Stability of N- and Ga-polarity GaN surfaces during the growth interruption studied by reflection high-energy electron diffraction

Electrotechnical Laboratory (ETL), 1-1-4 Umezono, Tsukuba, Ibaraki, 305-8568, Japan

(Received 9 August 2000; accepted for publication 23 October 2000)

GaN films with N- and Ga-polarity were grown on sapphire (0001) substrates using different buffer layers by plasma-assisted molecular-beam epitaxy. The surface stability of each lattice-polarity film during the growth interruption was studied by reflection high-energy electron diffraction (RHEED).

It was found that the surface of N-polarity film was unstable against the exposure to the nitrogen plasma flux, while that of Ga-polarity one was stable. This provides a method to clarify the lattice polarity by the in situ RHEED observation directly. A model is proposed to explain the observed phenomenon, where the origin of the phenomenon is mainly attributed to the differences in surface dynamics processes and morphologies between the two kinds of lattice-polarity films. © 2000 American Institute of Physics. [S0003-6951(00)00452-6]

Recently, lattice polarity of III-nitride epitaxial films becomes a hot topic due to its great influence on the optical and electrical properties of the films. GaN films with Ga polarity have been demonstrating many advantages over those with N polarity, such as smooth surface morphology, stable chemical property, enhancement of two-dimensional electron density in AlGaN/GaN heterostructures, and so on. It is well accepted that Ga-polarity GaN films are usually obtained by metalorganic chemical vapor deposition growth, which is known to be the most suitable technique for III-nitride growth at present. The lattice orientation of molecular-beam epitaxy (MBE) films grown on a c-face sapphire substrate is known to be mainly N polarity. We have proposed that even in MBE technique, the GaN growth should be carried out under the Ga-polarity growth mode to obtain high quality films. Recently, excellent results of GaN films and AlGaN/GaN heterostructures have been reported by MBE growth technique, where the lattice polarity of those films are thought to be Ga polarity.

It is well known that growth interruption and migration enhanced epitaxy techniques are useful to improve the interface flatness for the GaAs growth and arsenic-related heterostructures. These techniques have been shown to be also promising in the III-nitride growth. Therefore, the surface stability problem during the growth becomes important, because the earlier techniques include the operations of source shutters. However, there is less information concerning the stability of the GaN surface, especially the comparison of the films with different lattice-polarity in MBE. Furthermore, there is no in situ characterization method to clarify the lattice polarity.

In this study, we investigated the stability of GaN surface with different lattice polarity by reflection high-energy electron diffraction (RHEED) technique. As a result, it was found that the surface of Ga-polarity film is much more stable than that of N-polarity one, by interrupting the growth against the plasma nitrogen flux exposure. A model considering the surface dynamics and surface morphology is proposed to explain the phenomena.

The GaN films with different lattice polarities were grown on sapphire (0001) substrates by rf plasma-assisted MBE (rf–MBE). We checked the lattice polarity by direct measurements by coaxial impact collision ion scattering spectroscopy, together with RHEED, secondary electron microscope (SEM) observations of surface morphologies, and chemical etching in NaOH solution. Details of the N- and Ga-polarity GaN films preparations and the clarifications of the lattice polarity will be published elsewhere. In our plasma nitrogen source apparatus, an ion deflector is equipped. We applied 500 V on the deflector to remove the ion damage from the growing surface. During the growth of GaN films, the N2-plasma power was 350 W and the N2 flow rate was 5.0 sccm. The growth temperature was fixed at 700 °C and the growth rate was 0.6 μm/h. In situ RHEED observations along the [11-20] azimuth during the growth were carried out and the RHEED images were recorded using a charge coupled device camera. The intensity change of specific RHEED spot/streak was measured from the recorded data using an image processor with a computer.

It is well known that if the growth is carried out under the N-rich condition in rf–MBE, the grown surface becomes rough resulting from the three-dimensional growth. Therefore, we avoided this growth regime and concentrated our interest on the two-dimensional growth regime with Ga-rich conditions. To study the stability of the GaN surface, we interrupted the growth by closing the Ga shutter and let the surface expose only to the plasma nitrogen flux at the growth temperature after 1.0 μm GaN growth. Figure 1 illustrates the RHEED patterns of N- and Ga-polarity films before and after the Ga shutter closing. During the growth, it is clear that both the N- and Ga-polarity films show streak RHEED patterns indicating the flat GaN surface under the two-dimensional growth [Figs. 1(a) and 1(b)]. However, the difference between the N- and Ga-polarity surface occurred significantly after closing the Ga shutter and exposing the surface only to the plasma nitrogen flux. First, for both of the

---

a)Author to whom correspondence should be addressed; on leave from New Energy and Industrial Technology Development Organization (NEDO); electronic mail: sxq@etl.go.jp
cases, the pattern became brighter than that during the growth, indicating the reduction of excess Ga atoms on the surface since the growth of GaN was under the Ga-rich condition. The similar phenomenon has been previously reported in the GaAs system. Second, the RHEED pattern of N-polarity surface changed from a streak pattern to a spotty one, illustrating the surface roughening. On the other hand, the RHEED pattern of Ga-polarity surface remained streak referring that the flat surface morphology was maintained. The earlier phenomena clearly demonstrated that the surface of Ga-polarity film is much more stable than that of N-polarity one against the plasma nitrogen flux exposure. This conclusion provides a method to directly clarify the lattice polarity by the in situ RHEED observations. Surface stability problem is specially important for InGaN/Al, GaN quantum well fabrications, because interruption of the growth to decrease the growth temperature is necessary for InGaN growth.

It is obvious that the RHEED patterns of both N- and Ga-polarity films became bright after closing the Ga shutter as shown in Fig. 1. Monitoring the RHEED intensity change could pick out the differences between them. Figure 2 illustrates the in situ intensity changes of RHEED patterns of N- and Ga-polarity films. It is clear that the intensity for the N-polarity film increases gradually (dotted line) to reach the saturation level. On the other hand, the intensity for the Ga-polarity film (solid line) is enhanced rapidly followed by a gradual increase to the saturation level. The difference of the RHEED intensity change between the N- and Ga-polarity film is attributed to the different dynamic processes on the different surface features as discussed later. Concerning the surface morphology, it is well known that N-polarity films always show grain features, while each grain surface is flat. On the contrary, Ga-polarity films illustrate uniform and smooth surface morphology. We actually observed such morphological difference between our N- and Ga-polarity films as shown in Fig. 3.

Figure 4 illustrates our proposed model schematically to explain qualitatively the surface stability phenomena. It is necessary to notice that the growth was carried out under the Ga-rich condition. Therefore, the excess Ga atoms exist on the growing surface. Smith et al. reported that N-polarity surface with a Ga adlayer showed a $(1\times1)$ RHEED pattern and adatom-on-adlayer resulted in a $(3\times3)$ pattern. To sim-
In the extremely favorable reactions between GaN and N to since a single Ga–N bond is exposed to the N flux resulting GaN may be greatly enhanced in the grain boundary region, as shown in Fig. 3.

Further, surface morphology containing grain features as shown in Fig. 3(a) is also considered. In the case of Ga-polarity surface, we suppose that the excess Ga atoms on the top surface form a Ga metal layer, which are weakly bonded to the Ga atoms just below them. Earlier images of the different polarity surfaces are believed to be reasonable for the growth under the Ga-rich conditions.

First, we argue the N-polarity case. As illustrated in Figs. 4(a) and 4(b), crystallization and re-evaporation processes of the excess Ga atoms within the Ga adlayer would take place after closing the Ga shutter. Since only nitrogen atoms come to the surface, a part of the excess Ga atoms on the surface would be incorporated into the GaN crystal. Meanwhile, the other Ga atoms evaporate from the surface, leading to the surface roughening. In addition, grain boundary area including a high dislocation density is considerably unstable. Recent in situ transmission electron microscopy results proved that nitrogen preferred to desorb along the cores of dislocations. Furthermore, the thermal decomposition of GaN may be greatly enhanced in the grain boundary region, since a single Ga–N bond is exposed to the N flux resulting in the extremely favorable reactions between GaN and N to form Ga and N₂. Therefore, the boundary region will expand and act as another cause for the surface roughening. The earlier two mechanisms are believed to coexist during the surface roughening processes.

It is much easier to understand the surface dynamic process of Ga-polarity films based on our model. It is assumed that the Ga–Ga bond is weaker than the Ga–N one (2.17 eV). Therefore, Ga atoms within the metal layer during the growth are easily evaporated from the surface after stopping the Ga flux supply as shown in Figs. 4(c) and 4(d), resulting in a quick increase of the RHEED intensity to the saturation level as shown in Fig. 2. Since the Ga atoms below the metal layer have three Ga–N bonds to the N beneath them, these Ga atoms are much stable on the surface. In addition, uniformly flat surface will prevent the decomposition of GaN.

In summary, the surface stability of GaN films with N and Ga polarities during the growth interruption was studied by RHEED. It was found that the surface of Ga-polarity film was much more stable than that of N-polarity one against the nitrogen plasma flux exposure. This provides a method to clarify the lattice polarity by the in situ RHEED observation directly. The origin of the stability problem was attributed to the differences in surface dynamic processes and morphologies between the two kinds of lattice-polarity films.