Luminescence from growth topographic features in GaN:Si films

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Cathodoluminescence (CL) in the scanning electron microscope is used to investigate the nature of defects responsible for the luminescence associated with round and hexagonal-like topographic features of GaN:Si films. Round hillocks of the size of a few microns, which sometimes have a nanopipe related central hole, do not influence the luminescence emission of the film. Hillocks with sizes of several tens of microns show a marked CL contrast at the center and at the border. The origin of the observed contrast is attributed to a growth induced inhomogeneous distribution of point defects and impurities. Radiation with the electron beam of the scanning microscope causes a decrease of the CL intensity without spectral changes. © 1998 American Institute of Physics.

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I. INTRODUCTION

The fabrication of blue light emitting diodes and lasers from GaN-based materials is the cause of an increasing interest in the study of the optical properties of defects in III-V nitrides. One of the techniques used to investigate the recombination properties of defects in semiconductors is the cathodoluminescence (CL) in the scanning electron microscope (SEM). It has been recently applied to characterize epitaxial GaN films and has provided information on the spatial distribution of band edge, yellow and red emission bands, and their association to extended defects. One of the structural features often reported in microscopic observations of the surface of GaN epitaxial films is the presence of growth hillocks with a wide range of sizes, typically several microns, with a hexagonal shape. Qian et al. have reported observations relating the hexagonal hillocks to the presence of nanopipes emerging at their centers and concluded that nanopipes are the open cores of screw dislocations. Middleton et al. in a CL study of pyramidal hillocks found that in the center only the yellow emission is present and suggested that it originates from defects associated with inversion domain boundaries. In the present work CL microscopy is used to investigate the luminescence of hexagonal-like and round topographic features in Si doped GaN films. The results are discussed in terms of the structural defects associated with the hillocks.

II. EXPERIMENTAL METHOD

The samples used were GaN films grown on (0001) sapphire. The films consisted of an 8-μm-thick GaN buffer layer and two 2-μm-thick epilayers. The upper layer was Si doped with a carrier concentration of about 10^{19} cm^{-3}. The films were observed in the secondary electron and cathodoluminescence (CL) modes of scanning electron microscopy by using an Hitachi S-2500 or a Leica S-440 scanning electron microscope with an electron beam energy of 15 keV. CL measurements were performed at sample temperatures between 77 and 300 K in the visible range with a Hamamatsu R928 photomultiplier. An Oriel computer controlled monochromator was used for spectral analysis of the emission.

III. RESULTS

Secondary electron images show a distribution of round hillocks of a size of about 5 μm in a smooth background (Fig. 1). Most of the hillocks show an almost circular shape and in some of them some corners indicate the appearance of a not well defined hexagonal shape. In general, no steps or terraces have been observed in the surface of the hillocks with the exception of a central circular step in some of them. In some cases high magnification SEM images (Fig. 2) revealed a hole, with a size of about 500 Å, in the center. The hexagonal shape reported in the literature is here observed only in the relatively few hillocks present with sizes of about 40 μm or higher (Fig. 3).

CL spectra of an extended area of the sample show luminescence bands centered at about 360 and 545 nm which correspond to the band edge emission and to the defect related yellow emission, respectively. The relative intensity of both bands is a function of temperature and of excitation density. Figure 4(a) shows a series of CL spectra at different temperatures under constant high excitation conditions. A decrease of the excitation intensity leads to a relative increase of the band edge emission and to the appearance of weak bands centered at about 430 and 730 nm [Fig. 4(b)]. CL images show a contrast with bright and dark regions which seems to be related to the grain structure of the films (Fig. 5). The presence of the small hillocks does not modify the CL contrast or cause slight variations relative to the background as in Fig. 6. On the contrary, the big size hillocks present a well defined CL contrast at the hillock boundary in panchromatic and monochromatic images (Fig. 3).

IV. DISCUSSION

The inhomogeneous distribution of CL intensity shown in Fig. 5 is similar to that reported in Refs. 3 and 9. This contrast, which has not corresponded with surface features, is similar to that observed in diamond films (e.g., Ref. 10) and could be related to the grain structure and to the differences

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of emission among the different grains as well as to grain boundary effects. The CL images of Figs. 5(b) and 3 show that only hillocks with a big size markedly influence the CL intensity distribution in the film. In these hillocks three regions—center, large intermediate region, and border—with different luminescent behavior are observed. Sometimes a hole appears in the center, as in Fig. 2, while in other cases (Fig. 3) a region of several microns shows a topographic as well as a CL contrast which is shown in detail in Fig. 7. These different hillocks, or islands, have been described by Liliental-Weber et al., in a study by atomic force microscopy and transmission electron microscopy. They reported that at the center of some islands there is either an inversion domain or a twinned precipitate, which appears to be the case shown in Fig. 7, while other islands show a central hole as in Fig. 2. The central hole had been related to nanotubes which are the open cores of screw dislocations, but in Ref. 11 it was found that a fraction of the hollow defects is not associated with dislocations, but to inversion domains. The association of inversion domains with pyramidal hillocks has also been reported by Daudin et al., who found many tiny columnar inversion domains in the pyramids. These structural observations open the question of the relationship between the yellow luminescence at the center of the hillocks and inversion domains. Our results clearly show that the enhancement of the yellow luminescence takes place only at the center of the hillocks which seems to exclude the inversion domains, in principle distributed all over the hillock, as the origin of the enhancement. On the other hand, calculations of electronic structure have shown that the presence of an inversion domain boundary does not induce electronic states in the band gap while stacking mismatch boundaries lead to electronic states in the gap with transition energies in the yellow range. It appears possible that the yellow emission at the center of some of the hillocks is related to some stacking defect. The main part of the hillock—the intermediate region—shows an enhancement of the 430 nm emission relative to the film background. It has been previously observed that the centers responsible for this band decorate the grain boundaries, which indicates that they are formed by point defects and/or impurities. The concentration of point defects appears then to be different in the hillocks than in the film matrix. Inhomogeneity of the impurity/dopant distribution in the hillocks has been detected by Raman measurements in Si doped GaN films. The Raman results showed a reduced dopant concentration in the center and in the border of the hillock as well as a general reduced concentration in the hillock surface as compared with the film background. In Ref. 15 it is mentioned that segregation of oxygen could

FIG. 1. Secondary electron image of round hillocks.

FIG. 2. Secondary electron image of a round hillock which reveals the presence of a hole with a size of about 500 Å.

FIG. 3. A big hexagonal hillock. (a) Secondary electron image (b) panchromatic CL (c) CL at 360 nm (d) CL at 430 nm, and (e) CL at 550 nm.
FIG. 4. CL spectra recorded at: (a) high excitation conditions and different temperatures (b) low excitation conditions.

contribute in part to the contrast of the Raman scattering image which shows three regions with similarity to the regions observed in CL images. These results support the possibility that the CL contrast of the hillocks, and in particular the contrast at the border, is due to a distribution of point defects and impurities produced during growth. The fact that in the smaller hillocks almost no CL contrast is observed indicates that they are in a state of the formation in which the rearrangement of dopant atoms or point defects has not taken place at an appreciable scale.

CL features of pyramidal hillocks were described in Ref. 8 and are only partially coincident with the contrast of the big hillocks described here. For instance, in our case the 360 nm band appears only enhanced at the center and at the border while in the pyramidal hillock all the surface, with the exception of the center, shows enhanced emission in a close spectral-range. As stated above, the centers of the hillocks can have a different nature which would lead to different CL contrast. On the other hand, the hillocks observed here have a flat surface which suppresses any edge or orientation effect and enables the obtention of CL images in which luminescent regions are more clearly identified.

FIG. 5. (a) Secondary electron image with some round hillocks, and (b) panchromatic CL image of this zone.

FIG. 6. (a) SEI and (b) panchromatic CL image of a small hillock.
Round shaped hillocks of a few microns size, in some cases related to nanoparticles, show the same CL contrast than the film background. Hillocks with sizes of several tens of microns show a complex CL contrast involving the 360, 430, and 550 nm bands. Analysis of the contrast indicates that an enhancement of the yellow emission observed at the center of the hillocks could be related to some stacking defect. CL contrast at the hillock surface and at the border is attributed to an inhomogeneous distribution of point defects and impurities. Electron irradiation in the SEM does not induce changes in the defect structure of the films.

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