaN/sapphire under different operating pressures (150, 100, and 50 torr) and (b) calculated Al-composition/ thickness versus operating pressures for the growth time of 100 min.

a fixed TMGa/TMAI flow rate of 105/70 μ mol. The separation of the GaN and Al_xGa_{1-x}N peaks became systematically wider when the reactor pressure was increased for the same growth period of 100 min. Since AlN has a smaller lattice constant compared with GaN, the sharp peak at a diffraction angle of $\sim 17.06^{\circ}$ corresponds to the characteristic of a highquality underlying GaN layer, while the complex peak structure of Al_xGa_{1,x}N at higher diffraction angles represents the variation in the Al composition, x within the layer. On the assumption of a single peak, the calculated Al composition was approximately 14% for the 50 torr sample, 15% for the 100 torr sample, and 18% for the 150 torr sample, as shown in Fig. 1(b). In the experiment, the Al incorporation of 18% into the (Ga, N) layer seemed to reach a saturation level under the growth conditions with a reactor pressure over 150 torr. At higher operating pressures than 150 torr, the prereaction between the TMAI and NH_3 reactants⁽⁷⁾ can be a main cause of the limited incorporation of Al into the (Ga, N) layer. On the same basis, the significant reduction of Al_xGa_{1,x}N film thickness from 1.3 μ m at 50 torr to 0.4 μ m at 150 torr (Fig. 1(b)) can be understood as a combined result of the enhanced parasitic reaction between TMAI and NH₃ and the decrease in growth kinetic energy⁽⁷⁻¹⁰⁾ due to a reduced gas velocity. As the reactor pressure decreased from 150 to 50 torr, average surface roughness of the $Al_xGa_{1-x}N$ layer also decreased from 1.5 to 0.4 nm with a distinct reduction in the small pits and pit line densities associated with pure edge dislocation and defects.^(11,12) Thus, in addition to the strain in the Al_xGa_{1-x}N layer, the growth kinetic activity depending on the reactor pressure was proved to be an important factor in determining the degree of Al incorporation and defect generations.

To further increase the Al incorporation into a smooth Al_xGa_{1-x}N layer, the TMAl flow rate was adjusted to 70 μ mol/min and 105 μ mol/min with a fixed reactor pressure of 50 torr. HRXRD measurements of the resulting Al_xGa_{1-x}N layers indicated that a simple increase in TMAI flow rate was found not efficient enough to enhance the Al incorporation due to the increased parasitic reaction between TMAI and NH₃ leading to adduct formation.^(9,10) Instead, the change in TMGa flow resulted in a significant change in the amount of Al incorporation. Fig. 2(a) displays the series of HRXRD spectra from the Al_xGa_{1-x}N layers grown on GaN as a function of TMGa flow rates of 60, 40, and 30 μ mol/min under a reactor pressure of 50 torr and TMAl flow rate of 70 μ mol/min. Only two main peaks were observed with a significantly increased separation, while the estimation indicated a marked increase in Al incorporation from 21% for a TMGa flow rate of 60 μ mol/min to 47% for 30 μ mol/min, as plotted in Fig. 2(b). Since the thickness of the grown Al_xGa_{1-x}N layers was decreased from 0.85 μ m for 60 μ mol/min to 0.4 μ m for a 30 μ mol/min, the growth kinetics would be much less active for lower TMGa flow rates and responsible for a slightly degraded surface morphology with a roughness from 0.6 nm for 60 μ mol/min to 1.8 nm for 30 μ mol/min. In spite of the increased surface roughness, the HRXRD spectra for the lower TMGa flow rates revealed a very homogeneous distribution of the incorporated Al species. After all, the activity of the growth kinetics is a key parameter for the reliable preparation of homogeneous $Al_xGa_{1-x}N$ films with a high Al composition of ~50%. To further improve the surface morphology, H₂/NH₃ flow rates were adjusted from 3/3 slpm to 6/6 slpm at a reactor pressure of 50 torr and TMAI/TMGa flow rates of 70/40 μ mol/ min. Compared to the H_2/NH_3 flow rate of 3/3 slpm revealing a high density of surface cracks, the 6/6 slpm resulted in a much improved surface morphology with an Al mole



Fig. 2. (a) HRXRD spectra for Al_xGa_{1-x}N/GaN layers grown at different TMGa/TMAl flow rates (60/70, 40/70, 30/70 μ mol/min) and (b) calculated Al composition/thickness versus TMGa/TMAl flow rates.

fraction of 44%. Surface roughness of $Al_xGa_{1-x}N$ layer on GaN was also decreased from 1.45 nm for 3/3 slpm to 0.95 nm for 6/6 slpm but surfaces cracks were not completely avoidable as shown in Fig. 3(a).

In addition to appropriate control of the growth kinetics, an introduction of a thin LT-AlGaN interlayer between the AlGaN and GaN layers was promising for the growth of device-quality layer structures appropriate for Schottky-type UV sensors as shown in Fig. 3(b); Al_{0.33}Ga_{0.67}N (0.5 μ m)/GaN surface (Fig. 3(a)) and Al_{0.33}Ga_{0.67}N (0.5 μ m)/10 nm LT-



Fig. 3. SEM photographs of (a) AlGaN/GaN layer and (b) AlGaN/10 nm LT-AlGaN interlayer/GaN layer

AlGaN/GaN surface (Fig. 3(b)). In practice, the undoped GaN layer was replaced with an n⁺-GaN layer for reliable ohmic contact and a crack-free Al_{0.33}Ga_{0.67}N active layer was grown up to a thickness of 0.5 μ m for sufficient UV light absorption. Since the AlGaN layer surface showed no more cracks, a 10-nm-thick LT-AlGaN interlayer between the Al_{0.33}Ga_{0.67}N and GaN layers was understood as an adequate buffer layer to reconcile substantial lattice and thermal mismatches between Al_xGa_{1-x}N and GaN and thereby, essentially cancelled out the compressive stress of the GaN layer via the tensile stress of the AlGaN layer.

Figure 4 shows a schematic cross section of the designed AlGaN photodetector structure. Top-illuminated Schottky-type mesa-structure photodiodes, with a thin (100 Å) Pt Schottky contact and Ti/Al/Ni/Au ohmic contact prepared by electron-beam evaporation, were designed and fabricated, as explained previously in detail.⁽¹³⁾ The photosensors had a circular AlGaN mesa structure with diameters of 500 – 1000 μ m. To form the bonding pad, an Au layer was deposited to a thickness of 2000 Å.

Figure 5 shows the I-V characteristics of the Schottky-type Al_{0.33}Ga_{0.67}N UV detector fabricated on both samples without (cracked surface, Fig. 5(a)) and with a 10 nm LT-AlGaN interlayer (crack-free, Fig. 5(b)). The specific resistivity was approximately $3.4 \times 10^{-4} \Omega$ -cm² for the n-type ohmic contacts and the forward series resistance was in a range of ~1 k\Omega for the fabricated diodes. Detailed analysis of the measured I-V curves showed much better electrical properties for the samples with an interlayer; Schottky barrier height of ~0.8 eV and ideality factor of 1.6 for thc Pt/AlGaN sample without an interlayer and Schottky barrier height of ~1.0 eV and an ideality factor of 1.1 for the Pt/AlGaN sample with an interlayer. The reverse leakage current of Fig. 5 was also highly comparable for two samples; 509 nA at –5V for the sample without an interlayer and 1 nA at –5V for the



Fig. 4. Schematic cross section of the designed device.



Fig. 5. I-V characteristics of (a)Pt/Al_{0.33}Ga_{0.66}N/GaN diode and (b)Pt/Al_{0.33}Ga_{0.66}N/LT-AlGaN/GaN diode.

sample with an interlayer. Under reverse-bias conditions, the leakage current of the diode sample with the thin LT-AlGaN interlayer was much smaller and more stable than that of the diode sample on the AlGaN layer without an interlayer. Since the HRXRD spectra of two AlGaN samples indicated a very similar crystal quality revealing a homogeneous incorporation of Al species in the (Ga, N) layers, the differences between the electrical properties can be explained by the additional leakage through the surface cracks within the Schottky contact for the sample without an interlayer. However, the Pt/AlGaN/GaN sample still maintains the I-V characteristics of a Schottky diode revealing a high degree of degradation compared with the Pt/AlGaN/LT-AlGaN/GaN sample. Consequently, we can conclude that a thin LT-AlGaN interlayer is a good buffer for adjusting high-thermal and lattice mismatches between the GaN and AlGaN layers.

Figure 6 shows the spectral responses of $Pt/Al_{0.33}Ga_{0.67}N$ UV photosensors with a diameter of 500 μ m under a Xenon lamp emitting a UV photon with a wavelength of 250~480 nm. Based on this optical measurement, the $Pt/Al_{0.33}Ga_{0.67}N$ diode with an interlayer produced a UV-visible extinction ratio of 1.5×10^4 and responsivity of 150 mA/W at a wavelength of 280 nm.

4. Conclusions

For the successful preparation of crack-free, less defective, homogeneous $Al_xGa_{1-x}N$ layers with a high Al composition, two thermodynamic growth parameters were combined; 1) introduction of a 10 nm LT-AlGaN interlayer for stress release between the $Al_xGa_{1-x}N$ and GaN layers and 2) lowering the growth rate to approximately 5 nm/min by adjusting three key parameters, namely, operation reactor pressure, TMAl/TMGa flow rate and H₂/NH₃ flow rate. Less defective and homogeneous $Al_xGa_{1-x}N$ epilayers revealing some cracks were repeatedly grown on a 2- μ m-thick GaN layer at a reactor pressure of 50 torr,



Fig. 6. Spectral responsivity of Pt/Al $_{0.33}$ Ga $_{0.66}$ N Schottky diode measured at radiation wavelength of 280 nm.

TMAI/TMGa flow rate of 70/40 μ mol/min, and H₂/NH₃ flow rate of 6/6 slpm. Then, the introduction of a 10-nm-thick AlGaN interlayer grown at 700°C on the 2.0- μ m-thick GaN layer proved to be sufficiently effective to release the compressive stress of the GaN layer and tensile stress of the Al_xGa_{1-x}N layer, resulting in a crack-free, less defective, homogeneous Al_{0.33}Ga_{0.67}N film with a thickness of 0.5 μ m. In conclusion, a typical Schottky diode consisting of Pt/Al_{0.33}Ga_{0.67}N/LT-AlGaN interlayer/n⁺-GaN was proved as a promising UV selective photosensor with a reverse leakage current of 1 nA at -5 V, a UV-visible extinction ratio of ~10⁴, and a responsivity of 150 mA/W at a radiation wavelength of 280 nm.

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