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Optical and electrical properties of (1-101)GaN grown on a 7° off-axis (001)Si substrate

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Uniform growth of (1-101)GaN was performed on coalesced stripes of GaN which had been prepared by selective metalorganic vapor phase epitaxy on a 7° off-axis (001)Si substrate via an AlN intermediate layer. The cathodoluminescence spectra at 4 K exhibited a donor bound excitonic emission at 358 nm followed by defect-related emission peaks at 363, 371, and 376 nm. The 363 and 376 emission bands are observed upon the coalescence region. The Hall measurements exhibited *p*-type conduction at 80–300 K (the hole carrier density 6.3×10^{12} cm^{-2} and hole mobility 278 $cm^2/V\,s$ at 100 K). The activation energy of the acceptor was estimated to be 60 meV. The possible origin of the *p*-type conduction is discussed in relation to the unintentionally doped carbon. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1758300]

Most of the III-nitrides utilized for optical and electronic devices are of wutzite structure and prepared on a *c* plane of sapphire or SiC. A heterostructure grown on such a crystal is subject to the strong piezoelectric and/or pyroelectric nature of the crystal, which often determines the device performance.[1,2] If the growth is performed on a crystal plane other than (0001), we might reduce these effects to improve the device performances. Several attempts have been reported on the growth/properties of GaN on a different crystal plane, to reduce the stress/strain or dislocations.[3,4] Honda *et al.*[5] performed selective growth of GaN on a patterned Si substrate and achieved highly qualified crystal with less dislocation density. They demonstrated the growth of uniform (1-101)GaN on a 7° off-axis (001)Si substrate.[6] By tilting the crystal axis on the substrate, they reduced the thermal expansion coefficient mismatch and achieved a flat (1-101) surface without cracks. Moreover, because of the low growth rate as compared to the C face, the surface morphology has been much improved. In the conventional metalorganic vapor phase epitaxial growth (MOVPE) of wurtzite GaN, the growth is along the $+C$ axis. Therefore, the top face of the (1-101) GaN is of the N plane. This will modify the nature of the impurity incorporation as well as the growth modes. In this letter, the behavior of the band-edge emission and electrical properties of the (1-101) GaN are studied in detail.

First, the growth of (1-101) face GaN stripes were achieved on a 7° off-oriented (001) silicon substrate by a selective MOVPE method with an AlN intermediate layer.[6] A 70 nm thick SiO_2 film was deposited on the silicon surface. By conventional photolithography, a stripe mask pattern was made (the mask and window widths were 1 μm, or the period is 2 μm.) Then, the wafer was immersed in a KOH solution to get grooves. By virtue of the etching anisotropy, we got a groove of which bottom face was (001) and the side faces were of (111) and (-1-11) of the Si. In order to prevent the growth of GaN on the (-1-11) face, we deposited a thin SiO_2 film on this face. Thus, we obtained a patterned substrate which has (111) facets exposed to the air. The MOVPE growth was performed on this substrate. Trimethylaluminum, trimethylgallium, and ammonia were used as the source gases. A 70 nm AlN was grown on the patterned Si substrate at 1180 °C as an intermediate layer. Following the growth of intermediate layer, GaN was grown at 1120 °C for 20 min. By this procedure, we achieved coalesced GaN stripes of which the top face is (1-101). The surface of the sample was smooth and no cracks were found as shown in Fig. 1(a). The mean roughness measured with atomic force microscopy was as small as 0.3 nm.

On the thus prepared GaN templated Si substrate, a uniform growth of GaN was performed via a low-temperature-grown 40 nm thick AlN intermediate layer (LT-AlN). Figure 1(b) displays a picture of the sample grown with an LT-AlN layer. The thickness of the top (1-101)GaN layer was typically 500 nm. For the Hall measurements, Ti/Al or Ni/Au was deposited to form *p*- or *n*-type ohmic contacts followed by relevant heat treatment.

The cathodoluminescence (CL) spectra were analyzed at various temperatures. At low temperatures, the sample exhibited several near-band-edge emission peaks. The yellow band was very weak suggesting high crystal quality. Figure 2 shows typical spectra at 4 K obtained on different cross sections in the sample. Near the sample surface, the main peak was at 358 nm which is due to the neutral donor bound exciton (D_0X) followed by a subsidiary peak at 363 nm [Fig. 2(α)]. On the other hand, in the stripe structure region near

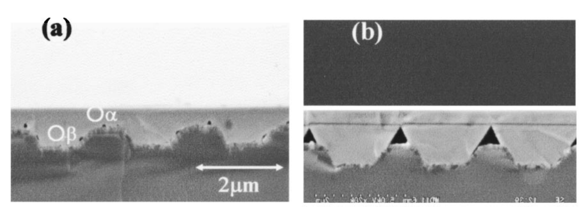

FIG. 1. Cross-sectional scanning electron micrograph image of the samples (a) without and (b) with an LT-ALN layer. The CL spectra shown in Fig. 2 are obtained at positions α and β.

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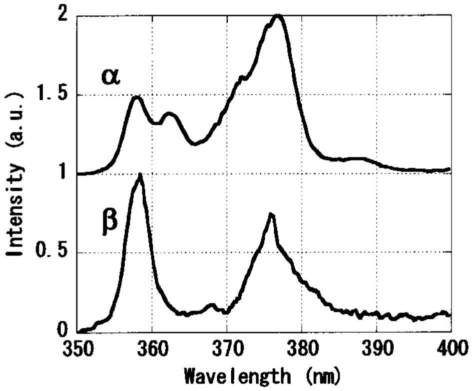

FIG. 2. CL spectra measured at 4.2 K. Top trace (α) is near the surface and bottom trace (β) is on the stripe structure [see Fig. 1(a)]. The top trace has two subsidiary peaks at 363 nm and 372 nm.

the Si substrate [Fig. 2(β)], the subsidiary peak at 363 nm does not appear. In both cases, we have donor-to-acceptor pair (DAP)-like emission at 376 nm which is followed by a longitudinal optical (LO) phonon replica. The cross-sectional CL images obtained at 358 nm, 363 nm, and 376 nm are displayed in Fig. 3. Apparently, the 363 and 376 emissions are strong upon/from the coalesced regions while the emission at 358 nm is strong in the region uniformly grown along the ⟨1-101⟩ axis. Note that Fig. 3 showed only the contrast of the emission intensity, and the 358 nm as well as 376 nm emissions were observed all over the sample.

The enhancement of the D_0X 358 emission band in the uniformly grown region on the (1-101) plane is in accordance with the observation by Shibata *et al.*[7] in an epitaxial-lateral-overgrowth (ELO) GaN by hydride vapor phase epitaxy, which has been adopted to get a GaN with less dislocation densities.[8] In the present case, the method is not ELO but uniform growth on the (1-101) plane. The strong 358 nm emission is the indication of the high optical quality of the sample, i.e., less density of dislocations/nonradiative centers.

On the origin of the 363 nm emission band, various reports have been published. Most of the studies suggest that it is related to some kind of defect.[9,10] Honda *et al.*[11] suggested it is related to oxygen. In Fig. 2(b), we found a shoulder peaking at 372 nm, which was occasionally enhanced in several samples. Sun *et al.*[12] studied the 371 nm emission in a sample grown on an *a*-plane substrate, and attributed the peak to stacking faults in GaN. Recent transmission electron microscopy analyses showed that we have stacking faults in the coalesced region in agreement with the present observation.[13]

In Fig. 2, we observed a DAP-like emission band at 376 nm followed by a weak LO phonon replica. Considering the strain due to the thermal expansion coefficient difference between GaN and the Si, the slight redshift observed in the D_0X emission should appear also in the DAP band. According to previous reports, the DAP emission band should appear at around 380 nm in our sample. Therefore, the present peak obtained at 376 nm is at a slightly higher energy than expected. Fisher *et al.*[14] attributed this band not to the DAP emission but to the electron acceptor emission. They obtained strong LO phonon replicas, which is, however, not the case here as is shown in Fig. 2. Reshchikov *et al.*[15] studied unusual emission lines in GaN. They found rich peaks with energies between 3.0 and 3.5 eV. Though we did not find corresponding energy in the report, the fact that the peak is associated with weak LO phonon replica suggests that the 376 nm band is due to a localized center which is specific to the (1-101) face.

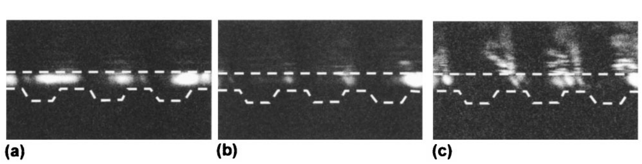

FIG. 3. Cross-sectional CL images for a sample shown in Fig. 1(a): (a) 358, (b) 363, and (c) 376 nm. The broken lines indicate the boundary of the GaN section.

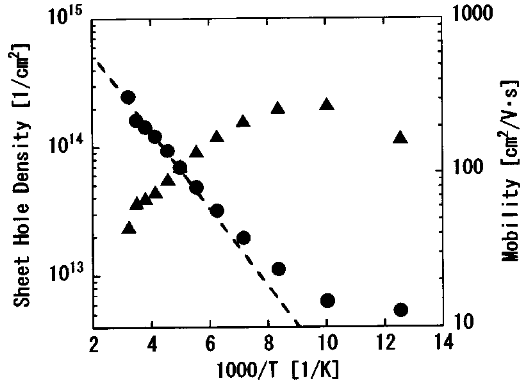

FIG. 4. Hole density (●) and mobility (▲) vs $1/T$ for sample (A).

The electrical characteristics of the sample were investigated by the van der Pauw method. The sample was set on a cold finger in a cryostat and the electrical resistivity and the Hall mobility was investigated at various temperatures. The Si substrate was *n*-type doped (the carrier concentration and mobility at room temperature were 1.6×10^{15} cm^{-3} and 1320 cm^2/V s, respectively.) We prepared three samples; (A) GaN grown on the LT-AlN layer [as shown in Fig. 1(b)], (B) GaN capped with a 30 nm thick $Al_{0.1}Ga_{0.9}N$ top layer, and (C) GaN grown without the LT-AlN intermediate layer [as shown in Fig. 1(a)]. All samples were not doped intentionally. But, we found that the type of conduction depends on the structure/temperature. In each sample, we tested both types of electrodes to determine the conduction type.

The sample (A) exhibited *p*-type conduction. Figure 4 shows the density and mobility of "holes" as a function of the temperature. Because of the insertion of "highly resistive" 40 nm thick LT-AlN layer, the *p*-type conduction might display the properties of 500 nm thick GaN. The sample resistivity increases by lowering the temperature due mainly to the decrease of the hole concentration. We could not get reliable data at temperatures lower than 80 K. We tested samples where the GaN top layer is 100 or 200 nm thick. But, the hole mobility and sheet carrier density were nearly equal to each other. These facts suggest that the conduction is not in the bulk GaN but near the GaN/LT-AlN interface. At low temperatures, the carrier density decreases gradually and we got the activation energy of 60 meV from the temperature dependence shown in Fig. 4. This suggests that we have a kind of acceptor level in the sample even though the sample was not intentionally doped.

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In sample (B), the type of conduction was *n* type. We got high "electron" density more than 1×10^{13} cm^{-2} irrespective of the temperature (40–300 K). The apparent mobility at room temperature was 70 cm^2/V s, which was higher than those obtained at lower temperatures. A slight increase (local maximum) of the apparent carrier density and the decrease (local minimum) of the apparent mobility were observed at temperatures between 80 and 200 K. The behavior suggests the parallel conduction (we should have more than two types of carriers) in the sample (B). In sample (C), the behavior was more complicated. At low ($T<80$ K) or high ($T>200$ K) temperatures, the sample exhibited *n*-type conduction, while it was *p* type at temperatures between 80 and 200 K. The apparent hole mobility was typically 35 cm^2/V s at 120 K.

Since the Si substrate is *n*-type doped, the contribution of the substrate is ruled out for the occurrence of *p*-type conduction. The effect of diffusion of Al and/or Ga into the Si substrate is not the case, because we have found high electrical resistance in a GaN/AlN/Si diode with more than 40 nm thick AlN layer in between.[16] The complicated transport in samples (B) and (C) is attributed to the parallel conduction and/or compensation of donors and acceptors. In addition, we should have the contribution of the transport near the AlN/Si interface.

We should have a piezo- or pyroelectric field at the GaN/AlN or AlGaN/GaN heterointerface in samples (A) and (B), respectively. The carrier might be induced there. Judging from the *p*-type conduction in the sample (A) and *n*-type conduction in the sample (B), the carrier transport is supposed to be dominated by the two-dimensional carrier gases in these samples. In sample (C), on the other hand, contribution of *n*-type transport in the bulk GaN and *p*-type transport near the GaN/AlN/Si interface might be responsible for the behavior observed.

The fact that the *p*-type conduction is associated with an activation energy of 60 meV suggests that we should have an acceptor level in sample (A). The hole mobility higher than 100 cm^2/V s along with high sheet carrier density obtained at low temperatures indicates the formation of two-dimensional hole gas. Since the GaN/LT-AlN is of inverse heterostructure in the conventional sense, we might get high hole accumulations at the heterointerface. But it is only possible if the material has been *p*-type doped. Secondary ion mass spectroscopy was performed and we found the incorporation of C as high as 1×10^{18} cm^{-3} throughout the sample. Previous studies showed that the C can be an acceptor in GaN. But in case of wurtzte GaN grown on a (0001) Ga face, no report on the *p*-type conduction has appeared in spite of the estimated activation energy lower than Mg. This is believed to be due to the amphoteric doping. In the case of cubic GaN, on the other hand, *p*-type conduction was achieved if the growth was under a N-rich condition.[17] In the case of AlN, C can be an active acceptor to provide *p*-type conduction which will be enhanced by codoping of oxygen.[18]

In the present case, the heterostructure doped with C might be responsible for the *p*-type conduction. The doping of C to be a donor will be limited on the (1-101) face, because it is a highly qualified N face. Because of the high V/III ratio, we have always kept a N-rich condition, that is to say, the (1-101) surface is filled with N immediately after the adsorption of Ga. The incorporation of C is more provable to replace N on the top most atomic layer. This should be achieved only if the surface is highly qualified (atomically smooth) and should be in contrast to the rough surface obtained on the (000-1) N face. The activation energy as low as 60 meV might be attributed to the heterostructure adopted. That is, most of the carriers are offered by the highly carbon-doped AlN layer and the transport is in the GaN layer. The situation is very similar to the *p*-type conduction in a Mg-doped GaN/AlGaN superlattice.[19] In order to establish the scenario, the doping property of Si and Mg on the (1-101) GaN is now under study, the results of which will be published elsewhere.

In summary, the optical and electrical properties of a uniformly grown (1-101)GaN have been investigated. Optical spectra suggested the high quality of the sample. In samples grown on a LT-AlN layer, the Hall measurement showed *p*-type conduction with an activation energy of 60 meV. The possible origin of the *p*-type conduction is discussed in terms of unintentionally doped C.

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