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Correlation between the residual stress and the density of threading dislocations in GaN layers grown by hydride vapor phase epitaxy

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ABSTRACT

The correlation between the residual stress and the density of threading dislocations was investigated in polar GaN layers that were grown by using hydride vapor phase epitaxy (HVPE) on three different GaN templates. The first template type was GaN grown on sapphire by metal-organic vapor phase epitaxy. The second template type was a closed GaN nucleation layer grown on sapphire by HVPE. The third template type was a non-closed GaN nucleation layer grown by HVPE, which formed isolated pyramids on the sapphire surface. The residual stress was determined using the combination of micro-Raman spectroscopy and modified $\sin^2\psi$ method. The interplanar spacings needed for the $\sin^2\psi$ method were obtained from the reciprocal space maps that were measured using high-resolution X-ray diffraction. The density of threading dislocations was concluded from the broadening of the reciprocal lattice points that was measured using high-resolution X-ray diffraction as well. The fitting of the reciprocal space maps allowed the character of the threading dislocations to be described quantitatively in terms of the fractions of edge and screw dislocations. It was found that the threading dislocation density increases with increasing compressive residual stress. Furthermore, the dislocation density and the residual stress decrease with increasing thickness of the GaN layers. The edge component of the threading dislocations was dominant in all samples. Still, some differences in the character of the dislocations were observed for different templates.

1. Introduction

Gallium nitride is regarded as a promising semiconductor material for production of light emitting diodes, blue lasers, solar cells etc. [1]. Due to the lack of appropriate native substrates, GaN is typically grown on foreign wafers such as sapphire or silicon carbide. Sapphire is the most frequently used substrate for the GaN deposition. Still, the direct deposition of GaN films on the sapphire substrates is a challenging task because of the large lattice misfit between GaN and Al_2O_3. The (0001)-oriented GaN films usually grow on the (0001)-oriented sapphire with the mutual in-plane orientation $\langle 01\bar{1}0\rangle_{GaN} \| \langle \bar{1}100\rangle_{sapphire}$ [2]. In the corresponding directions, i.e. in $\langle 01\bar{1}0\rangle$ for GaN and in $\langle \bar{1}100\rangle$ for Al_2O_3, the distances between the metallic atoms (Ga and Al) match best, but they still differ of about 14.9%. The respective interatomic distances in GaN and Al_2O_3 are equal to the lattice parameter a of GaN (0.31895 nm) and to $a\sqrt{3}=0.27476$ nm for Al_2O_3. As the interatomic distances are larger in GaN than in Al_2O_3, compressive residual stress in GaN is expected.

At the growth temperature of about 1000 °C, the above lattice misfit is slightly reduced, because the thermal expansion coefficient (α_a) of sapphire is larger than the thermal expansion coefficient of GaN. Still, reduction of the lattice misfit is negligible (approximately 0.1%), as calculated for $\alpha_a(\mathrm{GaN})=7.9\times10^{-6}\,\mathrm{K}^{-1}$ and $\alpha_a(\mathrm{Al_2O_3})=9.2\times10^{-6}\,\mathrm{K}^{-1}$ [3]. Therefore, the large remaining lattice misfit promotes the nucleation and growth of individual and isolated islands instead of the growth of a closed film. Böttcher et al. correlated the average islands diameter of the islands with the residual stress in the layer [4].

The in situ wafer curvature measurements performed at the growth temperatures showed in GaN the presence of tensile stresses up to hundreds of MPa in many cases [5]. These tensile stresses were explained by the formation and recovery of cracks in the GaN films during the growth [6]. Due to the different thermal expansions of GaN and Al_2O_3, the low tensile residual stress in GaN turns to be compressive upon cooling from the deposition temperature to room temperature. For GaN layers having a thickness between 1 and 2 μm that were grown using metal-organic vapor phase epitaxy (MOVPE) at 1050 °C and cooled down to room temperature, Hearne et al. [5]

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and Böttcher et al. [4] reported the contribution of the thermal stress of about –(660 ± 100) MPa and –(710 ± 100) MPa, respectively. In thick GaN layers, the residual stress can be compensated by structure defects. A lot of studies reveal that the polar GaN heteroepitaxial layers grown by hydride vapor phase epitaxy (HVPE) possess large number of threading dislocations (TDs) [7,8].

In this study, we describe the interplay between the layer thickness, the density of threading dislocations and the residual stress in GaN layers, which were grown by HVPE on different GaN templates. The first template type was GaN produced by MOVPE on sapphire. The second and third template types consisted of closed and non-closed GaN nucleation layers, respectively, both deposited by HVPE directly on a sapphire substrate [2]. The residual stress was determined at room temperature by using high-resolution X-ray diffraction (HRXRD) and micro-Raman spectroscopy. The HRXRD recognizes the residual stress as a shift of the diffraction lines from their intrinsic positions. For the calculation of the residual stresses from the HRXRD data, a modified $\sin^2\psi$ method [9,10] in conjunction with the single-crystalline elastic constants from [11] was applied. The micro-Raman spectroscopy determines the residual stress from the shift of the peak positions of the observable Raman modes. The stress coefficient needed for recalculation of the peak shift into the residual stress [12] was calculated from the same elastic constants that were used for the modified $\sin^2\psi$ method [11]. The density of threading dislocations was obtained from the XRD line broadening that was measured via reciprocal space mapping using HRXRD. A Monte Carlo method was employed to simulate the intensity distribution in the reciprocal space [13]. The fitting of the simulated reciprocal space maps to the measured ones revealed the densities of screw and edge dislocations.

2. Experimental details

2.1. Sample preparation

The GaN layers were grown in a commercial vertical HVPE reactor from Aixtron [14] using the conventional HVPE process with elemental gallium, hydrogen chloride (HCl) and ammonia (NH_3) [15]. In most cases, a mixture of H_2 and N_2 was used as carrier gas, and the growth temperature was 1040 °C. Within each sample series, the HVPE GaN layers with different thicknesses were deposited on the same template, see Table 1. For most samples, the deposition was performed at the standard ratio of the partial pressures in the vapor phase known as V/III ratio [2]. Samples 1 and 4 were deposited a three-time lower V/III ratio.

Samples S1–S3 were grown on 2-inch (0001)-oriented MOVPE GaN templates. The second sample series comprising samples S4–S6 was deposited on closed HVPE GaN templates. These templates were grown directly on (0001)-oriented, 2-inch sapphire substrates by using a special two-step HVPE technology, which was described in more details in Ref. [2]. In the first step, a closed nucleation layer with a thickness up to 2 μm was deposited at 780 °C. In the second deposition step performed at 1040 °C, this nucleation layer was overgrown by the actual HVPE GaN template layer. The last sample (S7) was deposited at similar deposition conditions like S5 and S6, but the non-closed HVPE GaN nucleation layer grown at 780 °C was employed directly as template for the further growth. Consequently, the surface of sample S7 is relatively rough, having a typical thickness variation of 1–2 μm with a lot of small pits.

2.2. Sample characterization

The high-resolution X-ray diffraction (HRXRD) measurements were carried out at a triple-axis diffractometer (Seifert/FPM) with an Eulerian cradle, which was equipped with a sealed X-ray tube with copper anode and two perfect, i.e. dislocation-free, (111)-oriented Si crystals. The first Si crystal was used as a monochromator in the primary beam, the second one as an analyzer of the diffracted beam. The cross section of the primary X-ray beam was reduced by a set of slits to 0.09 × 2 mm² in order to reduce the effect of the sample bowing on the measured line broadening. The instrumental line broadening of the diffractometer was below 10″ as estimated using a (111)-oriented Si single-crystal. The penetration depth of X-rays in GaN was estimated as 5 μm for $\lambda = 0.154056$ nm.

The density of threading dislocations was determined from the reciprocal space maps (RSMs) that were recorded in coplanar diffraction geometry on the symmetrical reflection 0004 and on the asymmetrical reflections $10\bar{1}4$ and $10\bar{1}5$. During the reciprocal space mapping, a set of radial ($2\theta/\omega$) scans was measured for different values of ω. The quantity 2θ denotes the detector angle, ω the angle between the primary beam and the sample surface. As a result of the reciprocal space mapping, a distribution of the measured intensity in the angular (ω, 2θ) space was obtained for each diffraction line that was converted into the (q_x, q_z) representation of the reciprocal space using the transformation

$$q_x = \frac{2\pi}{\lambda}[\cos(2\theta-\omega) - \cos\omega]; \quad q_z = \frac{2\pi}{\lambda}[\sin(2\theta-\omega) + \sin\omega] \qquad (1)$$

In Eq. (1), $\lambda = 0.154056$ nm is the wavelength of the X-ray beam. In addition to RSMs, radial ($2\theta/\omega$) and azimuthal (ω) scans were performed through the intensity maxima of the symmetrical diffractions 0002, 0004 and 0006 and the asymmetrical diffractions $10\bar{1}4$, $10\bar{1}5$ and $11\bar{2}4$ or $20\bar{2}4$ for each sample. These radial and azimuthal scans were used to obtain exact line positions (in the q_x and q_z coordinates, see Eq. (1)) and interplanar spacings ($d_\psi = 2\pi/\sqrt{q_x^2+q_z^2}$) that are needed for the residual stress calculation.

The lateral distribution of threading dislocations at the sample surface was visualized via etch pit technique. The samples were

Table 1
Characteristics of the samples under study: template type, average sample thickness (t), screw TDs density (ρ_s), edge TDs density (ρ_e), total TDs density (ρ_{tot}), residual stress determined from XRD (σ_{XRD}), residual stress determined from Raman spectroscopy (σ_{Raman}). The densities of screw and edge threading dislocations from Monte Carlo simulation were determined with an error of ± 15%. The values of residual stress from X-ray and micro-Raman experiments possess an error of approx. ± 50 MPa. The asterisks mark the samples, which were deposited at three-time lower V/III ratio than the standard deposition conditions are.

Sample	Template	t [μm]	ρ_s [10^8 cm^{-2}]	ρ_e [10^8 cm^{-2}]	ρ_{tot} [10^8 cm^{-2}]	σ_{XRD} [MPa]	σ_{Raman} [MPa]
S1*	MOVPE GaN	900	0.13	0.38	0.5	−55.1	−44.4
S2		50	0.28	1.04	1.3	−190.6	−274.1
S3		5	0.77	1.9	2.7	−566.4	−496.3
S4*	Closed HVPE	13	0.68	3.3	4	−231.2	−203.7
S5	GaN template	9	0.9	8.9	9.8	−496.6	−444.4
S6		15	0.86	5.4	6.3	−366.5	−348.1
S7	Non-closed HVPE GaN template	8	6.7	17.6	24.3	−658.9	−507.5