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Composition and interface quality control of AlGaN/GaN heterostructure and their 2DEG transport properties

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Abstract – The effects of NH_3 flow rate and reactor pressure on Al composition and the interface of AlGaN/GaN heterostructure were studied. Equilibrium partial pressure of Ga and Al over AlGaN alloy was calculated as a function of growth pressure, NH_3 flow rate and temperature. It was found equilibrium vapor pressure of Al is significantly lower than that of Ga, thus, the alloy composition mainly controlled by Ga partial pressure. We believe that more decomposition of Ga occur at lower NH_3 flow rate and higher growth pressure leads to preferred Al incorporation into AlGaN. The alloy composition gradient became larger at AlGaN/GaN heterointerface at higher reactor pressures, higher Al composition and low NH_3 flow rate. This composition gradient lowered sheet carrier concentration and electron mobility as well. We obtained an AlGaN/GaN heterostructure with sheet carrier density of $\sim 2\times 10^{13}$ cm^{-2} and mobility of 1250 and 5000 cm^2/Vs at 300 K and 100 K, respectively.

I. Introduction

The AlGaN alloy are used as electron barrier and wave guide cladding layers for LED [1] and LD [2] and UV detector [4] where relatively low Al concentration are needed in these AlGaN layers. Thus the growth of AlGaN with low composition is relatively easy. A structure of particular interest for the electronic devices is the AlGaN/GaN two-dimensional electron gas (2DEG) for use in high electron mobility transistors (HEMTs) with applications for high voltage, high power operation at microwave frequencies [5-9]. These devices need high sheet carrier density and high mobility for high device performance to obtain large channel currents, breakdown voltages and cut off frequencies and high output powers. To obtain high sheet carrier density and high mobility in AlGaN/GaN heterostructure, it must growth AlGaN with higher Al content with carefully control the interface quality. It is reported that MOCVD growth pressure and ammonia flow rate profoundly influences the growth rate and alloy composition of AlGaN film [10-15]. In this work Al solid incorporation behavior was investigated systematically at different growth pressure and ammonia flow rate. Equilibrium vapor pressure of Al and Ga was calculated for analysis the alloy composition. We found that gallium decomposition has much effect on aluminum incorporation. The composition gradient at interface was discussed with consideration of strain barrier induced composition pulling effect and parasitic reaction between Al source material and ammonia. 2 DEG transport properties were characterized for the samples growth at 300 Torr.

II. Experiment

The 30~100 nm AlGaN layers were grown at 1060~1100°C on 2 mm-thick undoped GaN layers (with room temperature mobility of ~500 cm^2/Vs and background carrier of $\sim 5\times 10^{16}$ cm^{-3}) on c-plane sapphire substrates by EMCORE D-180 rotating disc MOCVD system. NH_3 flow rate was varied from 15 to 40 L/min, reactor pressure from 100 to 300 Torr, and Al/(Ga+Al) input ratio from 0.15 to 0.40, respectively. Al composition was measured by high-resolution x-ray diffraction and secondary ion mass spectrometry (SIMS). SIMS and Auger electron spectroscopy (AES) depth profiling were used to determine the compositional profile at the AlGaN/GaN interfaces. The mobility and the sheet carrier concentration were measured by Van der Pauw method at liquid nitrogen temperature.

III. Results and discussion

3.1. Al solid incorporation at different NH_3 flow rate and different reactor pressure

To investigate effect of NH_3 flow rate on Al incorporation behavior, samples were grown at different NH_3 flow rate ranging from 15 L/min to 40 L/min at reactor pressure 300 Torr and with the same gas phase input ratio of $Al/Ga + Al = 0.26$. Fig. 1 shows the Al composition as a function of NH_3 flow rate, i.e. NH_3 input mole ratio. The composition was measured by HRXRD (inset at upper right corner). It was found that the Al was incorporated more efficiently into AlGaN alloy as NH_3 flow rates decreased from 25 to 15 L/min and above 25 L/min the Al composition was independent of NH_3 flow rate and it seem to became saturated. We try to explain the NH_3 flow rate dependent of Al composition in AlGaN alloy with equilibrium thermodynamic model [16] by calculation of gallium and aluminum equilibrium partial pressure P_{Al} and P_{Ga} assuming that the growth was at masstranport region and neglecting the pre-reaction between metal organic source and NH_3. The results show that equilibrium vapor pressure of Al over the growth surface is significantly lower than that of Ga. This is due to the equilibrium constants of reaction for binary AlN and GaN are in the order $K_{AlN} \ll K_{GaN}$. The calculated Al and Ga partial pressure are in the range of $\sim 10^{-17}$ atm and $\sim 10^{-6\sim-7}$ atm respectively as shown in Fig. 2. Thus the solid composition $x = (P_{Al}^{o}-P_{Al})/[(P_{Al}^{o}-P_{Al}) + (P_{Ga}^{o}-P_{Ga})]$ can be expressed as $x \sim P_{Al}^{o}/[P_{Al}^{o} + (P_{Ga}^{o}-P_{Ga})]$. So the Al composition mainly controlled by gallium equilibrium partial pressure P_{Ga}. Fig. 3 is the calculated equilibrium Ga partial pressure as function of input NH_3 mole fraction (of total flow). It shows that the P_{Ga} much sensitive to NH_3 input mole fraction at lower input NH_3 flow

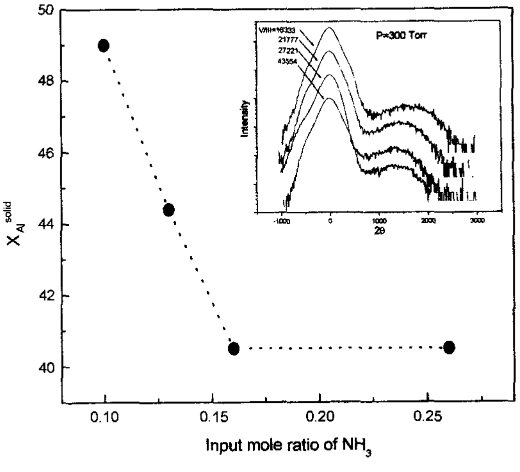

Fig. 1. Solid composition of $Al_xGa_{1-x}N$ as a fuction of input NH_3 mole ratio (flow rate) growth at 300 Torr. Inset shows the peak split of (002) between GaN and AlGaN measured by HRXRD.

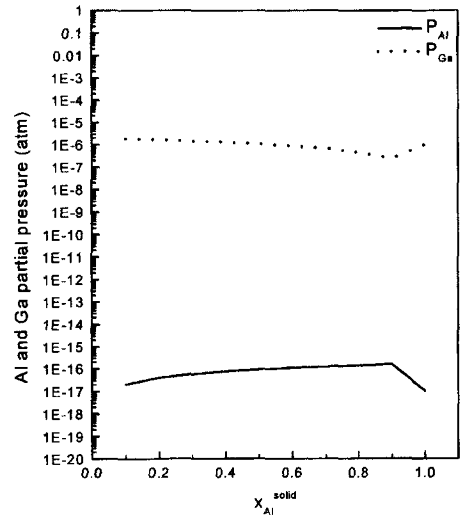

Fig. 2. The calculated equilibrium partial pressures as a function of the solid composition in the $Al_xGa_{1-x}N$ alloy. Calculation parameters: $T = 1050°C$, $P_{tot} = 1$ atm, $V/III = 10000$, input group III partial pressure $P_{III}^{o} = 1 \times 10^{-5}$ atm.

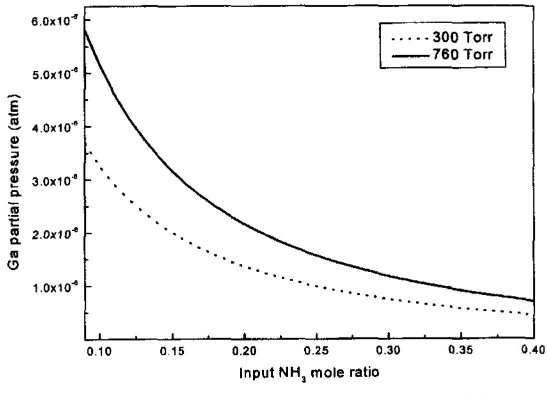

Fig. 3. Calculated equilibrium partial pressure of Ga as a function of input NH_3 mole ratio. $T = 1100°C$, $P_{tot} = 300$, 760 Torr, initial input Ga partial pressure $P_{Ga}^{o} = 5 \times 10^{-6}$ (atm).

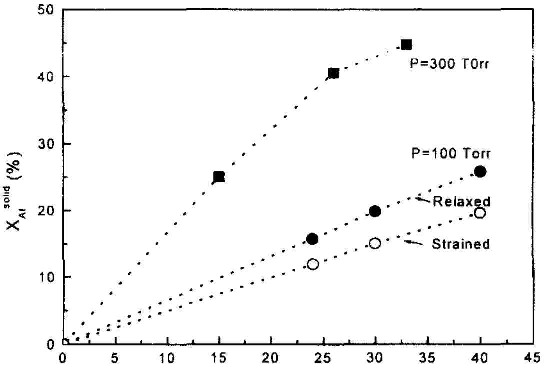

Fig. 4. Solid-vapor composition relation for samples growth at different reactor pressure 100 Torr and 300 Torr.

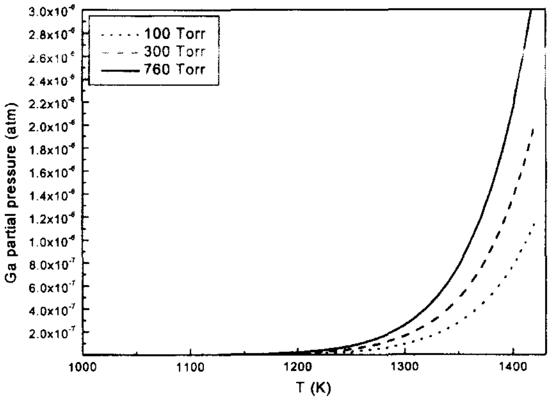

Fig. 5. Calculated equilibrium partial pressure of Ga as a function of temperature at various reactor pressures of 100 Torr, 300 Torr, 760 Torr. V/III = 10000.

rate. The P_{Ga} soaring up when the input fraction of NH_3 below 0.1 (correspond to V/III = 10000 in this calculation) and it change slowly at higher input NH_3 fraction. It qualitatively explained the Al incorporation behavior with respect to NH_3 flow rate. At high growth temperature (1100°C) of AlGaN alloy we have to consider the decomposition of Ga from the alloy because the gallium equilibrium partial pressure much increased and it comparable to input gallium partial pressure of 10^{-6} (atm) which is normally used growth conditions.

To study the effect of growth pressure on the growth of AlGaN alloy, we growth two series of AlGaN layers at growth pressure of 100 Torr and 300 Torr. Fig. 4 shows the solid vapor relation of compositions for AlGaN alloy at growth pressure P_{tot} = 100 Torr and 300 Torr respectively. It clearly shows preferred incorporation of Al in AlGaN at higher growth pressure than at low pressure. The composition tend to saturate at gas phase input ratio above 35% for sample growth at 300 Torr, on the other hand it show linearity for the samples growth at 100 Torr. This results is different from others reported growth pressure dependent of AlGaN composition where they reported that Al incorporated much efficiently due to reduced gas phase reaction between TMA and NH_3 at lower growth pressure [10]. Our results are explained by analyzing the equilibrium gallium partial pressure. Fig. 5 shows calculated temperature dependent of gallium equilibrium partial pressure at different growth pressure for our growth condition. At high temperature of 1100°C, the gallium equilibrium partial pressure becomes of comparable to input gallium equilibrium partial pressure (~10^{-6} atm). Thus we have to take in to account gallium decomposition at this growth range. Especially, we find that the gallium equilibrium partial pressure increase much higher at higher total growth pressure. Thus, the decomposition effect of gallium became large at higher growth pressure compare to that at lower growth pressure. This mean more gallium species remain in gas phase rather than incorporated into the alloy at higher growth pressure. This results in relatively higher Al solid composition in AlGaN alloy for samples growth at higher reactor pressure than at lower reactor pressure. With above discussions, we conclude that during the growth of AlGaN alloy at the high temperature, the decomposition of gallium species must take into account. Gallium equilibrium partial pressure tend to increase significantly at high temperature and become of comparable to initial input gallium partial pressure especially at lower NH_3 flow rate and higher total growth pressure. This helps gallium decomposition and results in preferred incorporation of Al in AlGaN alloy accordingly.

3.2. Alloy composition gradient over AlGaN/GaN heterointerface

We find that growth conditions of NH_3 flow rate and growth pressure not only affect the alloy incorporation behavior but also have much effect on composition gradient at interface. Fig. 6 shows the SIMS depth profiles over interface region for the

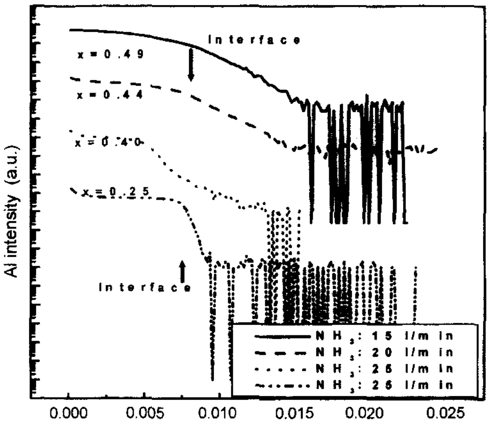

Fig. 6. SIMS depth profile over AlGaN/GaN heterointerface grown at 300 Torr and T = 1100°C. NH_3 flow rates were varied from 15 to 25 L/min.

samples growth at 300 Torr with different NH_3 flow rate. Compositional interface abruptness became worse with decreasing NH_3 flow rate and accordingly with increasing Al alloy composition. We believe that strain barrier formed at interface due to lattice mismatch between AlGaN and GaN can be responsible to this composition pulling effect at the interface. And this strain barrier increased with increasing Al composition and it consist with large compositional pulling at high Al concentration show in this figure. Also we find sample growth at lower pressure has better interface quality by means of compositional abruptness compare to samples growth at higher pressure as in Fig. 7. It reported there are some extent of gas phase pre- reaction between NH_3 and TMA, specially at high growth pressure [10]. Thus the larger composition gradient at higher growth pressure can be explained by depletion of Al species at the growing surface caused by prereactions between source material and NH_3.

3.3. 2 DEGs transport properties

The electrical transport properties of the AlGaN/GaN heterointerface were characterized by measuring the temperature dependent of mobility and the sheet carrier concentration. Fig. 8 shows the temperature dependent sheet carrier density and mobility AlGaN/GaN with compositions of x = 25%, 40% and 45% grown at 300 Torr. It is clearly show that two dimensional electron gas is well confined at AlGaN/GaN heterointerface judging from the typical temperature dependent behavior 2 DEGS sheet carrier density and mobility in x = 25% sample. The sheet carrier density and mobility are 2×10^{13} cm^{-2} and 1245 cm^2/Vs at room temperature and 1.38×10^{-3} cm^{-2} and 5000 cm^2/Vs at 100 K respectively. With further increasing Al composition (x>40%), both sheet carrier density and mobility were drastically reduced. When the Al composition above 40%, biaxial strain maybe partially relaxed which causes the reduction of sheet carrier induced by piezoelectric field. The reduction of sheet carrier also can be coming from free electron trap due to the misfit dislocation at high

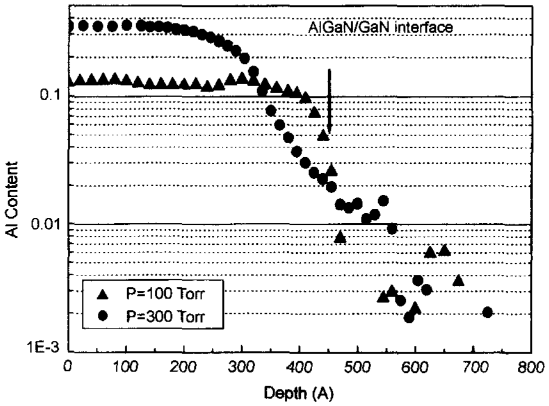

Fig. 7. AES depth profile (interface region of AlGaN/GaN heterostructure) for samples growth at 100 Torr and 300 Torr. NH_3 flow rate and gas-phase input ratio is 25 L/min and 0.26, respectively.

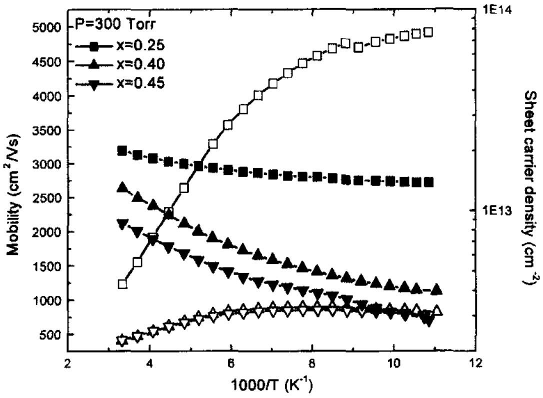

Fig. 8. Electron mobility and sheet carrier concentration of AlGaN/GaN heterostructure, x = 0.32~0.50 grown at 300 Torr. T = 1100°C.

Al composition. We noticed from Fig. 6, that the interface became worse with increase Al composition. These composition gradient causes the broadening of interface region and result in reduction of 2 DEG confinement and hence reduce the sheet carrier density. Especially the broadening of interface region due to compositional gradient can strongly scat the 2 DEGs confined near interface and cause profound reduction of mobility. It is concluded that to obtain high sheet carrier concentration as well as high mobility in the AlGaN/GaN heterostructure, the alloy composition and interface must be carefully controlled during growth with optimizing important growth parameters of NH_3 flow and reactor pressure.

IV. Conclusion

Al incorporation behavior was investigated at various growth pressures and NH_3 flow rates. It was found that Al was incorporated more efficiently in the AlGaN films at higher growth pressure and at lower NH_3 flow rate. We believe that more decomposition of Ga occur at growth conditions of lower NH_3 flow rate and higher growth pressure leading to preferred Al incorporation into AlGaN at relatively higher temperature. Severe composition gradient in AlGaN/GaN heterointerface was observed at higher reactor pressure and higher Al composition and lower NH_3 flow rate. This composition gradient over the heterostructure interface lowered sheet carrier concentration and electron mobility as well. We obtained an AlGaN/GaN heterostructure with sheet carrier density of $\sim 2\times 10^{13}\,\mathrm{cm}^{-2}$ and mobility of 1250 and 5000 $\mathrm{cm}^2/\mathrm{Vs}$ at 300 K and 100 K, respectively.

Acknowledgment

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Reference

[1] S. Nakamura, T.Mukai, and M. Senoh, Appl. Phys. Lett. **64**, 1687 (1994).

[2] S. Nakamura, M. Senoh, S. Nakahama, N. Iwasa, T. Yamada, T. Matsushita, S. Sukimoto, and H. Kiyoku, Appl. Phys. Lett. **70**, 616 (1997).

[3] M. A. Khan, J. N. Kusnia, D. T. Olson, G. M. Van Hove, M. Blasingane, and L. F. Reitz, Appl. Phys. Lett. **60**, 2917 (1992).

[4] D. walker, X. Zhang, P. Kung, A. Saxler, S, Javadpour, J. Xu, and M. Razeghi, Appl. Phys. Lett. **68**, 2100 (1996).

[5] O. Ambacher, J. Phys. D **31**, 2653 (1998).

[6] M. A. Khan, Q. Chen, M. S. Shur, B. T. McDermott, J. A. Higgins, J. Burm, W. J. Schaff, and L. F. Eastman, IEEE Electron Device Lett. **17**, 584 (1996).

[7] R. Gaska, Q. Chen, J. Yang, A. Osinsky, M. A. Khan, and M. S. Shur, IEEE Electron Device Lett. **18**, 492 (1997).

[8] Y. F. Wu, S. Keller, P. Kozodoy, B. P. Keller, P. Parish, D. Kapolnek, S. P. DenBaars, and U. K. Mishra, IEEE Electron Device Lett. **18**, 290 (1997).

[9] R. Dimitrov, L. Wittmer, H. P. Felsl, A. Mitchell, O. Ambacher, and M. Stutzmann, Phys. Status Solidi A **168**, R7 (1998)

[10] A. E. Wickenden, D. D. Koleske, R. L. Henry, R. J. Gorman, M. E. Twigg, M. Fatemi, J. A. Freitas, Jr., and W. J. Moore, J. Electron. Mater. **29**, 21 (2000).

[11] C. H. Chen, H. Liu, D. Steigerwald, W. Imler, C. P. Kuo, and M. G. Craford, Mat. Res. Soc. Symp. Proc. **395**, p. 103 (1996).

[12] J. Han, J. J. Figiel, M. H. Crawford, M. Banas, M. E. Bartram, R. M. Biefeld, Y. K. Song, and A.V. Nurmikko, J, Cryst. Growth **195**, 291 (1998).

[13] T. G. Mihopoulos V. Gupta, and K. F. Jensen, J. Cryst. Growth, **195**, 733 (1998).

[14] F. Nakamura, S. Hashimoto, M. Hara, S. Imanaga, M. Ikeda, and H. Kawai, J. Cryst. Growth **195**, 280 (1998).

[15] S. C. Choi, J. H. Kim, J. Y. Choi, K. J. Lee, K. Y. Lim, and G. M. Yang, J. Appl. Phys. **87**, 172 (2000).

[16] A. Koukitu, N. Takahashi, T. Taki and H. Seki, Jpn. J. Appl. Phys. **35**, L673 (1996).