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IMPACT OF GROWTH TEMPERATURE, PRESSURE AND STRAIN ON THE MORPHOLOGY OF GaN FILMS

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ABSTRACT

GaN films grown on sapphire at different temperatures are investigated. A Volmer-Weber growth mode is observed at temperatures below 1000K that leads to thin films composed of oriented grains with finite size. Their size is temperature dependent and can actively be influenced by strain. Largest grains are observed in compressed films. It is argued that diffusing Ga ad-atoms dominate the observed effects with an activation energy of 2.3 ± 0.5 eV. Comparably large grain sizes are observed in films grown on off-axes sapphire substrates and on bulk GaN. This assures that the observed size limitation is a consequence of the 3D growth mode and not dependent on the choice of the substrate. In addition, the grain size and the surface roughness of the films depend on the nitrogen partial pressure in the molecular beam epitaxy (MBE) chamber,most likely due to collisions between the reactive species and the background gas molecules. This effect is utilized to grow improved nucleation layers on sapphire.

INTRODUCTION

GaN films are usually grown at temperatures that are low compared with the melting point ($< 0.5\ T_m$) of the material. This, as well as the growth on lattice mismatched substrates (e.g. sapphire or SiC) with largely different thermal expansion coefficients, greatly affects the crystal quality and introduces strain into the layers. Amano et al. [1] and Nakamura [2] introduced the growth of AlN and GaN buffer layers to improve the film quality and to relax strain. A coexistence of hydrostatic and biaxial strain components in the GaN films was recently found [3] that allows to strain engineer the films. The design of an appropriate buffer layer structure is crucial for this purpose. However, the impact of growth parameters on the film morphology is not well understood. Wu et al. reported on the evolution of the morphology of GaN buffer and main layers [4]. However, its dependence on growth temperature, pressure and strain remained hidden. In this paper we report on the impact of growth temperature, strain, substrates and pressure on the morphology of GaN films.

EXPERIMENTAL

The GaN films were grown by ion-assisted MBE on c-plane and off-axes sapphire substrates. Homoepitaxial growth was performed on bulk GaN. Details of the growth procedure are reported elsewhere [5]. The films were homogeneous and well reproducible. Here, ~ 1.5μm thick GaN layers were investigated. An n-type carrier concentration of ~ 10^{19} cm^{-3} was present in

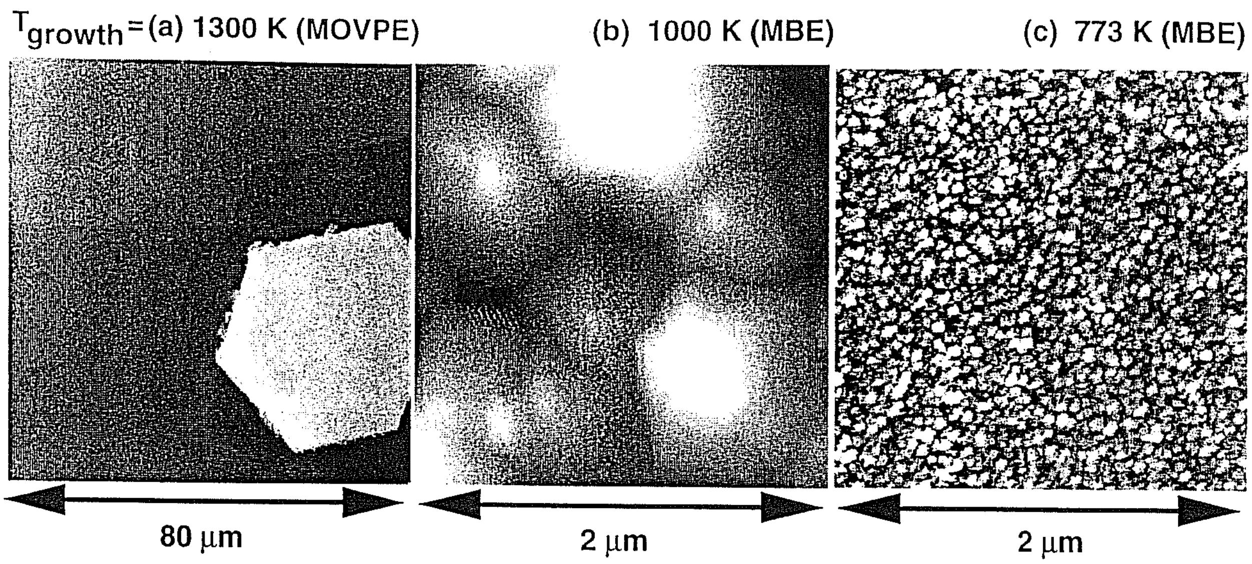


Figure 1: AFM plan-view images of GaN films on sapphire substrates grown at different temperatures by different methods as indicated.

all investigated MBE-grown films that did not change with the growth conditions. The n-type conductivity might be influenced by impurities. Oxygen, carbon and hydrogen were detected by SIMS at concentrations larger than 10^{18} cm^{-3}.

Here, we report on results obtained by AFM, by PL spectroscopy and by TEM. AFM measurements (contact mode) were done in air with an AFM tip force of 1 nN and a scan rate of 1 Hz. We used the root mean square (RMS) value of a 2 µm x 2 µm area to quantify the surface roughness. The feature sizes were evaluated statistically.

PL measurements were done at 4 K using the excitation of a 325 nm He-Cd laser line. The shift of the dominant donor bound exciton from its zero strain position at 3.467 eV [3] was evaluated to measure strain at 4 K. A narrow half width of the lines (best full width at half maximum: 1.8_5 meV at 3.468_8 for GaN film grown by MBE on sapphire) allowed to measure strains as small as 10^{-4}.

RESULTS AND DISCUSSION

Figure 1 shows AFM images of GaN films grown on sapphire at different growth temperatures. We compare the surface morphology of the MBE grown films (figure 1b,c)

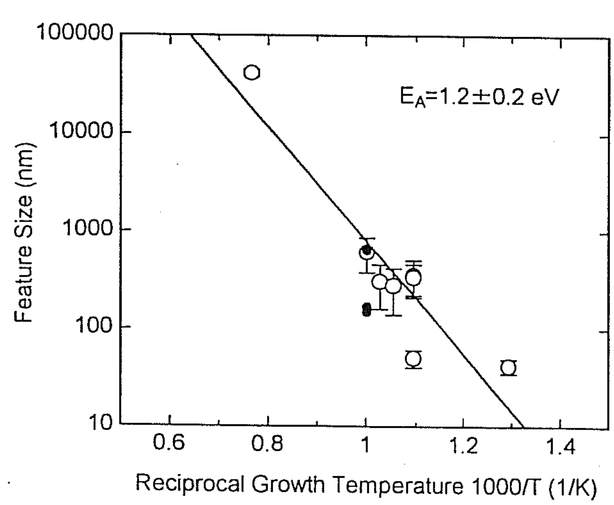


Figure 2:
Feature size as a function of the reciprocal growth temperature. Open circles: Films grown by MOCVD and by MBE. Solid circles: strain engineered films grown by MBE.

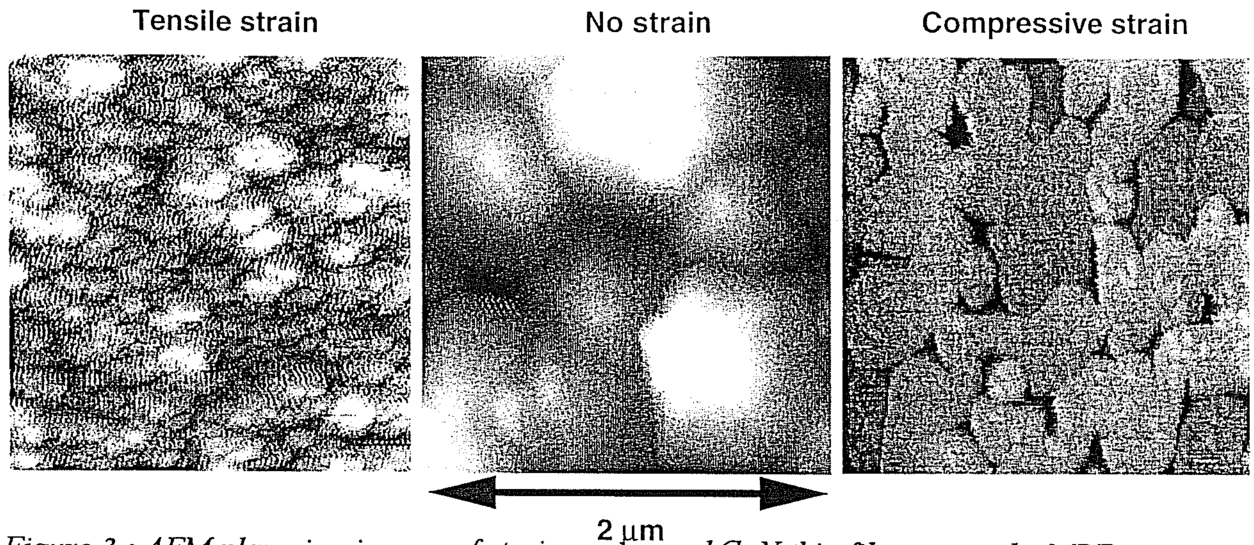
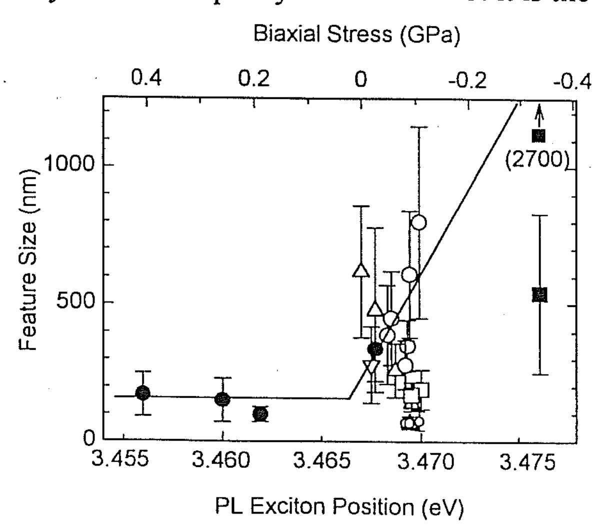


Figure 3 : AFM plan-view images of strain engineered GaN thin films grown by MBE at 1000K.

with the surface of a GaN film grown by MOVPE grown at 1300K (figure 1a). Details of the film morphology are given elsewhere [6]. Here, we stress that in figure 1a a Stranski-Krastanov [7] growth mode can be seen that causes two-dimensional (2D) plateaus and a three-dimensional (3D) grain. Step flow growth is observed in the flat areas. In contrast, figures 1b and 1c depict a Volmer-Weber [8] growth mode with 3D features only. Cross-section TEM reveals that the features seen in figures 1b and 1c are in fact oriented grains that form the GaN film [5]. The results shown in figure 1 are typical for all other investigated samples and demonstrate that the sizes of grains in the films depend on the growth temperature. Figure 2 depicts that the logarithm of the grain diameter is approximately a linear function of reciprocal growth temperature. Open circles show GaN films grown by MOVPE and by MBE. Solid circles are examples from a series of GaN films grown by MBE with different strains in the films. The grain size appears to be thermally activated. An activation energy $E_A = 1.1_5 \pm 0.2_5$ eV can be estimated from this Arrhenius plot. If we assume that the grain size is limited by a surface *diffusion length* $x \sim (Dt)^{1/2}$, where D is diffusion coefficient and t is the time, the activation energy of the diffusion coefficient is 2.3 eV. Brandt et al. reported on an activation energy of 2.4 eV for the *diffusion rate* of Ga ad-atoms on cubic GaN surfaces [9]. However, they could not specify whether or not it is the

Figure 4:
Dependence of feature size on the donor bound exciton position that measures strain. MBE growth. Open circles: variation of nitrogen flux. Open triangle (upwards): variation of buffer layer thickness. Solid square: Growth on MOCVD film. Open squares: pressure controlled buffer layer structure. Open triangle (downwards):Different main layer growth temperature.



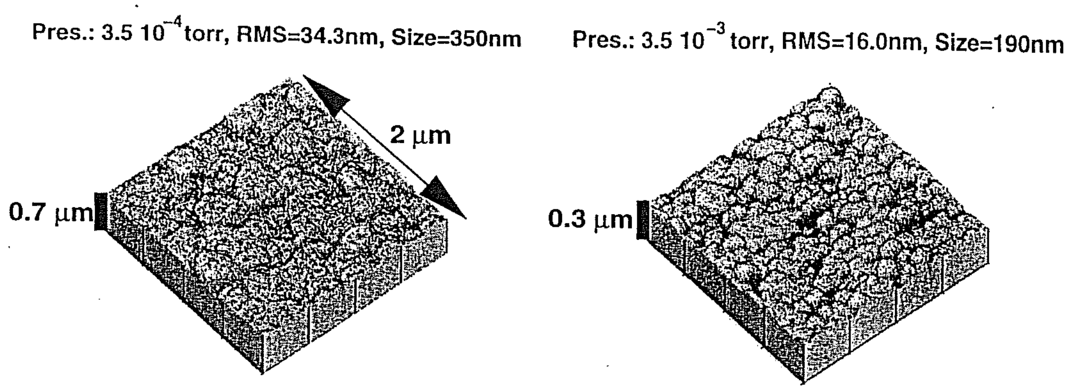


Figure 5: AFM 3D images of GaN films grown on sapphire substrates with a different buffer layer structure controlled by the nitrogen partial pressure in the MBE chamber.

diffusion length itself or the diffusion rate that depends on temperature. The similarity of the data suggests that it is the *diffusion length* of Ga ad-atoms which is temperature dependent and limits the grain sizes in our case. It is also seen in figure 2 that strain modifies the feature sizes as shown by the solid circles.

Next, we describe a correlation of the feature size with strain. The strain in the films was engineered by compositional changes and by the growth of buffer layers of different thickness [10]. Figures 3 shows that for a given growth temperature the feature sizes depend on strain that was extracted from the energetic position of donor bound excitons [3]. We investigated this correlation for a variety of films. From the caption of figure 4 it is clear that very differently grown samples were investigated. Yet, an analysis in terms of strain gives consistent results: the feature size in GaN films under compression can be larger than in films under tension. Thus, tensile and compressive strains affect the grain sizes in an opposite manner. Also, only films that are little strained or strain free exhibit a pyramidal surface structure while strained films are atomically flat [5]. We argue that it is the activation energy for the diffusion of Ga ad-atoms that is likely to be strain dependent. In fact, a tensile strain *widens* the in-plane lattice spacing of GaN and this may result in an increased trapping of the "large" Ga ad-atoms. However, compressed films may also exhibit smaller surface features at the same time. Ongoing investigations indicate that such small features can be related to the presence of extended defects that locally modify the strain. It should be noted that the differential thermal expansion of a GaN layer and its sapphire substrate will add a compressive component to the strain present at the growth temperature upon post-growth cooling. This was recently measured to occur up to 600K [11]. Thus, the strain measured at 4K is not equal to the strain state of the sample during growth.

Figure 5 shows AFM 3D images of GaN films grown on sapphire substrates with different buffer layer structures controlled by the nitrogen partial pressure during MBE growth. The nitrogen partial pressures during buffer layer growth was 3.5 x 10^{-4} Torr (left) and 3.5 x 10^{-3} Torr (right), respectively. The pressure during main layer growth is the same for both samples (3.5 x 10^{-3} Torr). It is seen that the film grown on the buffer layer with the larger nitrogen partial pressure is considerably smoother though it also exhibits smaller surface features. a nitrogen partial pressure in the range 10^{-3} - 10^{-4} Torr reduces the mean free path of the activated ions to a value comparable with the source-substrate distance. Thus, collisions with gas molecules reduce the kinetic energy of the adsorbed reactive species. Consequently, surface diffusion is reduced

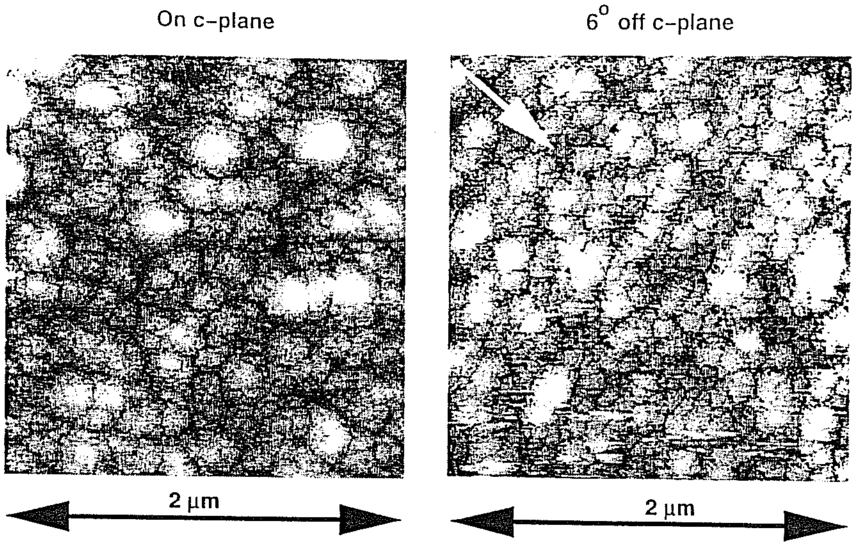


Figure 6:

AFM images of MBE grown GaN films on c-plane sapphire and on substrates 6° inclined towards the a-plane. The misorientation direction is indicated.

resulting in the formation of a uniform nucleation layer with grains reduced in size. This is favorable for dislocation annihilation during main layer growth. We exploit this effect to improve the quality of our MBE grown films [5].

Finally we evaluated a possible impact of the substrate material on the size of the grains in our films. Figure 6 shows AFM plan-view images of films grown by MBE at 1000K on c-plane sapphire and on 6° off c-plane sapphire towards [11-20](a-plane). The misorientation direction is indicated in figure 6. Similar small features are observed on all films grown on the other misoriented substrates. In the particular example of figure 6 the misorientation did not stimulate step flow growth but rather an alignment of the grains along terraces. The surface RMS roughness of the films increases with increasing off-angles from c-plane (figure 7). Using bulk GaN as a substrate material led to a morphology of the GaN thin film that is very simimilar to the one shown in figure 6. The results support our interpretation that the formation of grains in the GaN films grown by MBE at temperature below 1000K is caused by limited surface diffusion processes and not by the choice of substrates.

CONCLUSIONS

In conclusion, a three dimensional (3D) Volmer-Weber growth mode determines the morphology of GaN thin films grown by ion assisted MBE below 1000K. A transition to a step flow growth mode (2D/3D) occurs above this temperature. For the first time we show that the size of the grains in the films is temperature dependent and modulated by strain which induces an asymmetry with respect to

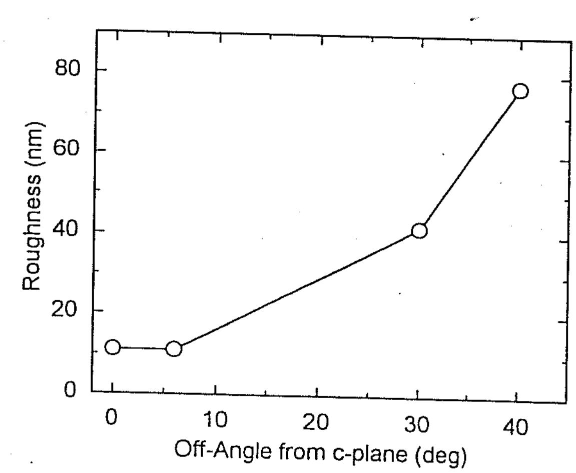


Figure 7:
Surface roughness as a function of the misorientation angle between the sapphire c- and a-plane.

compression or tension. If the formation of grains in the films is attributed to a limited surface diffusion length, we estimate a strain dependent diffusion coefficient of 2.3 eV. It is suggested to attribute this energy to the surface diffusion of Ga ad-atoms. The use of different substrates does not influence this growth mode significantly. An increase of the nitrogen partial pressure for buffer layer growth is found to be suitable for a better coverage of sapphire with small GaN nucleation sites.

ACKNOWLEDGMENTS

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REFERENCES

1. H. Amano, N. Sawaki, I. Akasaki and Y. Toyoda, Appl. Phys. Lett. **48**, 353 (1986).
2. S. Nakamura, Jpn. J. Appl. Phys. **30**, L1705 (1991).
3. C. Kisielowski, J. Krueger, S. Ruvimov, T. Suski, J.W. Ager III, E. Jones, Z. Liliental-Weber, M. Rubin, E.R. Weber, M.D. Bremser, R.F. Davis, Phys. Rev. B **54**, Dec, 1996, in press.
4. X. H. Wu, D. Kapolnek, E. J. Tarsa, B. Heying, S. Keller, B. P. Keller, U. K. Mishra, S. P. DenBaars and J. S. Speck, Appl. Phys. Lett. **68**, 1371 (1996).
5. M. S. H. Leung, R. Klockenbrink, H. Fujii, J. Krueger, C. Kisielowski, Sudhir G. S., A. Anders, Z. Liliental-Weber, E. R. Weber and M. Rubin, this volume.
6. H. Fujii, C. Kisielowski, J. Kruger, M. S. H. Leung, R. Klockenbrink, Sudhir G. S., E. R. Weber and M. Rubin, submitted to J. Appl. Phys. (1996).
7. I. N. Stranski and Von Krastanov, Akad. Wiss. Mainz L. Math-Nat. Kl IIb **146** 797 (1939).
8. M. Volmer and A. Weber, Z. Physik. Chem. **119** 277 (1926).
9. O. Brandt, H. Yang, A. Trampert and K. H. Ploog, Materials Research Society Symposium Proceedings, **395**, 27 (1996).
10. C. Kisielowski, J. Krueger, M. S.H. Leung, R. Klockenbrink, H. Fujii, T. Suski, Sudhir G.S., J.W. Ager III., M. Rubin and E.R. Weber, Proceedings of International Conference on Physics of Semiconductors , Berlin 1996, World Scientific, Singapore 1996, p.513.
11. T. Suski, J. W. Ager III, G. Conti, Z. Liliental-Weber, J. Kruger, S. Ruvimov, C. Kisielowski, E. R. Weber, C. P. Kuo, I. Grzegory and S. Porowski, Proceedings of International Conference on Physics of Semiconductors , Berlin 1996, World Scientific, Singapore 1996, p.2917.