Investigation of microstructure defects in HTVPE grown polar GaN layers

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Gallium nitride (GaN) is a typical representative of the group III nitrides that is widely studied nowadays. Due to the lack of native substrates and high costs for their production, GaN is grown heteroepitaxially on foreign (Al_2O_3 or SiC) wafers. Recently, a successful growth of (0001)-oriented GaN layers by a modified high-temperature vapor phase epitaxy (HTVPE) method on GaN templates was reported [1], which were produced by metal organic chemical vapor phase epitaxy (MOVPE). In contrast to the conventional growth techniques, this inexpensive method employs ammonia (NH_3) and thermally evaporated metallic gallium (Ga) as precursors at atmospheric pressure. However, a significant lattice misfit between the substrate and the GaN template leads to the formation of microstructure defects like threading dislocations (TDs) and stacking faults (SFs).

In this work, we determined the TDs densities in HTVPE GaN layers grown at different ammonia flows on commercial MOVPE-grown GaN templates and compared them with the TDs in the templates. Applying controlled ammonia flow during the growth of particular samples we observed planar defects that can be partial SFs on the (0001)-planes, which are characterized by the absence of nitrogen planes (a or b) in the …AaBbAaBbAaBb… stack. The residual stress inherent in the heteroepitaxially grown GaN layers is ascertained for all samples under study and MOVPE-GaN templates. The possible reason for the residual stress is the elastic deformation originating both from the lattice misfit and from the interplay between the lattice misfit, differences in the thermal expansion of substrate and GaN and the formation of microstructure defects. The values of the residual stress correlate with the layers thickness and the total density of TDs [2].

A series of three samples (P1-P3) having the thickness of 4.5 μm was grown in a vertical HTVPE reactor [1]. A 2.5 μm thick MOVPE-GaN deposited on the 3-inch c-oriented sapphire substrate was cleaved into the 15x15 mm^2 pieces and used as a substrate for the HTVPE growth. An in-situ deposited SiN_x mask at the initial stages of the MOVPE growth was used to reduce the density of TDs [3]. The growth temperature of all samples was 1080 °C. The ammonia flow, which defines the [N]/[Ga] ratio (V/III ratio), was chosen as a sole variable parameter. In our experiments, the ammonia flow varied between 0.05 and 0.5 standard liter per minute (slm), see Table 1. Additionally, an uncoated piece of MOVPE-GaN template denoted as N1 was investigated.

High-resolution X-ray diffraction (HRXRD), micro-Raman spectroscopy and transmission electron microscopy in scanning mode (STEM) were used as the main experimental techniques in our study.

Table 1. The characteristics of the samples: NH_3 flow (C), density of screw TDs (ρ_s), density of edge TDs (ρ_e), total density of TDs (ρ_{total}), residual stress determined by XRD (σ_{XRD}), averaged over the sample thickness residual stress determined by micro-Raman spectroscopy ($\sigma_{\mu Raman}$)

Sample	C [slm]	ρ_s [10^8 cm^{-2}]	ρ_e [10^8 cm^{-2}]	ρ_{total} [10^8 cm^{-2}]	σ_{XRD} [MPa]	$\sigma_{\mu Raman}$ [MPa]
P1	0.5	2.4±0.4	10.3±1.6	12.7±2.0	-608±84	-478±50
P2	0.2	2.0±0.3	7.8±1.2	9.8±1.5	-611±110	-574±50
P3	0.05	2.2±0.3	8.2±1.2	10.4±1.5	-598±65	-585±50
N1	-	2.0±0.3	10.1±1.5	12.1±1.8	-643±83	-650±50

First, we applied the reciprocal-space mapping in the HRXRD mode in combination with the Monte Carlo approach in order to determine the densities of edge and screw TDs [4]. For this purpose, the symmetrical 004 and the asymmetrical 105 reciprocal-space maps (RSMs) were recorded for each sample. As dislocation bunching is often observed in thicker GaN layers grown by hydride phase epitaxy, a correlation in the dislocation positions must be introduced into the Monte Carlo approach [5]. The results of Monte Carlo simulation shown in Table 1 revealed that the TDs have predominantly edge character. The density of the edge TDs was 4-5 times higher than that the density of the screw TDs. A small decrease of the density of the edge TDs in the HTVPE layers with respect to the MOVPE template was detected for samples P2 and P3 with the low ammonia flow. The density of screw TDs was nearly constant for all samples including the GaN template N1.

In order to determine the residual stress, XRD with a modified $\sin^2\psi$ method and the micro-Raman spectroscopy were used [2]. The first method recognizes the elastic lattice strain as a shift of the diffraction maxima along the 2θ direction from their intrinsic positions, whereas the second one detects the strain as a shift of the E_2(high) Raman mode. The X-ray based $\sin^2\psi$ method determines the averaged strain in the irradiated volume defined by the beam size and the penetration depth. The micro-Raman technique enables to focus the laser beam at a desired depth and to determine the strain locally. The residual stress measurement using both complementary methods is necessary, especially if a depth gradient of the residual stress is expected.

The values of the residual stress are summarized in Table 1. In general, we obtained a good agreement between the XRD and micro-Raman data, with exception of sample P1, where the confocal micro-Raman technique revealed a strong depth gradient of the residual stress. The samples under study possess compressive residual stresses that are comparable with the residual stresses measured in the uncoated MOVPE template. The residual stress in GaN grown on sapphire stems from the lattice misfit and different thermal expansion coefficients of GaN and Al_2O_3. To some extent, the residual stress results from the uncompensated strain fields of bunched TDs, thus it can be strongly affected by the bunching of the TDs [2].

The STEM investigations confirmed the presence of dislocation bunching in the samples (see Figure 1). Here, one can see several areas located close to the interfaces (depicted as red rectangles), where several dislocations converge and form a bunched object. Still, the dislocation lines of the bunched dislocations are oriented nearly perpendicular to the sample surface, i.e., in the [0001] direction.

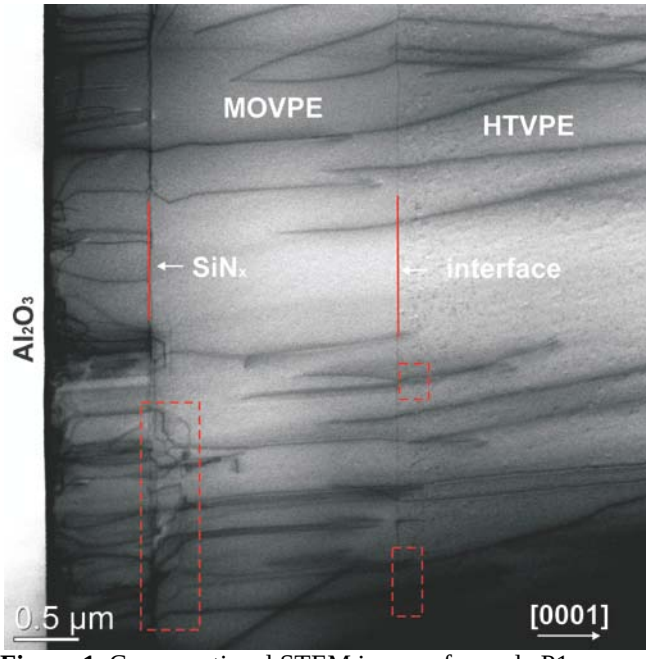

Figure 1. Cross-sectional STEM image of sample P1. The red rectangles refer to the areas where dislocation bunching takes place.

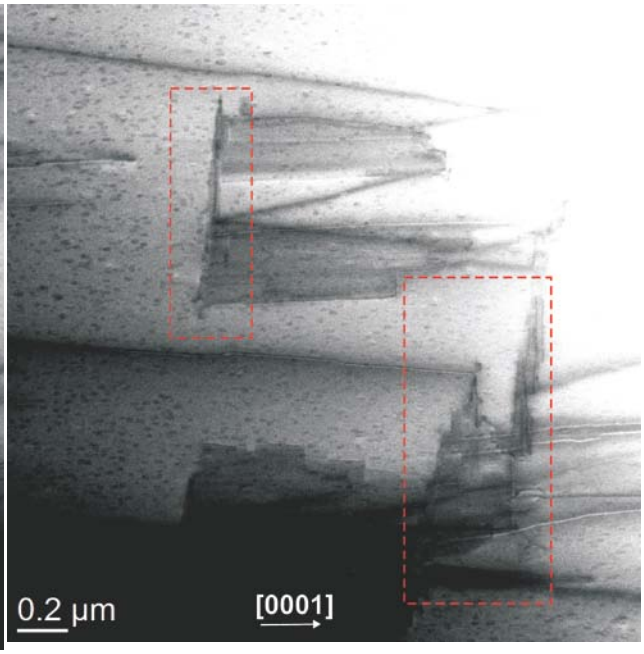

Figure 2. Cross-sectional STEM image of sample P3. The red rectangles mark the areas, which probably contain partial stacking faults terminated by partial dislocations.

In sample P3, which was grown under low V/III ratio, the detailed STEM investigations revealed several microstructure defects that deserve our attention (see Figure 2). They are aligned in the basal planes perpendicular to the surface and surrounded by partial dislocations. Unlike the common basal stacking faults (BSFs), these defects do not provide the broadening of the electron diffraction maxima and they do not disappear if the specimen is tilted so that the visibility condition of the BSFs is not fulfilled. If sample P3 is considered as a Ga-rich layer, than the revealed type of planar defects can refer to partial SFs characterized by the absence of nitrogen layers. The investigation of these defects using electron energy loss spectroscopy (EELS) can clarify their nature, whereas the combination of HRXRD with the DIFFaX software [6] can give their quantitative description.

Summarizing, a successful deposition of the low expensive HTVPE GaN layers on the thin MOVPE templates was revealed. The density of TDs in the HTVPE GaN is comparable or even less than the TDs density that can be achieved in the MOVPE-GaN buffer layer grown by applying the SiN_x nanomask. That makes our method promising for the growth of GaN layers with large thickness. The typical in GaN layers TDs bunching was confirmed by STEM. The compressive residual stress was determined using two techniques. Additionally, planar defects that can probably refer to partial stacking faults were observed in the sample grown under the lowest ammonia flow. The study of their interaction with other defects and the determination of their density will be next steps in our investigations.

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