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Reduction of dislocation density in epitaxial GaN layers by overgrowth of defect-related etch pits

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GaN templates grown by the metal organic chemical vapor deposition method were etched in a defect-selective molten salts eutectic and were subsequently overgrown by a GaN layer using the hydride vapor phase epitaxy (HVPE) method. Optimized conditions of etching and of HVPE growth processes resulted in a significant reduction of the dislocations density (DD). Local areas virtually free of dislocations were obtained on ~50% of the surface, while the average DD was reduced from 3×10^9 cm^{-2} in the template to about 2×10^7 cm^{-2} in the HVPE-grown GaN layer. A model has been developed to explain the mechanism of reduction of the DD during the overgrowth process. The model was confirmed by the photoetching of cleaved layers. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3171928]

Production of large area GaN substrates with low dislocation density is still a challenging issue. The lack of large size single crystals enforces GaN heteroepitaxy. Low price and easy availability make sapphire one of the best substrates for GaN heteroepitaxy. In addition, GaN epitaxy on sapphire is quite mature and the material quality is comparable to that obtained on other substrates such as silicon carbide (SiC), silicon (Si), etc.[1] Large thermal and lattice mismatch lead, however, to various problems, one of which is high dislocations density (DD), i.e., of the order of 10^8-10^{10} cm^{-2} in the GaN layers, which is not acceptable for optoelectronic devices.

Several methods have been devised to improve the quality of metal organic chemical vapor deposition (MOCVD)-growth of GaN material with respect to the high DD, e.g., insertion of a SiN layer,[2] optimization of aluminum nitride (AlN) nucleation layer,[3] interrupted growth with annealing step in silane,[4] deposition of second phase nanoislands,[5] growth on nanoporous GaN templates,[6] epitaxial lateral overgrowth, and its modifications.[7–11] However, these methods are complicated and time consuming and usually only a small area of high quality material is available to fabricate devices.

In this communication we report a simple method to achieve reduced DD in GaN layers grown by hydride vapor phase epitaxy (HVPE). The main step in achieving this goal is to use defect-selective etching of MOCVD or HVPE-grown templates to produce pits which can be subsequently overgrown without continuation of the line defects into the homoepitaxial GaN layer. The quality of templates and the etching conditions required for obtaining reduced dislocation density are discussed.

MOCVD-grown samples obtained from commercial vendors and from scientific laboratories were etched in a eutectic (E) of NaOH and KOH at 400–450 °C for several minutes.[12,13] The etch pit density (EPD) in these templates was 3×10^9 cm^{-2} as calculated from scanning electron microscopy (SEM) images. Etched templates were cleaned ultrasonically in hot HCl, water and isopropanol to remove remaining etching products and dirt particles. GaN layers of thickness 15–20 μm were grown at a growth rate of 100 μm h^{-1} on the etched templates in a home-made horizontal HVPE reactor.[14] The precursors for the growth process were ammonia (NH_3) and gallium chloride ($GaCl_3$). In order to avoid decomposition of the GaN templates, NH_3 was introduced into the reactor at 400 °C. The growth was performed under optimized conditions to achieve a high lateral growth rate. A high lateral growth was achieved by optimizing the V/III ratio (83), the pressure (950 mbar) and temperature (as high as 1070 °C).

After the HVPE growth of 15–20 μm thick epilayers the samples were again subjected to molten E etching in optimal conditions for revealing the density, type, and distribution of dislocations. Some samples were cleaved in order to examine the interface between the template and the HVPE-grown layer. The as-cleaved and photoetched[12] cross sections and the growth surfaces were examined using differential interference contrast (DIC) optical microscopy and SEM.

From the previous detailed studies of selective etching of GaN in molten salts[12,13] it follows that orthodox etching allows us to distinguish edge, mixed, and screwtype dislocations from the different size of pits. Several overgrowth experiments on various types of substrates lead to the conclusion that the reduction of DD can be obtained only on templates with a relatively large number of dislocations with screw components. However, the standard etching procedure in molten E on these templates, which is used for revealing and counting dislocation density (EPD), yielded minor and only local decrease of dislocation density in the HVPE-grown layer. This effect is shown in Fig. 1: the overall EPD is similar in the template [Fig. 1(a)] and in the subsequently grown HVPE layer [Fig. 1(c)], with the exception of some local spots with markedly lower EPD [Figs. 1(b) and 1(c)]. Within the spots no large etch pits are observable, indicating the absence of dislocations with a screw component and the density of small pits is also slightly diminished. This obser-

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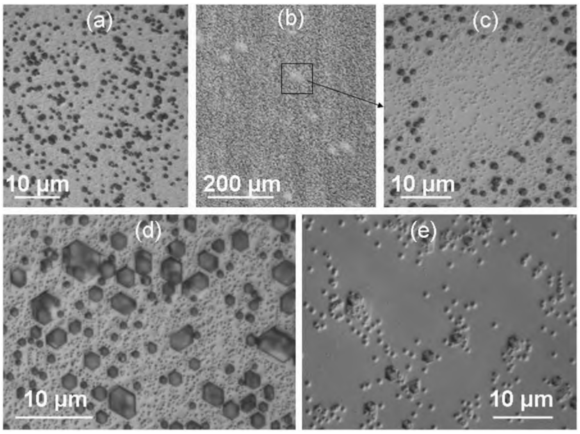

FIG. 1. DIC optical images of (a) MOCVD-grown template after E etching (note uniform distribution of large and small, background pits), (b) the same sample after HVPE growth of 20 μm thick GaN layer and E etching (this low magnification image demonstrates uniform overall distribution of pits with the exception of local spot marked by the frame), (c) fragment of image from (b), (d) MOCVD-grown template [(sample cleaved from the same wafer as that shown in (a)] after prolonged E etching, and (e) the same sample after HVPE growth of 20 μm thick GaN layer and standard E etching.

vation lead to the conclusion that mainly large etch pits formed on screwtype dislocations are actually overgrown without continuation of the underlying dislocations. The subsequent overgrowth experiment performed on a few samples containing a large number of screwtype defects and "overetched" to produce very large and deep pits [Fig. 1(d)] yielded GaN layer with markedly reduced dislocation density, as demonstrated in Fig. 1(e). Over the whole sample there are areas virtually free of dislocations (~50% of the surface) surrounded by a cellular network of material containing about 8×10^8 cm^{-2} EPD. The average density of dislocations is about 2×10^7 cm^{-2}, i.e., is two orders of magnitude lower than in the MOCVD template.

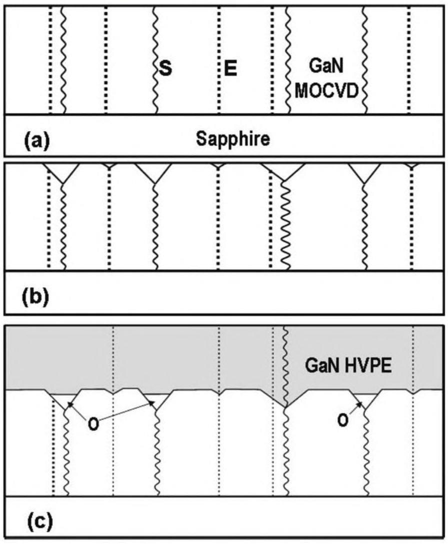

FIG. 2. Schematic drawing of steps involved in the process of reduction of dislocation density: (a) MOCVD GaN template (E—edge dislocations, S—screw/mixedtype dislocations), (b) template after deep etching in molten salts, (c) structure of the top HVPE-grown GaN layer: overgrown, i.e., empty large etch pits (**O**) are the reasons of reduction of dislocation density.

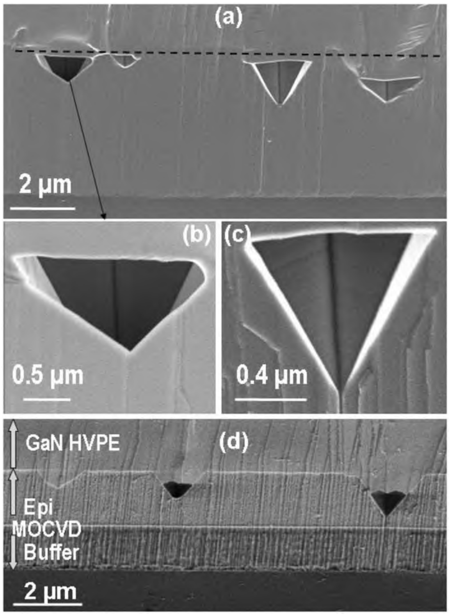

FIG. 3. SEM images of as-cleaved [(a)–(c)] and photoetched (d) sample after "overgrowth." The dashed line in (a) indicates the position of interface between MOCVD template and HVPE-grown layer. In (d) photoetching revealed exact interface between MOCVD- and HVPE-grown GaN layers.

On the basis of the experimental results a model was developed (see Fig. 2), to explain the mechanism of the observed reduction of dislocation density during the HVPE growth of GaN layer on etched GaN template. As in most GaN layers, both dislocations with edge (E) and screw (S) components of Burgers vector are present in the GaN templates [Fig. 1(d)]. Deep etching in molten E results in formation of very large and deep etch pits on screwtype dislocations (with side walls of pits inclined 30°–60°), while small and shallow pits (with side walls inclined 10°–25°, after[13]) are formed on edgetype dislocations, compare Figs. 1(d) and 2(b). During etching, these large pits "swept" small pits formed on edgetype dislocations situated in the vicinity of them [see Fig. 2(b)]. During subsequent growth on the etched template the largest pits were overgrown without complete filling and the underlying dislocations do not thread into the new GaN layer, while shallow etch pits formed on the edgetype dislocations are filled and the dislocations continue into the GaN layer [Fig. 2(c)].

In order to confirm the devised model several cleaved sample were examined. Characteristic images of overgrown (empty) pits are shown in Fig. 3(a). It is worth noting that these large pits are partially filled with GaN during HVPE growth. From SEM images and contrast analysis the position of the interface between the MOCVD and HVPE-grown material could be established and is marked by a dashed line in Fig. 3(a). Clearly, the top parts of the pits are filled with the HVPE-grown GaN, but the bottom of the pits remains empty, which prevents threading of the underlying dislocations into the HVPE-grown material. It can be assumed that the same holds for the edge dislocations which terminate at the sides of large pits. The inclination angle of the side walls is in the limits defined previously,[13] but can be measured exactly only on some pits, in which the cleavage plane is by coincidence exactly in the center [see e.g., the pit in Fig. 3(c): the inclination of the side walls is close to 60°]. As a rule, the pits with larger inclination angle of the side walls are overgrown and have empty bottom parts. This leads to the conclusion that both a large size (especially depth) and the geometry of the pits on screwtype dislocations are crucial for effective reduction of dislocation density in the second GaN layer.

Photoetching of the cleavage plane of sample after HVPE growth provided final confirmation of the model discussed above. Figure 3(d) shows one shallower etch pit completely filled and two deeper pits partially filled during HVPE growth, but with empty bottom parts. From this image it can be suggested that the pits should be at least 1.5 μm deep in order to be overgrown, to prevent threading of the dislocations from the template into the HVPE GaN layer.

It has to be mentioned here, that defect-selective etching before homoepitaxy was previously used to eliminate defects in other materials, e.g., basal plane dislocations (BPDs) in SiC.[15] In this case, however, the detrimental for PIN diodes BPDs were converted into threading edge dislocations and, as opposed to our overgrowth method, the overall dislocation density was not changed. The underlying mechanism to this concept relied on the gain of energy of dislocations, while the pits constituted the privileged sites for conversion of dislocation type and were filled during epitaxy.

In conclusion, reduction of dislocation density during HVPE growth of GaN on high dislocation density MOCVD template can be ensured when:

(a) there is a high density of screwtype dislocations in the template;
(b) defect-selective etching results in formation of large and deep pits, preferably with as large as possible inclination angle of the side walls; and
(c) the GaN layer of the template is thick enough (at least 3 μm) in order to ensure deep pits on screwtype dislocations.

It can be also suggested that further reduction of dislocations can be realized by repeated deep etching and growth procedure that could eliminate remaining screwtype dislocations [see large pits in Fig. 2(b)] and the vicinal edge dislocations threading into the HVPE layer.

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