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Epitaxy of GaN on silicon—impact of symmetry and surface reconstruction

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Abstract. GaN-on-silicon is a low-cost alternative to growth on sapphire or SiC. Today epitaxial growth is usually performed on Si(111), which has a threefold symmetry. The growth of single crystalline GaN on Si(001), the material of the complementary metal oxide semiconductor (CMOS) industry, is more difficult due to the fourfold symmetry of this Si surface leading to two differently aligned domains. We show that breaking the symmetry to achieve single crystalline growth can be performed, e.g. by off-oriented substrates to achieve single crystalline device quality GaN layers. Furthermore, an exotic Si orientation for GaN growth is Si(110), which we show is even better suited as compared to Si(111) for the growth of high quality GaN-on-silicon with a nearly threefold reduction in the full width at half maximum (FWHM) of the $(1\bar{1}00)$ ω-scan. It is found that a twofold surface symmetry is in principal suitable for the growth of single crystalline GaN on Si.

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1. Introduction

Since the first demonstration of metal organic vapor phase epitaxy (MOVPE) grown crack-free GaN layers in excess of 1 μm [1] GaN epitaxy on silicon has developed within a few years to a competitive low-cost alternative to growth on sapphire and SiC. With its good thermal conductivity it is especially interesting for electronic applications [2] but also for low-cost high brightness light emitting diodes (LEDs) if substrate removal is performed [3]. The typical Si surface on which GaN growth is performed is the Si(111) surface with a threefold symmetry and a sixfold atomic arrangement at the surface. On the other hand, Si(001) is the important material in the complementary metal oxide semiconductor (CMOS) industry, thus for integrating GaN electronics and optoelectronics with silicon this is the preferred surface. It has been already demonstrated that indeed single crystalline GaN can be grown on this surface with its fourfold symmetry after a few publications in the 1990s demonstrated *c*-axis oriented layers but extremely rough surfaces [4]–[9].

Here, we show how surface modifications of the Si(001) surface can be used to obtain smooth, single crystalline GaN and that silicon surfaces with twofold symmetry such as (110) can be used for the growth of high quality GaN.

2. Experimental

Nitride growth is performed in a horizontal AIXTRON AIX 200/4 RF-S or a vertical TSSEL $6 \times 2''$ showerhead reactor. All substrates are two inch Si substrates with surface orientations of (111), (001) 0°–4° off oriented towards (110), (110), (115), (117) and (119). Standard precursors such as NH_3, TMAl and TMGa in hydrogen atmosphere were used and growth temperatures ranged from ∼630 °C for low-temperature (LT) AlN seeding layers to 1050 °C for high-temperature (HT) AlN seeding and GaN buffer layers. Prior to growth, all substrates were cleaned with iso-propanol and etched using $H_2SO_4 : H_2O_2 : H_2O$ (3:1:1) followed by an HF (5%) etching step and rinsing in deionized water between these steps as described in [10]. GaN growth on silicon always starts with an AlN seeding layer grown at low (∼650 °C) or high (∼1050 °C) temperatures followed by GaN sometimes with additional strain engineering LT AlN layers [1]. The samples were characterized by x-ray diffraction (XRD) and transmission electron microscopy (TEM).

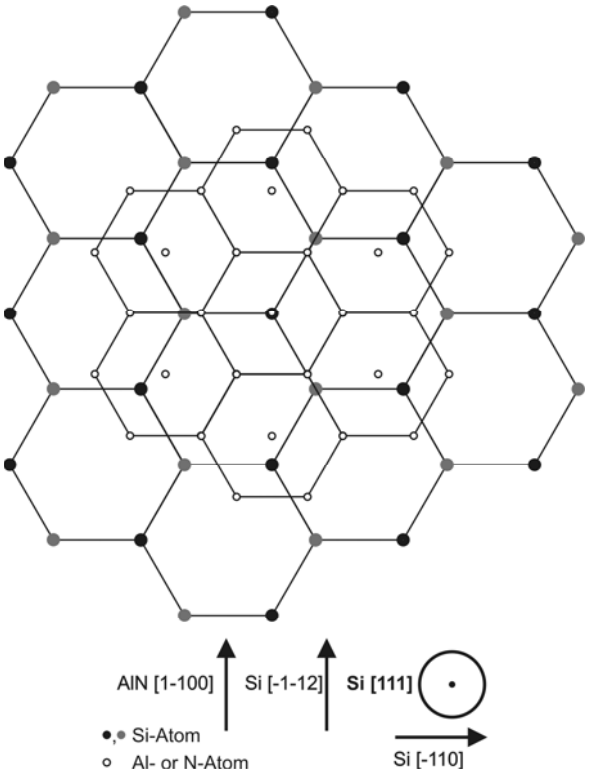

Figure 1. Atomic arrangement of AlN on Si(111).

3. Si(111)

The growth on Si(111) was always favored for the wurtzite nitrides due to the sixfold atomic arrangement (threefold symmetry) at the surface, which already gives a good rotational matching for AlN, the commonly applied seeding layer on Si. This matching is most likely also present during seed layer growth because the (1×1) surface reconstruction is the common reconstruction type in the presence of hydrogen [11], the commonly used carrier gas during AlN seed layer growth. The layer grows with an in-plane orientation of AlN $\langle 10\bar{1}0 \rangle \parallel \mathrm{Si}\langle \bar{1}12 \rangle$ and $\mathrm{AlN}\langle \bar{1}\bar{1}20 \rangle \parallel \mathrm{Si}\langle \bar{1}10 \rangle$ (figure 1). For this alignment lattice mismatch is 16.9% to GaN and 19% to AlN. The XRD tilt and twist ω-scan values for the (0002) and $(10\bar{1}0)$ reflections are typically around 500–800 and >800 arcsec, respectively [12]. For thick layers presently a best value for the (0002) ω-scan, reported for 5.4 μm thick LED structures, is 360 arcsec [2].

In regard of dislocations, it is still often argued that threading dislocations originate from lattice mismatch in a heteroepitaxial system. But this is not fully true for nitrides on Si: the high mismatch leads to an extremely high number of misfit dislocations at the interface ($\sim 10^{13}$ cm^{-2}) and only about every second Al–N molecule is bound to Si as already shown by Liu *et al* [13]. These misfit dislocations are in-plane dislocations and do not propagate vertically. But the poor matching of the atoms gives rise to twist and tilt since the low number of bonded atoms creates more possibilities for the nitride layer to be attached to the Si surface, especially when surface inhomogeneities such as steps are taken into account. Slightly twisted or tilted arrangements can then be more favorable than the idealized idea of growth as shown in figure 1 or by Liu *et al* [13]. Twist and tilt of the crystallites are the origin of most threading dislocations which

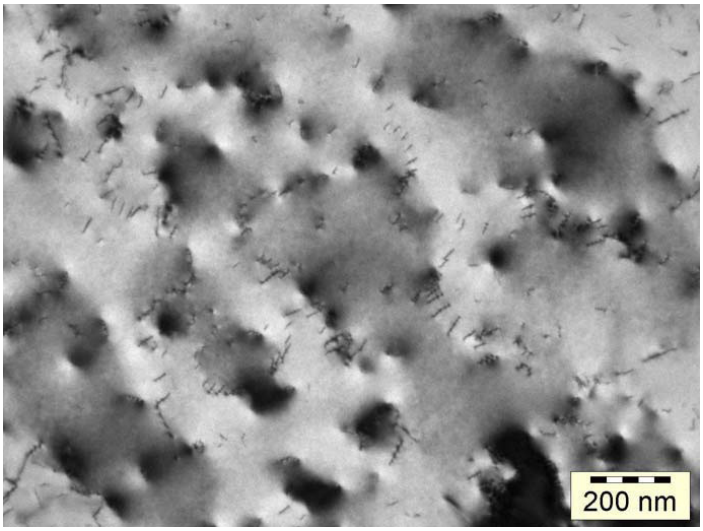

Figure 2. Slightly tilted plan view TEM of GaN on Si showing lines of dislocations at the boundaries between crystallites.

do originate at the boundaries of the crystallites [14]. In figure 2, a slightly tilted plan-view TEM image illustrates such dislocations which are aligned along a curved line which is the boundary between misaligned crystallites. The dark spots show dislocation clusters or strain induced bending of the thinned film. One method to reduce dislocations is to grow large diameter crystallites which can be performed, e.g. by *in situ* or *ex situ* SiN masking of the AlN surface and growing GaN on top of it [15] or by epitaxial lateral overgrowth (ELOG) [16] or pendeo-epitaxy [17]. Although the material quality has not reached the quality of layers on sapphire or SiC, electronic devices are already commercially available and LEDs based on GaN-on-Si are in the focus of industry.

4. Si(001)

Si(001) is favored for GaN growth due to several reasons: it is the material of the Si industry and wanted for a possible integration of GaN devices with Si electronics. It is easier to etch by wet chemical etching, and easier to cleave. Publications from the 1990s show that GaN growth on Si(001) leads to polycrystalline structures or at least very rough surfaces consisting of many grains [4]–[9]. The reason for this is the fourfold symmetry and the possibility for GaN with its sixfold symmetry to grow with two preferred rotational alignments on this surface when HT AlN seed layers are applied (figure 3). For such *c*-axis oriented material the in-plane alignment is AlN $\lfloor 10\bar{1}10 \rfloor \parallel \mathrm{Si}\lfloor \bar{1}00 \rfloor$ or $\mathrm{AlN}\lfloor 10\bar{1}0 \rfloor \parallel \mathrm{Si}\lfloor 0\bar{1}0 \rfloor$. The lattice mismatch is anisotropic and yields 15% for AlN $\langle 11\bar{2}0 \rangle \parallel \mathrm{Si}\langle \bar{1}00 \rangle$ and 0.7% for AlN $\langle 10\bar{1}0 \rangle \parallel \mathrm{Si}\langle \bar{1}00 \rangle$. While these mismatch values are lower than for Si(111) the material quality is inferior. We assume that both the possibility to grow single crystalline GaN, as well as the inferior quality compared to Si(111) substrates originates in the surface reconstruction of Si.

Although it is difficult to determine the surface reconstruction under MOVPE growth conditions calculations indicate that in a hydrogen atmosphere, even at high temperatures a (1×2) reconstruction is present on Si(001) surfaces [18]. The experiments support these

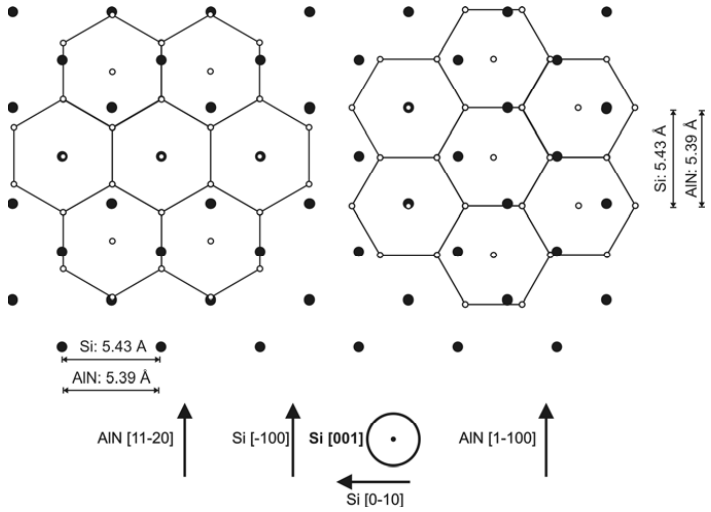

Figure 3. Atomic arrangement of AlN on Si(100).

calculations for low temperatures ($<350\,^{\circ}$C) [19]. On the other hand, in vacuum aluminium, present when AlN growth starts, can cause a roughening of the (001) surface at low temperatures ($<700\,^{\circ}$C) creating {310} facets and reversing to a (2×1) reconstruction when heating to $1000\,^{\circ}$C [20]. This is even below the seed layer temperatures used in our experiments for the growth on Si(001).

Since we obtain single crystalline GaN, we can expect that at the beginning of HT AlN seed layer growth ($>1000\,^{\circ}$C) the real Si(001) surface has a surface reconstruction which is reducing the fourfold symmetry of the crystal surface to a twofold symmetry, at least on a smaller scale. If we assume that at high temperatures the Si(001) surface rearranges in a (2×1) and (1×2) reconstruction, the reconstruction appearing depends on the Si surface layer. In general, the Si dimer chains are either aligned along the [110] or the $[1\bar{1}0]$ directions separated by one monolayer in height. Most reconstructions on the Si surface are aligned along these directions thus having two perpendicular directions separated by a monolayer in height. Therefore, if monatomic steps are present two reconstructions will appear with the reconstruction on the terraces between these monatomic steps showing only one type of reconstruction. One of these surfaces either with the (1×2) or the perpendicular (2×1) reconstruction can be used for single crystalline GaN growth if the other is suppressed. Suppression of one reconstruction type can be achieved by different means:

- forming biatomic steps using off oriented substrates [21],
- tensile or compressive strain [19].

While the latter method is difficult to apply, the first can be easily used for device quality layers. But even for misoriented substrates two differently aligned regions will most likely exist under MOVPE growth conditions. One reason is the high temperature resulting in disorder causing a low stability of only one reconstruction type on a large scale, as well as deviations from a perfect reconstruction [22]. Therefore, typical seeding layers do not consist of one orientation type but of two. The selection of one is determined by the one with a higher percentage as soon as GaN growth begins on the Al(Ga)N seeding/buffer layer as shown for a layer on a 4° off-oriented Si(001) substrate in figure 4. The growth of approximately 1 μm of GaN on an Al(Ga)N buffer layer on an exactly oriented Si(001) substrate gives rise to an island

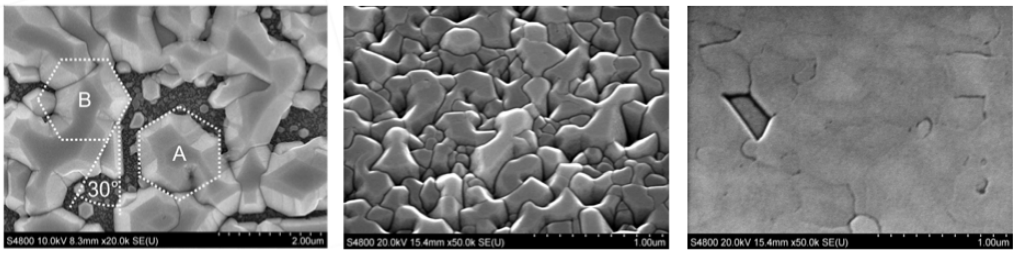

Figure 4. Left: GaN grown on an AlGaN/AlN buffer on Si(001). Two differently aligned types of crystallites (marked A and B) are observed. At the boundaries of such crystallites no coalescence is possible. Center: AlGaN on AlN on Si(001) 4° off. At this stage of growth no dominant crystallite alignment can be observed. Right: after 100 nm of GaN growth selection of one crystallite type has occurred suppressing the other. The dominant crystallite type usually grows faster and will overgrow remaining misoriented crystallites within a few hundred nanometres.

morphology with 90° twisted domains. The surface remains rough due to the impossibility of island coalescence for the two perpendicular types of domains (figure 4 left). Using a 4° misoriented Si(001) substrate the seeding layer and the subsequent AlGaN layer still exhibit the island type of growth with misoriented domains (figure 4 center), however, on a smaller scale. As soon as 100 nm of GaN are grown, a rather smooth surface has formed with only few remaining misoriented crystallites (figure 4 right) which disappear if growth is continued. Detailed XRD investigations have shown that indeed one orientation type dominates and the two different orientations in the AlGaN layer are differently tilted which seems to be sufficient to obtain one dominant orientation when GaN growth starts.

In contrast to the expectation that misoriented substrates show only one dominant reconstruction, which should result in only one orientation of the seeding and buffer layer (figure 4 center), the two orientations observed indicate the presence of two different reconstruction orientations on Si(001). As already mentioned, we attribute this to a thermally induced disorder of the surface reconstruction. But even for the subsequent GaN layer where only one orientation of the crystallites dominates and smooth surfaces are obtained the alignment of the crystallites is not good, especially twist is larger than for GaN Si(111), for best layers a factor of 2–3. This can be easily seen in cross-sectional TEM images (figure 5): the brighter and darker contrasts originate in a misalignment of the crystallites usually not observable for GaN on Si(111). It is found that within these columns no or only rarely dislocations are visible, as can be expected if threading dislocations in the nitrides do originate at the coalescence boundaries. For $\sim$2.5 μm thick GaN on Si(001) layers best twist and tilt values are around 720 arcsec for the (0002) and $\sim$0.8° for the (10$\bar{1}$0) reflections. They are worse, especially for the twist value, than for the growth on Si(111). However, we will show in the next section that indeed lattice mismatch plays an important role in regard of low twist and tilt.

The preference of one reconstruction type on the Si surface can also vary across the wafer. When the wafer is heated to high temperatures for growing the AlN seed layer the wafer shows a temperature gradient [23] and is typically concavely curved. The reason is that in MOVPE typically cold-wall reactors are used. If the curvature is strong enough regions with tensile and

Figure 5. Cross-sectional TEM of an LED structure on Si(001). The brighter and darker areas or domains in vertical direction originate in stress or a misorientation of the crystallites. Within these regions the dislocation density is very low.

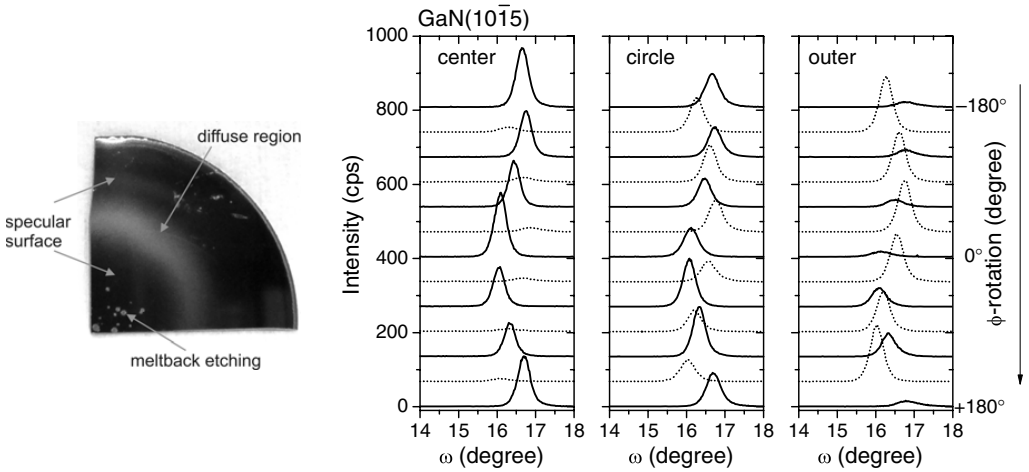

Figure 6. Picture of a sample with two differently aligned regions and a transition region visible from the diffuse appearance (left). XRD rocking scans of the (10$\bar{1}$5) reflection at different rotational angles Φ reveal the two orthogonal GaN orientations (full line and dotted line) which switch from center to the edge and are about equally distributed in the center.

compressive stress exist on the surface. Due to nonlinear effects the strain is anisotropic with different radial and azimuthal components [24]. This can change the preferred reconstruction from, e.g. (2×1) to (1×2) [19] and with it the preferred rotational alignment of the AlN crystallites of the seeding layer. We believe that we have observed this effect for multiple

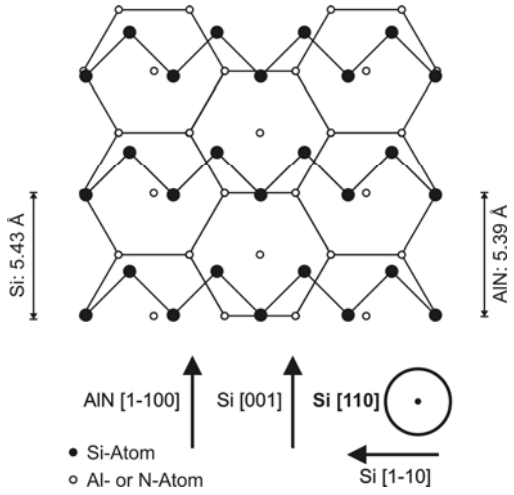

Figure 7. Atomic arrangement of AlN on Si(110).

samples as shown, e.g. for a quarter wafer in figure 6. Here the two different orientations are separated by a rough region in which both alignments are nearly equally present as shown by XRD ω-scans of the asymmetric $(10\bar{1}5)$ reflection for a whole rotation Φ of the wafer in the three different regions.

Applying low-temperature seed layers other GaN orientations were observed, mostly *r*-plane GaN [25]. But obtaining smooth layers is very difficult, e.g. *r*-plane GaN also shows four different orientations on this surface which can be reduced largely to one if off-oriented substrates are used. However, even with these methods the layer quality is still too poor for any device growth, in particular the surfaces are still rough and not continuous.

5. Si(110) and higher index planes

Si(110) is, besides Si(001) and Si(111), a surface orientation used in the silicon industry. Despite its good availability it has to our knowledge, not been investigated for GaN growth yet. An interesting point of this surface orientation is lattice mismatch which is very low in one direction for *c*-axis oriented AlN if it grows with $\mathrm{AlN}[1\bar{1}00] \parallel \mathrm{Si}[100]$ which is equal to $\mathrm{AlN}[11\bar{2}0] \parallel \mathrm{Si}[1\bar{1}0]$ (figure 7), as confirmed by the experiment. Here the lattice mismatch accounts to only 0.7% (when every second atomic plane is taken into account), as for one direction on Si(001), and 19% (matching every $\sim$10 Å), as for AlN on Si(111). In contrast to the before mentioned substrate orientations, this orientation likely enables a better rotational match. A direct comparison of the GaN XRD data for Si(111) and Si(110) with an identical growth structure confirms this assumption. Here, an approximately 600 nm thick GaN layer shows much better values for the XRD rocking curves in (0002) and especially for $(1\bar{1}00)$ reflections, namely 734 versus 1195 arcsec and 1030 versus 2830 arcsec for the layers on Si(110) and Si(111), respectively (figure 8). For GaN on Si(111) and the layer thickness grown these values are typical values we obtain for simple structures. We assume the comparably low FWHM for the $(1\bar{1}00)$ reflection is originating in the low mismatch in one direction namely $\mathrm{AlN}[1\bar{1}00] \parallel \mathrm{Si}[100]$. The better the match the higher the probability that the AlN seeding crystallites are growing in a well aligned manner.

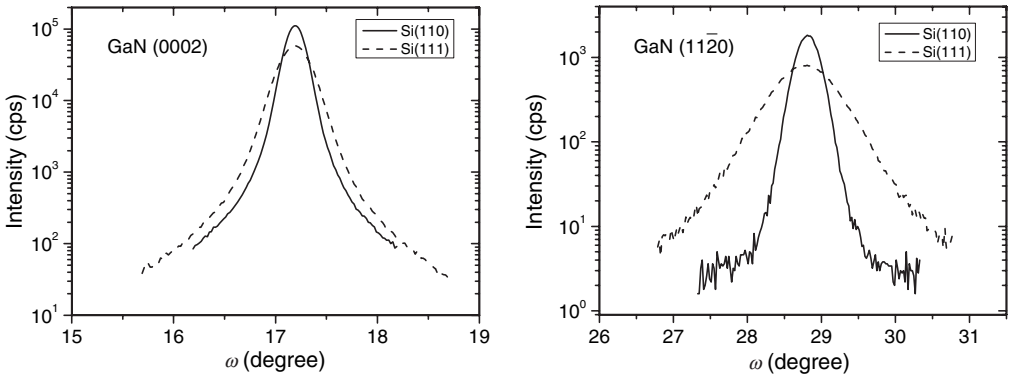

Figure 8. XRD ω-scans around the (0002) and (11$\bar{2}$0) reflections for a sample with 600 nm of GaN on Si(110) and Si(111).

In contrast to Si(001) and Si(111) with a rotational symmetry of four and three, respectively, the Si(110) surface has a symmetry of only two. While for the growth on Si(001) surface reconstruction is believed to play the important role for reducing the symmetry and in achieving only one GaN orientation, the reduced symmetry of Si(110) already exists without taking into account surface reconstruction. At room temperature the Si(110) surface usually exhibits a (16×2) reconstruction which transforms into a (1×1) at 790 °C [26] in vacuum. For HT AlN seed layers single crystalline growth is simply achieved, however, for low-temperature seed layers as successfully applied on Si(111) no successful growth was observed yet, which we assume to originate in an unfavorable surface reconstruction at low temperatures.

Besides Si(110) other higher index planes with a symmetry of two such as, e.g. (115), (117) and (119) yield single crystalline GaN, however, only comparable to Si(111), likely due to a higher lattice mismatch in both directions as compared to Si(110).

The better material quality on Si(110) also leads to a lower growth induced stress, e.g. from island coalescence which does add a tensile stress component to the tensile thermal stress always present for GaN on Si [27]: for a test sample even a weak compressive strain of −0.05% compared to 0.15% for Si(111) is found, which is most likely due to the Al-rich buffer inducing strong compressive stress during subsequent GaN growth. This largely fails for the growth on other surfaces.

Further benefits of growing on Si(110) are easier cleavage in orthogonal directions compared to Si(111) because there are primary {111} cleavage planes perpendicular to the (110) plane and easier etching, e.g. for thin film devices where the substrate has to be removed. This is very difficult to achieve for Si(111) substrates where an aggressive HF : HNO_3 etch is often applied which also etches the carrier the sample is bonded to and a possible metal bond between them which are difficult to protect against the acid. For Si(110), with even higher etch rates than for Si(100) for many etchants, many different etching solutions exist, giving more flexibility in finding a suitable etch for processing.

6. Summary

We have investigated the growth of GaN on different Si surfaces. The commonly used Si(111) surface has proven to be suited for device growth, however, with only an average crystalline

quality. Growth on Si(001) is most likely strongly influenced by surface reconstruction and requires off-oriented substrates to obtain single crystalline layers. Best results for GaN-on-Si can be obtained on Si(110) where a lower lattice mismatch exists in one direction, significantly improving twist values. First device data from such GaN layers on Si(110) are expected soon.

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