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D. Huang
*Virginia Commonwealth University*

P. Visconti
*Virginia Commonwealth University*

K. M. Jones
*Virginia Commonwealth University*

***See next page for additional authors***

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**Authors**
D. Huang, P. Visconti, K. M. Jones, Michael A. Reshchikov, F. Yun, A. A. Baski, T. King, and Hadis Morkoç

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# Dependence of GaN polarity on the parameters of the buffer layer grown by molecular beam epitaxy

D. Huang,[a] P. Visconti,[b] K. M. Jones, M. A. Reshchikov, F. Yun,
A. A. Baski, T. King, and H. Morkoç[c]
*Virginia Commonwealth University, Department of Electrical Engineering and Physics Department, Richmond, Virginia 23284*

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The polarity of GaN films grown using GaN and AlN buffer layers on sapphire substrates by molecular beam epitaxy were investigated by atomic force microscopy, hot wet chemical etching, and reflection high-energy electron diffraction. We found that the GaN films grown on high temperature AlN (>890 °C) and GaN (770–900 °C) buffer layers invariably show Ga and N polarity, respectively. However, the films grown using low temperature (~500 °C) buffer layers, either GaN or AlN, could have either Ga or N polarity, depending on the growth rate of the buffer layer. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1380399]

Wurtzitic GaN is predominantly grown on the *c* plane of sapphire by hydride vapor phase epitaxy, molecular beam epitaxy (MBE), and metalorganic chemical vapor epitaxy (MOCVD). Among the other incompatibilities, GaN does not share the same atomic stacking order with sapphire.[1,2] Consequently, the crystal direction [0001] of GaN film, the direction of the long bond along the *c* axis from Ga to N atoms, can be either parallel or antiparallel to the growth direction. The epilayer in the former case is conventionally referred to have Ga polarity or Ga face, while the latter have N polarity or N face.[3,4] Investigations have shown that these two polar films have vastly differing growth and surface properties. For examples, a Ga face is typically more smooth than a N face.[5–7] For MBE growth near stoichiometric conditions, the growth rate of N-polar domains may be slightly lower than that of Ga-polar matrix, leading to the formation of pits with inversion domains at their centers.[8] A *p*-type doping by Mg is easier in Ga-polar films, while C, O, Si, and other residual impurities are more likely to incorporate into N-polar films.[9,10] A Ga face is also more stable than a N face against wet chemical etching[5,6] and exposure to the nitrogen plasma at MBE growth temperatures.[11] In addition to the growth and the surface properties, the polarity also has significant effects on the electrical and optical properties of the films. The photoluminescence spectra,[12,13] the Pt/GaN Schottky barrier,[14] the band discontinuities,[15] and the two-dimensional charges in GaN/AlGaN heterostructures[16] are all affected by the polarity of the structures.

Different polarities can be identified by various techniques[6,7,17–23] including chemical etching and surface examination by atomic force microscopy (AFM).[24–27] For device applications, an understanding and control of the crystal polarity in the epitaxial growth is essential. For this purpose, the effects of the substrate nitridation,[28] buffer layer materials and growth conditions[29–32] such as III/V ratio[8,21] have been investigated. The change in film polarity by Mg doping was also observed.[33–35] It has been demonstrated that, in the case of MOCVD on sapphire substrates, either GaN or AlN low temperature buffer layers lead to Ga-polar films.[3,6,7,17,24,25,32] For MBE growth, however, the published results show that AlN buffer layers commonly lead to Ga-polar films while GaN buffer layers lead to N-polar films.[8,9,14,19,25,29,30–32] Although there are suggestions that low temperature GaN buffer layers may increase Ga domains[18] or lead to Ga-polar films in some cases,[28,36] a correlation between the film polarity and buffer layer growth is still not yet well established. In this letter, we report the dependency of GaN polarity on the parameters of the buffer layer growth by MBE. We show that both AlN and GaN buffer layers can lead to Ga- and N-polar films depending on the growth temperature and growth rate of the buffer layers.

Unintentionally doped GaN layers were grown by MBE on the *c* plane of sapphire substrates using radio frequency activated N. Nitridation was performed at both high (890–985 °C) and low (about 500 °C) temperatures by radio frequency N plasma, which have no apparent effect on the results. Four groups of samples were grown and investigated. The first and second sets utilized GaN buffer layers grown near 500 °C, and near 800 °C. The third and fourth groups utilized AlN buffer layers grown near 500 °C, and 890–930 °C. Following the buffer layers, typically 1-$\mu$m-thick GaN layers (active layers) were grown at a substrate temperature between 720 and 850 °C with growth rates in the range of 300–1000 nm/h under N-limited (Ga-rich) conditions.

The polarity of the films was initially determined by RHEED followed by wet chemical etching and AFM investigations of both as-grown and etched surface morphologies. As reported in the literature, the surface of an as-grown Ga-polar film is either very flat or shows stepped terraces, often with pits.[5,6,8,9,11,20,23] The surface of an as-grown N-polar film by MBE, however, often shows tall columns or terraces separated by deep troughs, without pits on the surface.[9,11,20,23] Similar to NaOH and KOH based etching,[5,6,9,20] we have found that hot (160 °C) $H_3PO_4$ etches

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[a] Also at: Physics Department, Fudan University, Shanghai 200433, China.

[b] Also at: Instituto per lo Studio di Nuovi Materiali per l'Elettronica, CNR, Via Arnesano, Lecce, Italy and INFM-Unita di Lecce, Dipartimento di Ingegneria dell'Innovazione, University di Lecce, Lecce, Italy.

[c] Electronic mail: hmorkoc@vcu.edu.

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N-polar GaN films very quickly resulting in either the complete removal or a drastic change in the surface morphology of the layer as revealed by AFM or even by optical microscopy.[27] The etching rate for N-polar GaN films is in the range of 0.1–0.5 $\mu$m/min. On the contrary, the acid attacks only the defect sites in Ga-polar films leaving the defect-free GaN intact and the morphology unchanged.[27] The polarity assignments were also consistent with *in situ* RHEED patterns.[3,20,21,22] A Ga face usually shows $2\times2$ RHEED pattern upon cooldown at temperatures between 280 and 650 °C after the entire structure was grown. The RHEED pattern of a N face upon cooling shows, however, only the bulk $1\times1$ structure, though $3\times3$ has been reported.

We first examine the polarity of the samples grown on GaN and AlN buffer layers at high temperatures. GaN films grown on high temperature (>770 °C) GaN buffer layers invariably turned out to be N polarity regardless whether a static or graded (with positive or negative slope) substrate temperature was employed during the buffer growth. The surface morphology investigated by AFM and the wet etching experiments confirmed the polarity assignment. The RHEED pattern upon cooling indicates only the bulk $1\times1$ structure. Conversely, GaN films grown on high temperature (~900 °C) AlN buffer layers with thicknesses in the range of 8–35 nm and growth rates of 40–60 nm/h led to Ga polarity. Also in this case, the polarity assignment was confirmed by AFM study of the surface morphology and the wet etching. The RHEED patterns of $2\times2$ reconstruction were observable.

The GaN films grown on low temperature (~500 °C) buffer layers (either GaN or AlN) were found to have either Ga or N polarity. A 60–150-nm-thick GaN buffer layer at a growth rate of about 600 nm/h can lead to Ga polarity as confirmed by the surface morphology and the characteristic $2\times2$ RHEED pattern. However, when the thickness of the GaN buffer layer was reduced to 30–40 nm, keeping the same growth rate, the layers turned out to be of mixed polarity with a faint $2\times2$ reconstruction observed upon cooldown. When about 110–220 nm thick GaN buffer layers were grown at 500 °C with a lower growth rate (220 nm/h), the resultant layers were of N polarity with consistent surface morphology and $1\times1$ RHEED pattern. For the GaN films grown on low temperature (~500 °C) AlN buffer layer, their polarities also depend on the growth rate of the buffer layer. When 20-nm-thick AlN buffer layers grown at a rate of 60 nm/h were employed, Ga-polarity films resulted. However, when ~20-nm-thick AlN buffer layers were grown at a lower rate of 20–30 nm/h, N-polarity films can be produced.

The typical surface morphologies of as-grown Ga-polar films with different buffer layers are presented in Fig. 1. In this case, a high temperature AlN buffer layer tends to result in a smooth, but pitted layer [Fig. 1(a)], consistent with the group III/V ratio employed. Higher group III/V ratios generally lead to disappearance of the pits. A low temperature AlN buffer layer grown at a high rate leads to a Ga-polar surface morphology with irregular stepped terraces, often with pits and/or a rough surface [Fig. 1(b)]. When a low temperature GaN buffer layer grown at high rate was used, we found a similar morphology to that shown in Fig. 1(b) with a more drastic variation in terrace height and shape [Fig. 1(c)].

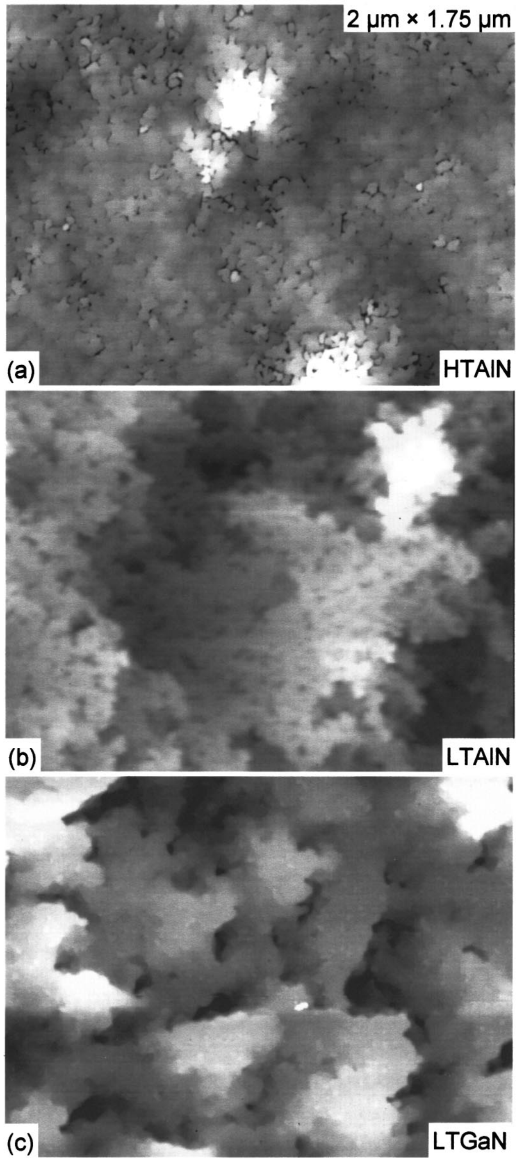

FIG. 1. AFM images of Ga-polar samples grown on three different buffer layers with different growth conditions: high temperature AlN buffer layer (a), low temperature AlN buffer layer at high growth rate (b), and low temperature GaN buffer layer at high growth rate (c). The vertical scales are 10, 20, and 20 nm for images (a), (b), and (c), respectively.

The surface morphologies of as-grown N-polar films with different buffer layers are presented in Fig. 2. With a high temperature GaN buffer layer, the film morphology is that of noncoalesced columns [Fig. 2(a)]. In general, smoother morphologies with stepped terraces were found when a low temperature AlN buffer layer grown at a low rate was used [Fig. 2(b)]. Using low temperature GaN buffer layers grown at a low rate, the morphologies vary from extremely rough surfaces with noncoalesced columns to a surface shown in Fig. 2(c), where very tall columns and terraces are separated by deep troughs.

Detailed investigations are necessary to get an insight as to the mechanisms involved. High growth rates mainly leading to Ga polarity and low growth rates to N polarity would indicate that there must be some atomic exchange or interaction that may be suppressed or promoted by large growth rates, depending on the case.

In conclusion, GaN films grown on high temperature

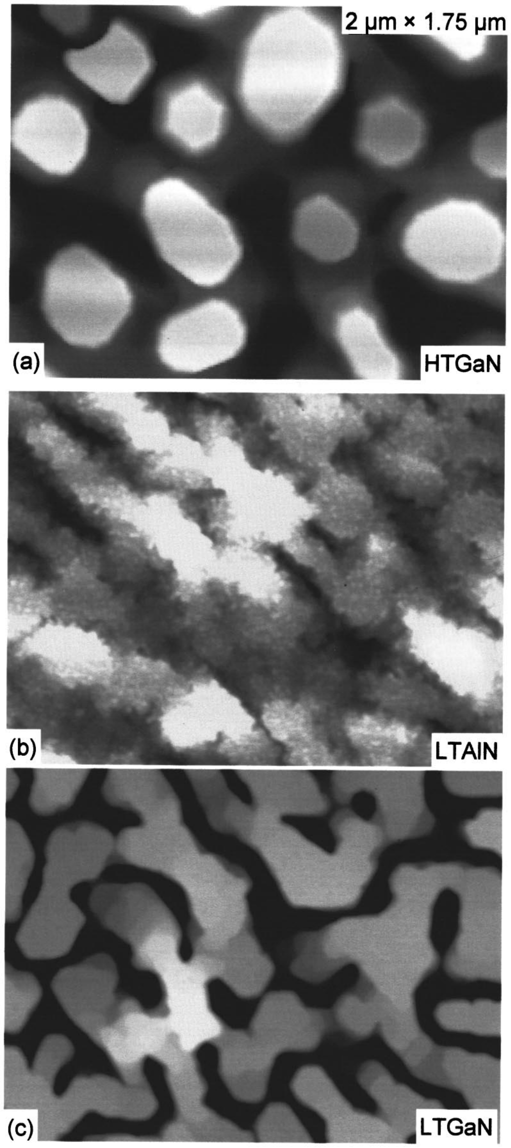

FIG. 2. AFM images of N-polar samples grown on three different buffer layers with different growth conditions: high temperature GaN buffer layer (a), low temperature AlN buffer layer at low growth rate (b), and low temperature GaN buffer layer at low growth rate (c). The vertical scales are 100, 30, and 75 nm for image (a), (b), and (c), respectively.

AlN ($>$890 °C) and GaN (770–900 °C) buffer layers by MBE invariably show Ga and N polarity, respectively. However, the polarity of the GaN grown on low temperature ($\sim$500 °C) buffer layers depends on the buffer growth rate. Consequently, if adequate care is not taken, mixed polarity growth with adverse consequences such as stacking mismatch and inversion domain boundaries can result.

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