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# High-temperature X-ray characterization of GaN epitaxially grown on Sc<sub>2</sub>O<sub>3</sub>/Y<sub>2</sub>O<sub>3</sub>/Si(111) heterostructures

Short title: High-temperature characterization of GaN

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**Abstract.** The thermal behavior of GaN epitaxially grown on a Sc<sub>2</sub>O<sub>3</sub>/Y<sub>2</sub>O<sub>3</sub> bilayer buffer on Si(111) substrate is studied between room temperature (RT) and 800°C by X-ray diffraction of symmetrical and asymmetrical reflections. It is found that the GaN layer is in-plane mediated by the oxide buffer completely fixed to the Si substrate, which indicates the absence of any temperature related compliance behavior of the oxide buffer. The GaN grows at 720°C already slightly tensile strained on top of the larger Sc<sub>2</sub>O<sub>3</sub> lattice, and this strain increases further during cooling down to RT due to different coefficients of thermal expansion of GaN and Si substrate.

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## 1. Introduction

The integration of GaN on large-size silicon (Si) wafers as substrate could offer new possibilities in the combination of micro and optoelectronics with the advantage of significantly lower costs compared to commonly used relatively small and expensive SiC and sapphire substrates. The great challenge is to develop a manufacturing process for crack free, low dislocation density GaN films. One approach to realize this is based on heteroepitaxial growth of thin GaN films on Si [1]. Due to a large lattice mismatch ( $\sim -17\%$ ) between GaN and Si, differences in the coefficients of thermal expansion (CTE) and the high reactivity of Si to impinging Ga, it is very difficult to grow GaN directly on Si. This led to the concept of using different semiconducting buffer layers as for example AlN [2,3].

Another idea is to use heteroepitaxially grown oxides as buffer [4]. Our group introduced the  $\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3$  bilayer as buffer and diffusion barrier that reduces the misfit between GaN and the top oxide layer ( $\text{Sc}_2\text{O}_3$ ) to about  $-8\%$  [5]. Since especially the cracking behavior of GaN layers on different substrates is strongly related to the thermal properties of all involved materials, it is very important to study their thermal interaction directly on the layer system. The thermal behavior of  $\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3/\text{Si}(111)$  heterostructures was investigated in [6]. Here we want to study the strain state of GaN on such a bilayer oxide buffer as a function of temperature.

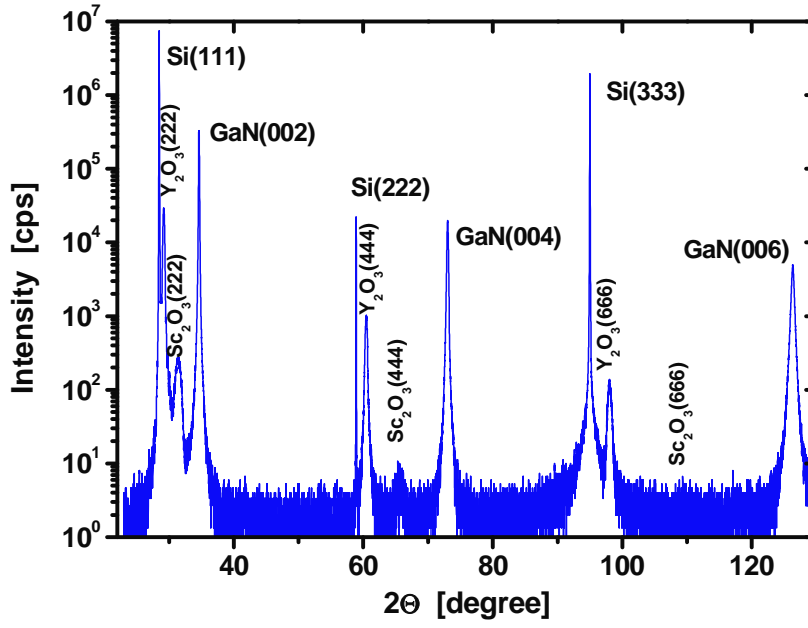
## 2. Experimental

The high temperature behavior will be demonstrated for a typical sample with a 600 nm thick GaN layer epitaxially grown on a 4-inch Si (111) wafer with oxide buffer consisting of about 30 nm  $\text{Y}_2\text{O}_3$  and 30 nm  $\text{Sc}_2\text{O}_3$ . GaN/ $\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3/\text{Si}(111)$  heterostructures were prepared in a multi-chamber molecular beam epitaxy (MBE) system following a recipe previously described in detail [5]. The deposition temperatures for  $\text{Y}_2\text{O}_3$  and  $\text{Sc}_2\text{O}_3$  were  $625^\circ\text{C}$  and  $500^\circ\text{C}$ , respectively, GaN of wurtzite structure was deposited at  $720^\circ\text{C}$ .

All X-ray measurements were performed with a Rigaku SmartLab diffractometer in a high-resolution setup with Ge(400)x2 crystal collimator and Ge(220)x2 crystal analyzer using  $\text{CuK}_{\alpha 1}$  radiation ( $\lambda = 1.54059 \text{ \AA}$ ). For high temperature experiments, a DHS1100 oven from Anton Paar was mounted on the diffractometer. The sample was kept in  $\text{N}_2$  ambient while heated.

## 3. Results and discussion

The thickness of the GaN layer of 600 nm is certainly too low for power device applications, but it was deliberately chosen. On one side the GaN layer should be much thicker than the buffer layers to represent possible applications, but on the other side it should not reach a thickness where cracking occurs. Figure 1 shows a specular  $\omega$ - $2\Theta$  scan of the GaN/ $\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3/\text{Si}(111)$  heterostructure that demonstrates the (111) orientation of both oxides and the (001) orientation of the GaN layer parallel to the (111) Si substrate normal. The in-plane orientation of the crystal lattices was analyzed in detail in [5] and is characterized by the relationship:  $\text{GaN}[100] \parallel \text{Sc}_2\text{O}_3[\bar{1}2\bar{1}] \parallel \text{Y}_2\text{O}_3[\bar{1}2\bar{1}] \parallel \text{Si}[\bar{1}\bar{2}1]$ .



**Figure 1.** Specular  $\omega$ - $2\theta$  scan of the investigated GaN/Sc<sub>2</sub>O<sub>3</sub>/Y<sub>2</sub>O<sub>3</sub>/Si(111) heterostructure

The two oxide layers in the buffer are nearly fully relaxed with lattice constants of 10.61 Å for Y<sub>2</sub>O<sub>3</sub> and 9.85 Å for Sc<sub>2</sub>O<sub>3</sub> as discussed in detail in [6].

The plane defined by the Si lattice directions [111] and  $[\bar{1}\bar{2}1]$  as well as by the GaN lattice directions [001] and [100] was used for the analysis of off-plane and in-plane lattice constants by measuring symmetrical and asymmetrical Bragg reflections. Figure 2 shows the superposition of the reciprocal lattices in these planes, where the used reflections (333) and (404) for Si and (002) and (104) for GaN are marked.  $\omega$ - $2\theta$  and  $\omega$  scans of these four reflections were measured at room temperature (23°C), from 100°C up to 800°C in steps of 100°C, and after cooling down to RT again. The measurements last about 40 minutes at each temperature step. Exact peak positions were adjusted by curve fitting and then translated into reciprocal space coordinates according to

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

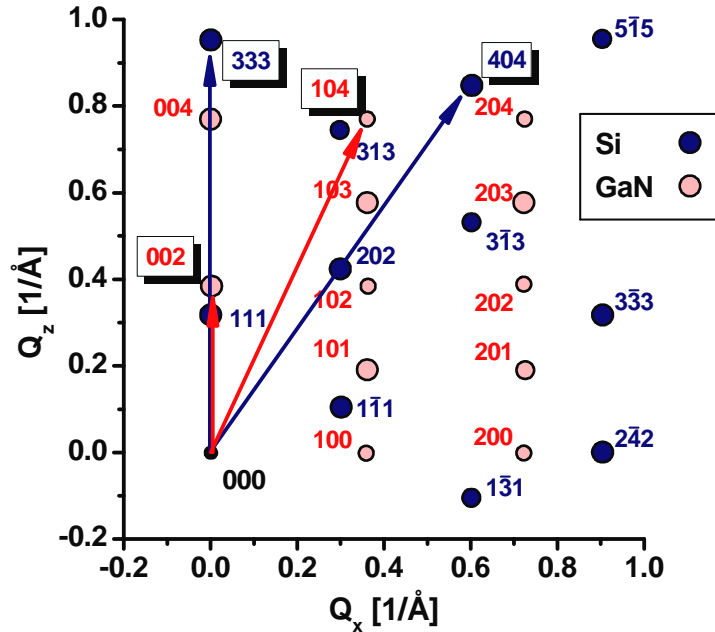
The off-plane ( $a_l$ ) and in-plane ( $a_0$ ) lattice constants of Si can be obtained by:

$$a_l = \sqrt{3} \cdot d_{(111)} = \frac{3 \cdot \sqrt{3}}{q_z^{(333)}} \quad , \quad a_1 = \sqrt{3} \cdot d_{(111)} = \frac{8 \cdot \sqrt{3}}{3 \cdot q_z^{(404)}} \quad , \quad a_0 = \sqrt{6} \cdot d_{(112)} = \frac{4 \cdot \sqrt{6}}{3 \cdot q_x^{(404)}} \quad (2)$$

and the lattice constants  $c$  and  $a$  of the hexagonal GaN by:

$$c = 2 \cdot d_{(002)} = \frac{2}{q_z^{(002)}} \quad , \quad c = 4 \cdot d_{(004)} = \frac{4}{q_z^{(004)}} \quad , \quad a = \sqrt{\frac{4}{3}} \cdot d_{(100)} = \sqrt{\frac{4}{3}} \cdot \frac{1}{q_x^{(104)}} \quad (3)$$

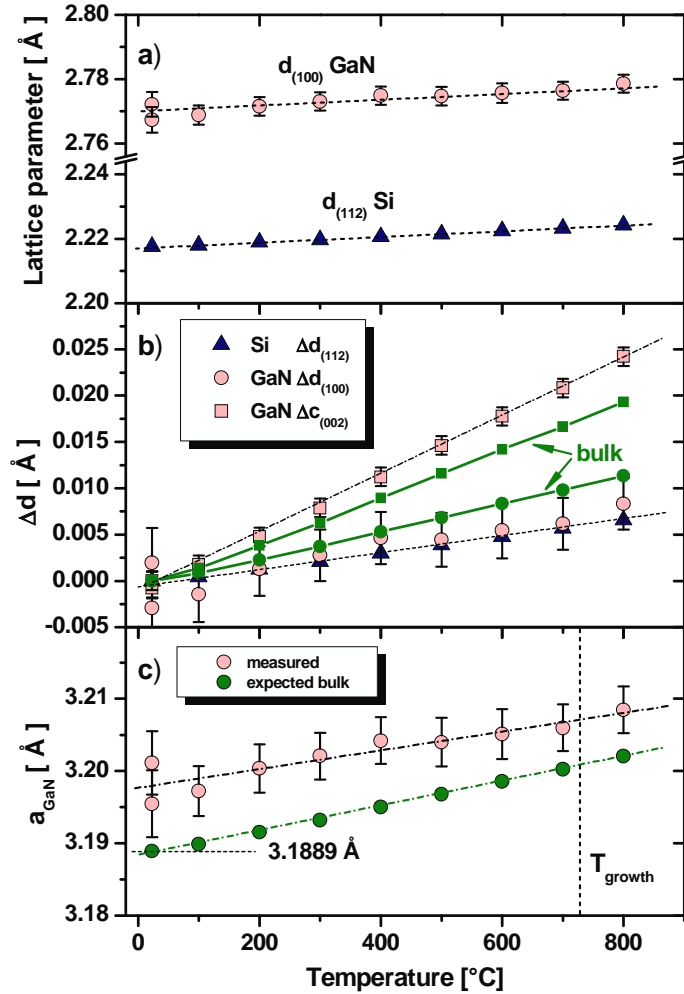
It is necessary to note that this method is not suited for a precise absolute determination of lattice constants, but it is acceptable for a relative comparison of data obtained at different temperatures. Nevertheless, the three  $a$  values obtained for Si agree within a maximum deviation of 0.0008 Å for all temperatures, which verifies the ability of this method and the unstrained state of the Si substrate.



**Figure 2.** Sketch of the Si and GaN reciprocal lattice in the plane of diffraction (hkl notation is also used for the hexagonal GaN lattice). The reflections used for the determination of off- and in-plane lattice constants are marked.

Most interesting is the behavior of the in-plane lattice parameter  $d_{(100)}$  of GaN compared to  $d_{(112)}$  of the Si substrate as demonstrated in figure 3a. The slope of the linear regression lines of both lattice parameters is almost equal. The absolute change of  $d_{(112)}$  of Si and  $d_{(100)}$  of GaN relative to the values at RT shown in figure 3b confirms that both parameters increase with increasing temperature by exactly the same amount. This indicates that the GaN layer on top of the two-layer oxide buffer is in-plane completely fixed to the Si substrate and follows the substrates thermal expansion despite of the fact that the coefficient of thermal expansion (CTE) for  $a$  of GaN [7] is significantly higher than the CTE of Si. This includes the fact that both oxide layers between Si and GaN are in-plane also fixed to the substrate as concluded in [6] for heteroepitaxial  $\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3/\text{Si}(111)$  structures, even this could not be directly measured by the used method due to a too low signal of the significantly thinner layers. There is no thermal compliance behavior of the oxide buffer detectable at least up to 800°C (slightly above the growth temperature of GaN). The  $a$  values of GaN at RT before and after high temperature treat-

ment differ slightly. But they still agree within the error limits, and since the corresponding  $c$  values do not show a similar behavior this can be simply explained by measuring inaccuracy. Additional,



**Figure 3.** a) Measured in-plane lattice parameter  $d_{(112)}$  of Si and  $d_{(100)}$  of GaN vs. temperature. b) Absolute change of lattice parameters  $d_{(112)}$  of Si and  $d_{(100)}$  and  $c$  of GaN relative to the values at room temperature. The curves marked with ‘bulk’ show the expected parameter change under bulk conditions. c) Comparison between the measured GaN lattice parameter  $a$  and the expected change of  $a$  starting from the bulk lattice constant at room temperature using estimated CTE values.

figure 3b shows that the difference of the lattice parameter  $c$  of GaN related to the value at RT of  $5.1810 \text{ \AA}$  increases much faster than that for  $a$ .

The estimated lattice parameters of the GaN layer for room temperature differ slightly from table values [8] (in brackets):  $c = 5.1810 \pm 0.002 \text{ \AA}$  ( $5.1855 \text{ \AA}$ ),  $a = 3.1979 \pm 0.003 \text{ \AA}$  ( $3.1889 \text{ \AA}$ ). The fact that the experimental  $c$  value is smaller and  $a$  larger than the table values indicates a tensile strain in the GaN layer. To decide whether this is the result of thermal mismatch only or it is caused by the growth

process, it is necessary to estimate the CTE values for bulk GaN from the performed measurement. The relation between strained ( $c_{str}$ ,  $a_{str}$ ) and bulk lattice parameters ( $c$ ,  $a$ ) of GaN is given by:

$$\frac{(c_{str} - c)}{c} = \frac{-2\nu}{(1-\nu)} \cdot \frac{(a_{str} - a)}{a} \quad (4)$$

with  $\nu = 0.212$  [8]. The temperature dependence of  $a$  and  $c$  is:

$$a = a_{T_0} (1 + \alpha^a \cdot \Delta T) \quad \text{and} \quad c = c_{T_0} (1 + \alpha^c \cdot \Delta T) \quad (5)$$

where  $\alpha^a$  and  $\alpha^c$  are the CTE values for both lattice parameters. The absolute change of the in-plane lattice constant  $a$  follows the Si lattice constant

$$\Delta a = (a_{str} - a) = a^{Si} \cdot \alpha^{Si} \cdot \Delta T \quad (6)$$

With the measured data for  $c_{str}$  and  $a_{str}$  and using equation (4) it is possible to estimate both CTE values for bulk GaN under the assumption that according to [7] the difference of CTE values  $\alpha^a - \alpha^c \approx 0.5 \cdot 10^{-6} \text{ K}^{-1}$  and the Poisson ratio  $\nu$  are constant independent of the temperature. Results obtained show the expected increase of  $\alpha^a$  and  $\alpha^c$  with increasing temperature and are with e.g.  $\alpha^a = (5.6 \pm 0.8) \cdot 10^{-6} \text{ K}^{-1}$  at 400°C in good agreement with data given in [7]. Although it is not the intention of this paper to measure the CTE values of GaN with high precision, since the used method is not suited for this, the roughly estimated CTE values can be used to calculate the change of lattice parameters under unstrained (bulk) conditions as shown in figure 3b. The comparison with the measured strained parameters shows qualitatively the expected situation that  $a$  is reduced in its expansion and  $c$  is additionally increased by in-plane compressive stress due to the fixed connection to the Si substrate.

Figure 3c shows that the in-plane lattice constant  $a$  of the GaN layer is larger than the expected value for unstrained GaN even at growth temperature. This indicates that the GaN does not grow fully relaxed on the oxide buffer. It grows already tensile strained on top of the larger  $\text{Sc}_2\text{O}_3$  lattice (in-plane lattice parameter  $a >$  bulk value), and this strain increases further during cooling down to room temperature due to different CTE values (difference between both values increases with decreasing temperature).

#### 4. Conclusion

Measurements of the in- and off-plane lattice parameters of GaN and Si in a heteroepitaxial GaN/ $\text{Sc}_2\text{O}_3$ / $\text{Y}_2\text{O}_3$ /Si(111) layer stack have demonstrated that the absolute change of in-plane lattice parameters of all layers is exactly the same as that of the Si substrate. Since the absolute CTE values

of GaN is larger than that of Si, this leads additionally to a growth-induced tensile strain in the GaN layer to a further increase of tensile strain during cooling down from growth to room temperature. Thus it becomes evident that the  $\text{Sc}_2\text{O}_3/\text{Y}_2\text{O}_3$  bilayer oxide buffer does not adapt the thermal behavior of GaN and Si substrate. Its advantages are more of structural kind to reduce the lattice mismatch, to act as a diffusion barrier, and to improve the surface wetting for GaN layer growth.

This finding leads to the more general question, whether an epitaxially grown crystalline buffer can have a compliant effect between GaN epi-layer and substrate at all. The critical situation for cracking is the cooling process from growth temperature down to room temperature. A suited crystalline buffer material should be able to compensate the CTE-related increasing lattice constant difference between GaN and Si substrate in the cooling process by adopting its lattice parameters to that of GaN. This requires a continuous plastic relaxation process over a wide temperature range, which seems to be quite unrealistic. Another possible approach could be the use of amorphous buffer structures to realize a ‘floating’ of the epi-layer on the substrate. First attempts were demonstrated using silicon semiconductor-on-insulator substrates [9-11]. But also rear earth oxides originally grown epitaxially as crystalline buffer layer on Si can be transformed by suited processing into amorphous silicate layers [6]. This will be the subject of our further investigations.

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