Cathodoluminescence and TEM studies of HVPE GaN layers grown on porous SiC substrates

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Plain-view-field transmission electron microscopy and cathodoluminescence are used to study the defect structure of GaN films grown by hydride vapour-phase epitaxy on porous and non-porous SiC substrates. It is shown that the use of porous substrate reduces the mosaic structure of the films. This finding supports the compliance of porous SiC substrates, which was proposed by the authors earlier.

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1. Introduction

In recent years, new methods of substrate engineering have been developed in order to lower defect density in heteroepitaxial layers. The concept of these methods is based on mismatch strain management in the epitaxial film/substrate system, in an attempt to reduce introduction of defects, which result from mismatch strain relaxation. In particular, recently it has been demonstrated that GaN growth over porous SiC (PSC) allows for improving the quality of the epitaxial films by reducing residual strain and threading dislocation density [1–4]. This results in the improvement of optical properties of the films [5]. In this paper, we investigate hydride vapour-phase epitaxy (HVPE) GaN growth over PSC by comparative analysis of the data acquired by cathodoluminescence (CL) and transmission electron microscopy (TEM). We show that the films grown on PSC possess more coherent mosaic structure as compared to those grown on non-porous SiC. This finding confirms the compliance of our porous SiC substrates.

2. Experiment

PSC layers were fabricated by surface anodization of commercially available *n*-type 6*H*-SiC wafers with 2 in. in diameter. The electrical resistivity of the starting wafers was in the range of $0.05-0.07\,\Omega\cdot\mathrm{cm}$. To compare the properties of GaN films grown on porous substrates with those grown on non-porous SiC, PSC was fabricated only on a half of each SiC wafer. PSC was prepared at anodization current density $j = 4-16\,\mathrm{mA/cm^2}$ in 3 vol% aqueous solution of fluorine acid. As we have shown earlier, such anodization conditions produce nano-porous SiC layers with 30% bulk porosity; these layers demonstrate no changes in stoichiometry (in terms of Si/C ratio) in respect to initial SiC, and have monocrystalline structure of the SiC compound [6,7]. $1\,\mu$m thick GaN films were grown directly on both PSC and non-porous SiC by HVPE [8].

TEM investigation was carried out on JEM-T6 (JEOL) electron microscope. Defect density in the films was determined on plain-view bright-field TEM images. Two-beam conditions were used in order to image dislocations with different Burgers vectors.

CL study was carried out at the room temperature using Camebax electron microprobe. The original CL spectrometer [9] was installed into an optical microscope port of the microprobe. CL spectra were recorded at electron beam energy of 1, 5 and 15 keV with electron current of 30 nA and electron beam diameter of $1\,\mu$m. Under these conditions the CL signals were registered from the depth of 0.1, 0.5 and $1.3\,\mu$m, correspondingly. CL images were obtained from the mid-part of the GaN films using defocused primary electron beam with a diameter of $100\,\mu$m at 5 kV accelerating potential and electron current of 30 nA. The combination of CL imaging and CL spectroscopy allowed us to get spatially resolved secondary electron images to characterize relatively large parts of the object under study, and to record CL spectra in specific areas.

3. Results and discussion

The CL spectra recorded at 1 and 15 kV accelerating potential are shown in Fig. 1. The depth-resolved CL measurements showed that for all samples under investigation the CL spectra were dominated by a luminescence band corresponding to donor-acceptor-pair (DAP) emission (3.24 eV). The average increase in the emission intensity by a factor of 2.5 was observed for GaN films grown on PSC as compared to those grown on SiC. This finding agrees with earlier obtained photoluminescence (PL) data, where the major observation was a substantial increase of the PL intensity for the films grown on PSC as compared to those grown on non-porous SiC [5]. This increase was associated with the reduction in the density of non-radiative recombination centers, namely, threading dislocations (TDs) [5].

TEM data acquired in the current study point out to one more substantial distinction concerning the crystalline structure of GaN layers grown on PSC and SiC. Fig. 2 illustrates the difference in defect structure of GaN films grown on porous and non-porous substrate. Presented in Fig. 2 plain-view TEM images indicate that the majority of TDs, which intersected GaN surface, were arranged in arrays. As can be seen in the insert in Fig. 2, *b*, each

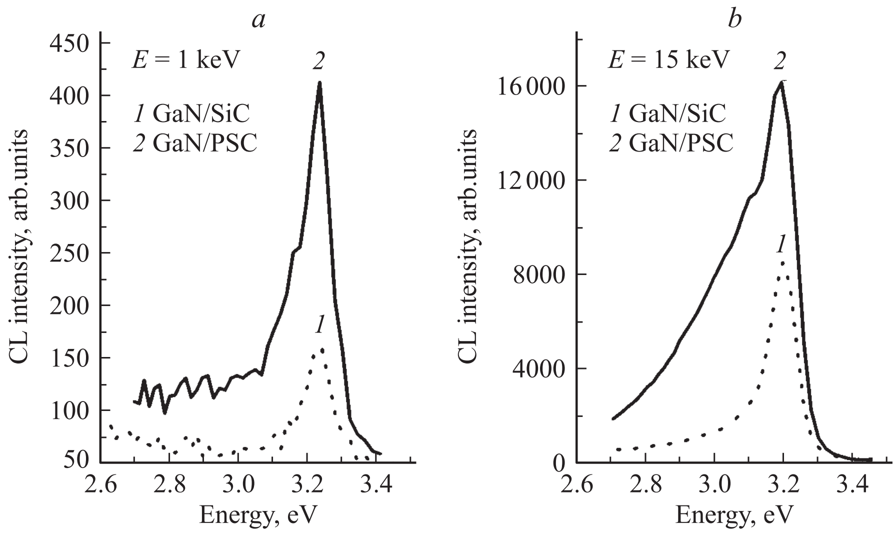

Figure 1. Cathodoluminescence spectra of GaN films grown on porous (PSC) and non-porous SiC, recorded at 1 kV (a) and 15 kV (b) accelerating potential.

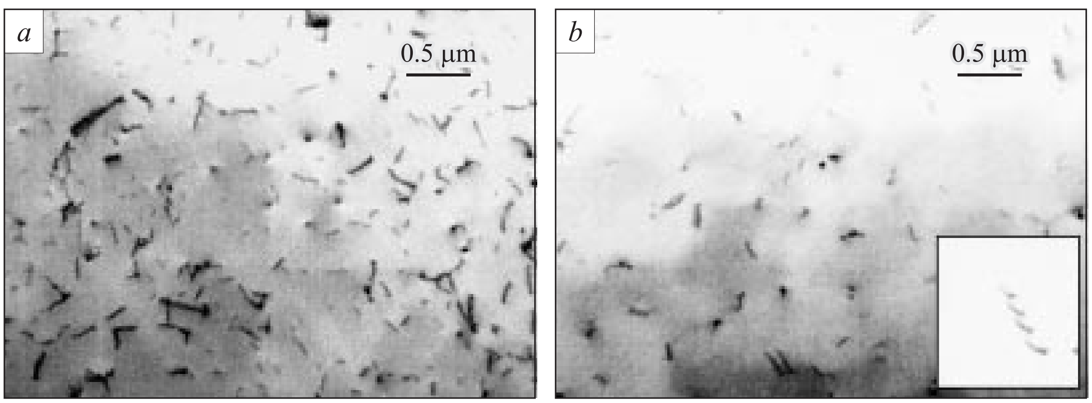

Figure 2. Plain-view TEM images of epitaxial GaN films grown on non-porous (a) and porous (b) SiC substrates.

array consisted of a set of parallel dislocations that had similar oscillatory contrast. This indicated that these TDs had similar direction, inclined relative to the normal to the layer surface. Such dislocations are known to form at grain boundaries between neighboring growth domains (associated with three-dimensional growth mode due to film/substrate mismatches) to accommodate slight tilt and/or twist between them. The clear difference in the dislocation density distribution in the films grown on conventional (non-porous) and porous substrates suggests that the use of PSC has the benefit of eliminating misorientation of the domains as compared to non-porous SiC. Hence, in addition to observation of drastic reduction in overall threading dislocations density reported earlier [4], TEM results acquired in this study suggest that using of porous substrate results in more coherent mosaic structure of the GaN films.

We consider this phenomenon as an extra evidence of the compliance of PSC substrate, which was discussed in our previous study, where we have shown that PSC allows for reducing the substrate/film mismatch strain in GaN/SiC system [4].

The validity of this conclusion was checked by a room-temperature panchromatic CL imaging. Fig. 3, a, b presents panchromatic CL images of the surface of GaN films grown on non-porous SiC and PSC substrates. The essential spatial inhomogeneity of luminescence was observed in the GaN/SiC layers (Fig. 3, a). This effect is known to be typical for GaN epitaxial films, and is associated with their columnar structure [10]. At the same time, more coherent emission was observed from the sample grown on porous substrate (Fig. 3, b). The difference between CL images presented in Fig. 3, a and 3, b maintains the conclusion on higher crystalline coherence of the GaN mosaic structure for the films grown on PSC as compared to those grown on non-porous SiC.

The CL spectroscopy study shows that the bright spots on the images (marked by black arrows) represent strong CL emission at 2.2 eV as shown in Fig. 4, a (curve *1*). It corresponds to yellow luminescence associated with the

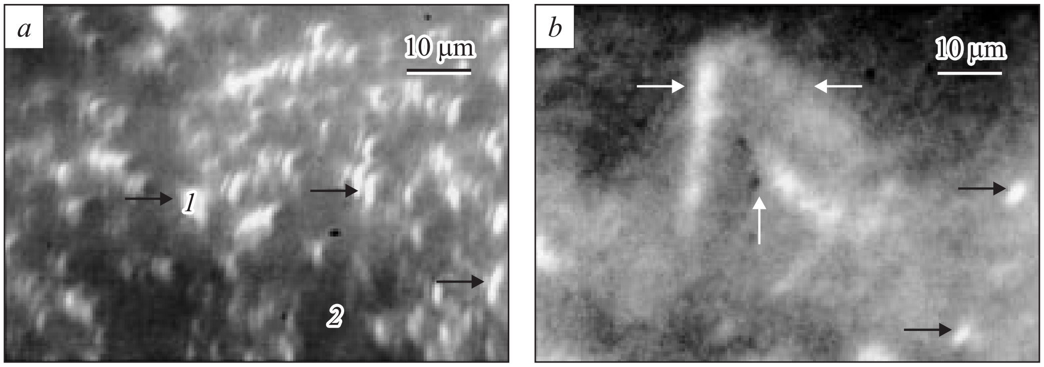

Figure 3. Panchromatic cathodoluminescence images of the surface on GaN films grown on non-porous SiC (a) and porous SiC (b).

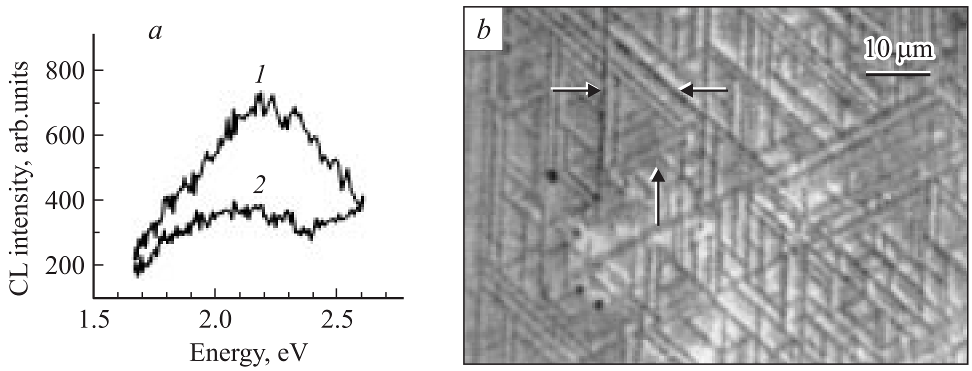

Figure 4. Cathodoluminescence spectra of an epitaxial GaN film (a) recorded from the spot with bright yellow emission (curve *1*) and from the dark area, where donor-acceptor-pair emission dominates (curve *2*). b — Lang topography image taken from the GaN/PSC sample.

presence of specific defects such as dislocations at low-angle grain boundaries [10]. The darker areas on the CL images from both GaN/SiC and GaN/PSC samples (as seen in Fig. 3, a, b) correspond to the DAP band emission. This emission most probably originates from the bulk of the columnar grains, which compose the microstructure of the GaN films. As shown in Fig. 4, a, the decrease of the yellow luminescence intensity by a factor of 2.5 was observed for the areas with darker emission. This emphasizes higher structural perfection of these areas. The image obtained from the GaN/PSC sample being dominated by the darker areas confirms the conclusion on more coherent mosaic structure of the films grown on PSC.

Yet another important feature was observed on CL images from GaN films grown on PSC, which showed „figure-of-line" luminescence sources indicated by white arrows in Fig. 3, b. As was reported earlier, GaN growth on PSC substrate is accompanied with spesific mismatch strain relaxation, which occurs via formation of planar defects (PDs). These defects form in GaN film region adjacent to the film/substrate interface, and suppress propagation of dislocations into the growing layer [4]. Fig. 4, b presents a Lang topogram of the as-grown GaN/PSC structure, which illustrates the occurrence of PDs within the GaN film in the vicinity of the substrate. The image shows the portion of the film containing PDs, where their boundaries are represented in the form of a cross-hatch pattern. The features corresponding to similar pattern observed on the CL image (Fig. 3, b) are indicated by arrows on the topogram. Thus, the „figure-of-line" CL features can be considered as originating at the PD boundaries. The PDs-related features, which appear on CL images, along with very low density of defects related to domain structure of GaN films confirm the validity of the conclusion of an ability of the porous SiC substrate to improve the quality of GaN films through the mechanism suggested in [4].

4. Conclusion

In conclusion, we have studied GaN films grown by HVPE on porous and non-porous SiC substrates. The complementary TEM and cathodoluminescence methods allow for providing the detailed study of the film quality. The experimental results acquired in this study give new supplemental data concerning improvement of crystalline structure of GaN films grown on porous SiC substrates.

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